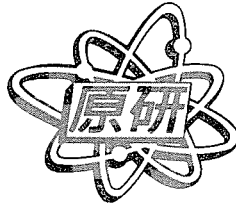


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NUCLEAR CRITICALITY SAFETY HANDBOOK, VERSION 2
(ENGLISH TRANSLATION)

August 2001

Working Group on Nuclear Criticality Safety Data

日本原子力研究所
Japan Atomic Energy Research Institute

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Nuclear Criticality Safety Handbook, Version 2 **(English Translation) ※**

Working Group on Nuclear Criticality Safety Data

Special Committee on Nuclear Criticality Safety
Nuclear Fuel Facility Safety Research Committee
Japan Atomic Energy Research Institute
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The *Nuclear Criticality Safety Handbook, Version 2* essentially includes the description of the *Supplement Report to the Nuclear Criticality Safety Handbook*, released in 1995, into the first version of the *Nuclear Criticality Safety Handbook*, published in 1988. The following two points are new: (1) exemplifying safety margins related to modeled dissolution and extraction processes, (2) describing evaluation methods and alarm system for criticality accidents. Revision has been made based on previous studies for the chapter that treats modeling the fuel system: e.g., the fuel grain size that the system can be regarded as homogeneous, non-uniformity effect of fuel solution, and burnup credit. This revision has solved the inconsistencies found in the first version between the evaluation of errors found in JACS code system and the criticality condition data that were calculated based on the evaluation. This report is an English translation of the *Nuclear Criticality Safety Handbook, Version 2*, originally published in Japanese as JAERI 1340 in 1999.

Keywords: Criticality Safety, Handbook, Burnup Credit, Criticality Accident,
Criticality Alarm System, Homogeneous, Error Evaluation,
Criticality Condition Data, Dissolution Process, Extraction Process

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※ Translated by Hiroshi OKUNO, Yasushi NOMURA (JAERI) and Susumu RYUFUKU (Visible Information Center, Inc.)

臨界安全ハンドブック第2版 (英訳) ※

日本原子力研究所核燃料施設安全性研究委員会臨界安全性専門部会
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「臨界安全ハンドブック」第1版(昭和63年刊行)に、「臨界安全ハンドブック改訂準備資料」(平成7年刊行)の内容を盛込んで、この第2版を作成した。第2版では、以下の2点を新規に追加した。(1)実際の化学プロセスが持つ安全裕度を溶解工程及び抽出工程に対するモデル計算の形で例示したこと。(2)臨界事故への対応として、臨界事故の評価方法及び臨界警報装置の設計・設置の考え方について記述したこと。また、臨界安全評価を行う際のモデル化について、均質と見なしてよい燃料粒径や、燃料濃度の不均一性の影響、燃焼度クレジットなど、これまでの研究成果を踏まえ、内容の充実を図った。さらに、第1版では、臨界条件データとその計算に用いられたJACSコードシステムの計算誤差評価結果との間に対応のとれていないものが一部含まれていたが、今回の改訂ではその整合を図った。この報告書は、1999年にJAERI1340として日本語で刊行した「臨界安全ハンドブック第2版」の英訳である。

この報告書は、電源開発促進対策特別法に基づく科学技術庁からの受託として行った研究成果である。

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1. INTRODUCTION

In 1988, the Nuclear Criticality Safety Handbook, Version 1, hereafter called the first version, was compiled by Nuclear Materials Regulation Division, Nuclear Safety Bureau, Science and Technology Agency and published from Nikkan Shobo. The first version played an important role in the criticality safety assessment of nuclear fuel facilities in Japan. Specifically, it provided the technical basis for both the applicant and the regulator in the safety assessment of the Rokkasho reprocessing plant whose construction was completed in 1992.

Being the first nuclear criticality safety handbook published in Japan, the first version did not include some issues to be added. Therefore, Japan Atomic Energy Research Institute (hereafter called JAERI) has started the preparation work for the second version since the publication of the first version in 1988. This preparation work was conducted by Working Group on Nuclear Criticality Safety Data (Leader: Kojiro Nishina, Professor of University of Nagoya (then)) under entrustment by the Science and Technology Agency. The results of this preparation work was released in 1995 as a JAERI technical report (Report number: JAERI-Tech95-048) titled "the Supplementary Report to the Nuclear Criticality Safety Handbook, Version 1" (hereafter called the supplement report).

The Nuclear Criticality Safety Handbook, Version 2 (hereafter called the second version) essentially incorporates the content of "the Supplementary Report into the first version". In addition, based on the experience of using the first version and the supplementary report, necessary revisions were made to some individual sections. The members of Working Group on Nuclear Criticality Safety Data (Leader: Yasushi Nomura, Head of fuel cycle safety evaluation laboratory), who participated in this preparation work, are listed in Table 1.1. Considering the nature of this document and the involvement of JAERI in the preparation work, it was decided that the second version would be published as a JAERI technical document.

The second version includes the following four features:

- (1) It described safety margins related to modeled dissolution and extraction processes (Chapter 5).
- (2) It described evaluation methods for criticality accidents (Chapter 6) and criticality alarm system design and installation principles (Chapter 7) (neither (1) nor (2) above were included in the first version).
- (3) Revision was made based on previous studies for the chapter that treats modeling the fuel system: e.g., the fuel grain size that the system can be regarded as homogeneous (3.1.11), non-uniformity effect of fuel solution, and burnup credit (3.5.2).
- (4) It solved the inconsistencies between the evaluation of errors found in JACS code system and criticality condition data that were calculated based on the evaluation.

"The Data Collection attached to Nuclear Criticality Safety Handbook " (hereafter called the data collection) was not revised for the second version. For new data added to the second version,

please refer to relevant reference documents quoted in the main text. However, the revision work for the data collection has been started since 1998, based on the criticality experiment data and other data at the JAERI Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF).

Table 1.1 Members of working group on nuclear criticality safety data (1995-1997)

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Introduction in the 1st Version of Handbook

In establishing nuclear fuel facilities, including nuclear fuel processing facilities and reprocessing facilities, it is important to pay attention to the conditions for location of the facilities, radiation control, environmental safety, criticality safety, and other safety measures. This Handbook describes the principles for addressing criticality control (criticality safety) and the methods for analytical evaluation of criticality safety.

The Science and Technology Agency has compiled and published this Handbook, which contains the results of studies and evaluations made by persons concerned with nuclear related education, research, and industry in Japan. These results are based on calculations and analyses using existing and newly developed codes from the Japan Atomic Energy Research Institute (JAERI). This Handbook is published with the hope that it will serve as a reference for criticality safety. This Handbook provides methodologies and examples of evaluations. The interpretation and application of this Handbook are not guaranteed and are strictly the responsibility of the reader. This Handbook is compiled specifically for the evaluation of criticality safety, but it also will serve as references for the design and for safe control of nuclear fuel facilities.

The methodology for securing criticality safety of nuclear fuel facilities, the methods of evaluation, and the required data are already arranged into handbooks, standards and guides including TID-7016 Rev. 1 and Rev. 2, AHSB(S), and CEA-R3114 in the U.S.A., the U.K., and France, respectively. These handbooks are based on experience with nuclear facilities, experimental data from criticality safety research facilities, and theoretical studies performed since the 1940s. Germany has compiled its own handbook based on information from the U.S., the U.K., and France.

While compiling and editing this Nuclear Criticality Safety Handbook, literature from various sources, including American and European handbooks, was studied. Some differences in criticality control methods and data were noted among the sources of literature during this study. Consequently, for values concerning various types of criticality parameters described in this Handbook, the values calculated or evaluated in this present work were principally selected. However, this does not restrict the use of procedures and data clearly described in other sources when evaluating criticality safety. Thus, this Handbook may be used for safety evaluation. However, it does not mean that the values in it must be used. When values from other sources of literature are to be used, the consistency of these values and restrictions on their use need to be sufficiently considered.

Neither chemical processes nor criticality accident phenomena are described in this Handbook. Examination of chemical processes is important for criticality safety control in reprocessing facilities. However, such an examination is not described here because it should specifically address each process individually. The method of safety assessment in criticality accidents is also an important theme, but this is not described in this Handbook. Thus, this Handbook primarily describes methods to be employed to secure subcriticality and methods to verify that the system is in the subcritical state.

Neutron multiplication factors are used in this Handbook to determine subcriticality. These

multiplication factors were obtained by considering the calculational errors obtained from the analyses of many benchmark tests. The shape and dimensions, volume, and mass of the nuclear fuel materials determined as subcritical were calculated using these neutron multiplication factors. Consequently, the familiar method that multiplies the criticality quantity by a safety factor to determine subcriticality is not used. The rapid development of computers has allowed subcriticality usually to be determined with the computed neutron multiplication factor of the system.

It was thought desirable that the judgment standards for dimensions, volume, and mass be the same as the judgment standards used in determining subcriticality from the calculation results obtained from computers. It was decided to consider safety margins other than calculation error corrections when setting up an object evaluation model. The methods for evaluation of criticality safety described in this Handbook are written simply and references are shown for the basis of such methods. In addition, "Nuclear Criticality Safety Handbook - Data Collection" is an attachment to this Handbook. Detailed analytical evaluation safety data are included in it.

The contents of the following chapters of this Handbook are: Chapter 2 describes the basic methodology for criticality safety control, the terminology used, the methods to secure subcriticality, and the standards used to determine subcriticality; Chapter 3 provides examples of models that are appropriate for safety analyses of complex systems; and Chapter 4 deals with specific methods of safety assessment and evaluation. These methods are classified into single unit analyses and multiple unit analyses.

The attached "Nuclear Criticality Safety Handbook - Data Collection" consists of the data needed for calculation of atomic number density, parameters of nuclear characteristics for various nuclear fuel compositions, criticality data for infinite systems, criticality data for single units, and data needed for analysis of multiple units.

Since more realistic safety margins may be adopted with the advancement of nuclear research and technology, it is desirable that this Handbook be revised consistent with the increase in knowledge about and utilization of atomic energy. However, since the methods and values described in this Handbook have sufficient margins included, it is believed that no phenomena that are undesirable for criticality safety will result from the long-term use of the methods and values described herein.

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2. TECHNICAL METHODS FOR CRITICALITY SAFETY CONTROL

2.1. Basic Methodology and Terminology

The basic principle of criticality safety control is to prevent criticality for all situations that are technically conceivable. To achieve criticality safety, sufficient margins are considered in each stage of design, fabrication, construction, and operation of applicable facilities. In the design stage, criticality safety is demonstrated to be achievable. In the fabrication and construction stages, criticality safety is assured by verifying that the design conditions are satisfied. In the operation stage, all operations are controlled so criticality safety can be maintained according to the design.

Nuclear safety for conditions of specified shape and dimensions of a single unit, in normal and abnormal states, is verified by scrutinizing the kinds, quantities, and physical and chemical forms of the nuclear fuel and other materials contained in the unit. To verify the criticality safety of multiple units, the neutron interaction effects in both the normal and abnormal states are evaluated taking the distance between units, the thickness of shielding material, or both, into consideration.

When verifying subcriticality by calculation, the evaluation must provide sufficient safety margins by considering the reliability of the data and the calculational method. The reliability of the data and calculational method used should be fully verified by comparison with experimental data from a system physically similar to the object system. When experimental data from a system the same as the object system can be obtained, the criticality safety may be evaluated based on this data.

The important terminology used in this Handbook is:

(1) Single failure

Single failure shall mean the condition where one system or piece of equipment loses its specified functions because of a single phenomenon. **Single failure** also shall include **multiple failures** that could occur because of a single phenomenon.

(2) Single failure, etc.

Single failure, etc. shall mean a **single failure or malfunction of a system or piece of equipment, or single mistake in operation** where the frequency of its occurrence is estimated to be one or more times during the service life of the facility concerned.

(3) Changes in peripheral conditions caused by external factors

Changes in peripheral conditions caused by external factors shall mean changes in peripheral conditions including reflection, absorption, moderation, and mutual interaction of neutrons attributable to **external factors** such as earthquakes, floods, or fires. All external factors whose occurrence cannot be neglected shall be included.

(4) All cases conceivable from a technical standpoint

All cases conceivable from a technical standpoint shall refer to all conceivable **single failure, etc. or changes in peripheral conditions caused by external factors**. The severest conditions for criticality safety for the assumed occurrence shall apply.

(5) **Criticality factor**

Criticality factors shall include all factors whose change influences the criticality of the system. Such factors include the shape and dimensions of the equipment containing nuclear fuel material, the concentration of the nuclear fuel material in solution, the mass and isotopic composition of nuclear fuel material, and the physical and chemical properties of the nuclear fuel material. Included also are the shape and dimensions, concentration, and material quality of neutron absorbers and the neutron reflection conditions.

(6) **Criticality control factor**

The **criticality control factor** shall apply to the **criticality factor** used to set **criticality limits** for design or operational control to secure criticality safety of nuclear fuel facilities.

(7) **Criticality limits**

A **criticality limit** shall be set for each **criticality control factor** to secure subcriticality of the system. These values shall be set to simplify the judgment of, and ensure the appropriateness of, the specific design and operating conditions of the equipment. The values shall be directly measurable values, such as dimensions, concentration, etc., or indirectly controllable values. Equipment shall not be fabricated that exceeds these values, and normal operating conditions shall not be established with values exceeding these values.

Examples of criticality limits include limits on mass, dimension (diameter, thickness), volume, concentration, array configuration, solid angle, etc.

(8) **Estimated lower limit criticality value and Estimated criticality lower limit multiplication factor**

The **estimated lower limit criticality value** shall be the value where a system is judged as subcritical if each of the criticality control factors of the system is less than this value. This value shall be determined considering the estimated criticality value and its calculational error where the system is believed to be critical. When a criticality value is to be calculated, the calculation accuracy of the calculation code or codes used must be evaluated in advance. This accuracy is to be determined according to the benchmark calculation using the criticality test data of systems similar to the object system. The upper limit of the neutron multiplication factor where the system may be judged as subcritical is obtained by this accuracy evaluation and is called the **estimated criticality lower limit multiplication factor**. The **criticality control factor** value corresponding to the **estimated criticality lower limit multiplication factor** is the **estimated lower limit criticality value**.

(9) **Single unit**

Single unit shall be defined as a unit for handling nuclear fuel material for which a set of **criticality limits** can be determined and criticality control can be performed with the limits. **Single units** for which **criticality limits** can be determined shall be those units for which peripheral conditions can be imposed to produce the severest conditions for criticality control and for which the kinds, compositions, physical and chemical properties, and quantities of the contained nuclear fuel and other materials can be specified.

A glove box or cell that is mass controlled is an example of a **single unit**. Even if various containers are in the glove box, the quantity and properties of the nuclear fuel inside can be specified and subcriticality can be maintained by controlling the total quantity of the nuclear fuel material present in the glove box to be within the specified mass limit value.

(10) **Multiple unit**

A system consisting of two or more **single units**, reflectors, and shielding materials shall be defined as a **multiple unit**. A **multiple unit** must be a system in which each of the **single units** contained therein and the arrangement of each of the single units can be specified. To secure criticality safety, the configuration of the **single units** and of the neutron shielding materials must be determined so no criticality can occur in any situation conceivable from a technical standpoint by considering the mutual interaction of leakage neutrons between single units.

(11) **Maximum permissible multiplication factor**

The maximum permissible multiplication factor is used for judging if a system is subcritical by calculating its multiplication factor. This value shall be set below the **estimated criticality lower limit multiplication factor**.

(12) **Maximum permissible limit**

In the criticality safety evaluation, a criticality control factor corresponding to the **maximum permissible multiplication factor** is called the **maximum permissible limit**. Examples of the **maximum permissible limits** corresponding to concentration, mass, and solid angle are "the maximum permissible concentration," "the maximum permissible mass," and "the maximum permissible solid angle." These values are also called "the subcritical concentration," "the subcritical mass," and "the subcritical solid angle," respectively.

(13) **Reactivity, Subcriticality, Reactivity worth**

Reactivity is an indicator of how far a system is away from criticality. The **reactivity** ρ is given by the following equation where k is the neutron multiplication factor of the object system.

$$\rho = \frac{(k-1)}{k} \quad (2.1)$$

When $k < 1$, $-\rho$ is called the **subcriticality**.

When the state of a system changes, that is, when a neutron absorber is added or the amount of a moderator decreases, the neutron multiplication factor changes accordingly. The difference in **reactivity** before and after the state change is called **reactivity worth** of the material for the state change. The **reactivity worth** associated with the state change is given by the equation below where k_1 and k_2 are the neutron multiplication factors of the system before and after the state change, respectively.

$$\rho_2 - \rho_1 = \frac{(k_2 - 1)}{k_2} - \frac{(k_1 - 1)}{k_1} = \frac{1}{k_1} - \frac{1}{k_2} \quad (2.2)$$

When obtaining the **reactivity** by a criticality calculation, we first calculate the neutron multiplication factor under the critical condition. Assume that the calculated value is k_1 , which is not necessarily 1.0. Next, we calculate the neutron multiplication factor for the system after the state change. Assume this value is k_2 . If we use a constant multiplier to force the neutron multiplication factor for the critical state to equal necessarily 1.0, we could estimate the neutron multiplication factor after the state change to be k_2/k_1 . Using this relationship, the **reactivity** is given by the equation below.

$$\rho' = \frac{\left(\frac{k_2}{k_1} - 1 \right)}{\frac{k_2}{k_1}} = \frac{(k_2 - k_1)}{k_2} \quad (2.3)$$

Even if the criticality condition is not known, we may calculate the difference or the change in neutron multiplication factor before and after the state change. In this case, the equation above is often used to represent the change ratio of the neutron multiplication factor, rather than the **reactivity**. In this case, k_1 is the reference neutron multiplication factor.

(14) **Convex geometry and concave geometry**

A single fuel unit with a geometry curving outward is said to have a **convex geometry**. Here, the geometry curving outward means that when two arbitrarily selected two points within the geometry are connected, all the points on the line are situated within the geometry.

A single fuel unit not having a convex geometry is said to have a **concave geometry**.

For a single convex fuel unit without a reflector, neutrons emitted from the unit do not go back to the unit and thus the **reactivity worth** of the reflector is always positive. In contrast, for a single concave fuel unit without a reflector, neutrons emitted from the unit may go back to the unit. A reflector situated in the concave portion may function as an absorber, and thus the reactivity worth of the reflector is not always positive. In addition, since the reflector in the concave portion may function as a moderator, care must be given when calculating the optimum moderating condition.

(15) **Homogeneous, heterogeneous, uniform and nonuniform**

A **homogeneous** system is a system in which the physical and chemical properties are constant or are changing continuously over the entire system, such as a solution system. A **heterogeneous** system is a system in which the physical and chemical properties change discontinuously, such as a system with fuel rods in water. The terms **uniform** and **nonuniform** are terms to distinguish whether or not the physical and chemical properties of a system are uniform. For example, a system in which the solution concentration is uniform everywhere is a **homogeneous uniform** system and a system in which the solution concentration distribution is not uniform is a **homogeneous nonuniform** system. A system in which fuel rods are uniformly arrayed in water is a **heterogeneous uniform** system. A system in which fuel rods are not arrayed uniformly is a **heterogeneous nonuniform** system.

Reflectors and vessels are not taken into account when judging a system homogeneous or heterogeneous.

(16) **Branch pipes (arms)**

Small pipes that intersect with a main nuclear fuel vessel are called **branch pipes (arms)**. Normally the **branch pipes** contain the same fuel as the main vessel, but often the **branch pipes** are ignored in calculating the criticality. This is because the **reactivity** of the **branch pipes** is relatively small and because the branch pipes complicates the calculation and may introduce many errors. When ignoring **branch pipes** in a criticality calculation, however, one must consider the magnitude of contribution of the **branch pipes** in neutron multiplication factor for criticality safety evaluation.

(17) **Burnup credit**

Burnup credit means accounting for a decrease in neutron multiplication factor associated with fuel depletion in criticality safety design and criticality safety control of a spent fuel handling facility.

The decrease in neutron multiplication factor associated with fuel depletion is attributable to a decrease of the fission rate caused by decreased masses of fissile materials and to an increase of neutron absorption caused by accumulation of fission products.

2.2. Classification of Installations for Criticality Safety Design

In criticality safety design, nuclear fuel facilities are categorized into facilities subject to criticality safety control and facilities not subject to criticality safety control according to the need of criticality safety control. The facilities in the former category are further divided into facilities for which one needs to consider measures for preventing criticality accidents and facilities for which one does not need to consider measures for preventing criticality accidents, depending on the need of considering measures for preventing criticality accidents.

2.2.1. Installations Requiring Criticality Safety Control

These are facilities required to adopt criticality safety control measures for preventing criticality accidents.

Here, the criticality safety control means taking measures for preventing criticality for all situations that are technically conceivable by controlling such criticality factors as shape, dimension, concentration, mass, isotope composition, neutron poison, and the combination of these. In other words, the criticality control means taking necessary measures so that criticality accidents can be prevented against all conceivable single failure, etc. and / or changes in peripheral conditions caused by external factors.

By taking such criticality safety control measures, probability of criticality accidents is considered very small. Nevertheless, for some facilities, one still needs to consider countermeasures against occurrence of criticality accidents, whereas for other facilities in which the counter criticality safety control is relatively easy and sufficient safety margins are provided, one can neglect occurrence of criticality accident.

(1) Facilities for which one needs to consider countermeasures against occurrence of criticality accidents

Facilities in this category are those facilities selected to guarantee residual uncertainties in the criticality safety control, though the probability of criticality accidents is very low. Specific countermeasures for a criticality accident are divided into two groups. One group of measures give workers an alarming signal during a criticality accident to immediately evacuate, and the other group of measures makes the accident subcritical.

Necessary alarm devices are installed to minimize the radiation exposure of workers. Whether one needs to take countermeasures for making accidents subcritical is determined by taking account of the severity of criticality accidents at a subject facility (e.g., important facilities from the points of criticality safety control such as fuel solution handling facilities) and exposure to the public.

(2) Facilities for which one does not need to consider counter measures against occurrence of criticality accidents

Examples of the facilities in which the criticality safety control is simple, sufficient safety margin is secured, and thus the occurrence of criticality accidents is neglected, are follows:

- Facilities adopting geometry control in the full concentration range
- Facilities adopting concentration control, where there are no factors increasing the concentration
- Facilities where the amount of fuel is small and dispersed
- Facilities where a sufficiently robust structure exists for maintaining enough distance between fuel assemblies

2.2.2. Installations Not Requiring Criticality Safety Control

Facilities in this category are facilities that contain only these fuel materials rendering no chance of going critical without criticality control measures, and facilities that never contain these fuel materials.

An example of fuel material rendering no chance of going critical is one that does not contain physical and chemical mechanism to reach more than 1.0 of infinite multiplication factor. Specific examples include uranium fuel with less than 0.88 % enrichment, uranium fuel with less than 11 g²³⁵U/L concentration (Refer to Fig. 4.2.), and plutonium fuel with less than of 7 g²³⁹Pu/L concentration (Refer to Table 4.9), in which precipitation and/or sedimentation do not have a chance of creating an local concentration increase leading to criticality. Vitrified high-level liquid waste contains plutonium and other fissile materials. However it is almost unlikely in canister storage, that these fissile materials are extracted from multiple canisters and concentrated in one region. For this reason, some vitrification canister storages are considered as facilities not subject to criticality safety control. In contrast, care must be taken for those facilities or devices that are not designed to contain fuel materials. These facilities are considered as facilities subject to criticality safety control, if there is a chance of fuel materials flowing into the facilities or devices.

2.3. Methods of Securing Subcriticality

Nuclear fuel facilities must be controlled so nuclear fuel will not become critical in any situation technically conceivable. This section briefly describes methods used to secure subcriticality. It also clarifies the nature of these methods and provides an outline and background of the physical mechanisms involved. Furthermore, the principles and procedures to attain subcriticality are described. To determine the specific value of each of the criticality factors for design and control when performing the methods described in this section, the procedures need to be followed based on the state of each facility concerning the method for determining subcriticality described in Section 2.4 and the method for analytical safety evaluation described in Chapter 4.

2.3.1. Securing Subcriticality of Single Units

Measures must be taken to prevent the nuclear fuel in **single units** from becoming critical in any case technically conceivable by controlling each of the criticality factors. These factors include the shape and dimensions of the unit, the concentration, mass, and isotopic composition of the nuclear fuel material contained in the unit, and the shape and dimensions, concentration, and material qualities of neutron absorbers. The absorption, moderation, and reflection conditions for neutrons around the **single units** also must be considered.

(1) Equation of the modified one-group theory

This section provides a general description of how these criticality factors and peripheral conditions will influence the neutron multiplication factor and criticality conditions. Equation (2.4) is used to illustrate these factors and conditions. This equation is a representative formula of the effective multiplication factor, k_{eff} , by the modified one-group theory.

$$k_{\text{eff}} = \frac{k_{\infty}}{1 + M^2 B^2} \quad (2.4)$$

where

k_{∞} : Infinite multiplication factor of the fuel materials comprising a single unit,

B^2 : Buckling dependent upon the shape and dimensions of a single unit, and

M^2 : Migration area of a neutron in the fuel material ($M^2 = \tau + L^2$ where τ : Age for a fission neutron slowing down to a thermal neutron in the fuel material and L : Diffusion length of the thermal neutron in the fuel material.)

This equation is thought to provide a high accuracy result for a bare nuclear fuel system. For this condition, buckling, B^2 , is given by the equation shown in Table 2.1. For a nuclear fuel system with a reflector, buckling is given by the equation shown in Table 2.2.

Despite the fact Eq. (2.4) is a very simple representational formula, the effective multiplication factor k_{eff} of a single unit can be calculated with considerable accuracy if k_{∞} , M^2 , and B^2 of the equation are determined by detailed calculations using recent calculational codes. In this Handbook, therefore, this theory is used not only for the following explanation but also for a simplified calculational method with the M^2 (migration area) and d_r (the extrapolation length of a system with a reflector) values obtained through more detailed calculations. The methods available to this theory are described in Chapter 4. The methods used there are based on Eq. (2.4). When a person is confronted by a special problem that is not covered by these data and calculation diagrams, or if these methods are believed to be unable to provide the required accuracy, calculations with detailed methods should be made referencing the value obtained from Eq. (2.4). Higher accuracy, detailed, calculational methods are described in Chapter 4. Furthermore, even if a calculation with higher accuracy is to be made by a detailed calculational method, this equation is suitable for systematically arranging a large quantity of data derived from the detailed calculation and for understanding the

fundamental physical mechanisms.

To obtain subcriticality conditions, it is necessary first to obtain the **estimated criticality lower limit multiplication factor**, k_L , through examination of calculational results of the benchmark systems similar to the object system. The ranges of various **criticality factors** when $k_{\text{eff}} < k_L$ are then obtained using Eq. (2.4). Allowable fluctuation ranges of various **criticality factors** are examined with an example of a sphere using the following equation.

$$k_{\text{eff}} < k_L \quad (2.5)$$

(2) Discussion of each of the **criticality factors**

Each of the **criticality factors** is discussed using UO_2 powder containing water as an example. A criticality buckling chart for homogeneous $\text{UO}_2\text{-H}_2\text{O}$ is shown in Fig. 2.1. The relation between the uranium concentration and H/U is shown in Fig. 2.1(a). Fig. 2.1(b) shows the relation between k_{∞} and H/U . Fig. 2.1(c) shows the relation between M^2 and H/U using uranium concentration as a parameter. When uranium concentration and enrichment are given, k_{∞} and M^2 are determined and the buckling corresponding to k_L can be obtained from Fig. 2.1(d). The shape and dimensions corresponding to k_L can be obtained using this buckling value and the relation between buckling and dimensions shown in Table 2.1 or Table 2.2.

(a) **Shape and dimension control**

Substituting Eq. (2.4) into Eq. (2.5) to eliminate k_{eff} , the following inequality is obtained for a spherical geometry system.

$$B^2 = \left(\frac{\pi}{R+d} \right)^2 > \frac{1}{M^2} \left(\frac{k_{\infty}}{k_L} - 1 \right) \quad (2.6)$$

Under the range of moderation conditions (for example, uranium concentration) and reflection conditions (those that influence the extrapolation distance, d , etc., which are selected that are the severest, yet are technically conceivable), the maximum value of dimension R that satisfies Eq. (2.6) is the **estimated lower limit criticality value**, R_{CL} . The method of securing subcriticality by performing control so no dimension of the object nuclear material becomes larger than R_{CL} is called **shape and dimension control**.

As a special case, if k_{∞} of the nuclear fuel material of the single unit being handled is smaller than k_L , the right side of Eq. (2.6) becomes negative. Thus, this inequality is satisfied, even if radius R is infinitely large. Concerning (b) in Fig. 2.1, if the enrichment is less than about 1 wt%, k_{∞} will not exceed 1.0 for any value of H/U . Examples are shown in Chapter 4 as the subcriticality conditions concerning infinite systems.

(b) Volume control and mass control

For the same conditions of nuclear fuel material, a sphere has the smallest volume where a nuclear fuel material can become critical. The volume corresponding to the **estimated lower limit criticality value**, R_{CL} , obtained by the shape and dimension control for a sphere is the **estimated lower limit criticality value**, V_{CL} , in volume control for the nuclear fuel material. **Volume control** is the method of securing subcriticality by controlling the volume of nuclear fuel material so it does not become larger than V_{CL} .

The critical volume usually becomes large if the concentration of nuclear fuel decreases. However, it must be stressed that the critical mass of nuclear fuel sometimes may decrease as the fuel concentration decreases. The smallest mass corresponding to k_L is the **estimated lower limit criticality value**, M_{CL} , in mass control. **Mass control** is the method of securing subcriticality by controlling the mass of nuclear fuel material so it does not become larger than M_{CL} .

(c) Concentration control and moderation control

When fuel concentration decreases below a certain value, the neutron multiplication factor will decrease due to an increase in the neutron absorption ratio in the solvent. As is evident from Fig. 2.1, when H/U is more than 20, k_{∞} decreases with increases in H/U . When the concentration of nuclear fuel in a container decreases below a certain value, ρ_{CL} , the neutron multiplication factor becomes less than k_L . The term ρ_{CL} is the **estimated lower limit criticality value** in concentration control in a finite system. **Concentration control** is the method of securing subcriticality by controlling the concentration of nuclear fuel material so it does not become higher than ρ_{CL} .

When the concentration drops to less than ρ_{CL} and the value of k_{∞} becomes less than k_L , this concentration is called the **estimated lower limit criticality value** of the concentration in an infinite system or merely the **estimated lower limit criticality concentration**. The **estimated lower limit criticality concentrations** for various nuclear fuel materials are shown in Chapter 4.

In contrast to concentration control, criticality safety may be secured by holding H/U at a low level. When H/U is less than 5, k_{∞} decreases with decreases in H/U , as shown in Fig. 2.1. Let us suppose that the neutron multiplication factor has become k_L when H/U in a specified container has become $(H/U)_{CL}$. This $(H/U)_{CL}$ is called the **estimated lower limit criticality value** for moderation control. The method of securing subcriticality by performing control so the H/U of the low-enriched uranium system does not exceed $(H/U)_{CL}$ is called **moderation control** for a low-enriched uranium system. Assume $k_L=0.95$, for the homogeneous UO_2 - H_2O in Fig. 2.1, where the enrichment of ^{235}U is less than 5 wt%, k_{∞} becomes less than k_L in the region of $H/U \leq 0.2$. It should be stressed that for many cases of highly enriched uranium and plutonium, the neutron multiplication factor increases with a decrease in water content.

(d) Isotopic composition control and criticality control using a neutron absorber

The isotopic composition of a nuclear fuel material and the quantity of neutron absorber in the fuel have a strong influence upon the value of k_{∞} . For example, criticality need not be controlled in

most cases for uranium compounds where the enrichment of ^{235}U is less than 1%. Plutonium containing a large amount of ^{240}Pu has a large critical mass. Thus, subcriticality is secured by controlling the isotopic composition of the nuclear fuel material. Cadmium, gadolinium, boron, hafnium, etc., are used as neutron absorbers. Mixing these neutron absorbers into nuclear fuel material will decrease the value of k_{∞} . As a result, the quantity of nuclear fuel material handled in a single unit can be increased. Thus, subcriticality is secured by maintaining a minimum quantity of neutron absorber mixed in the fuel.

The above offers a general view of the methods used to secure subcriticality of a single unit. The cautions for the respective methods of criticality control are described in Section 2.5. The various **estimated lower limit criticality values** needed for criticality safety control are described in Chapter 4.

2.3.2. Securing Subcriticality of Multiple Units

As for **single units**, criticality of **multiple units** also must be prevented for any case that is technically conceivable. Specific methods are:

- (1) To maintain the distance between **single units** above a certain value.
- (2) To locate neutron shielding material between the **single units**.

To determine subcriticality for a **multiple unit**, one or more of the methods described in Chapter 4 can be adopted. For subcriticality of a **multiple unit**, the subcriticality of each of the **single units** that are component elements of the **multiple unit** must be identified as described previously. The effective multiplication factor, k_i , of each **single unit** obtained previously, (where i represents the number of a **single unit**), is closely related to the effective multiplication factor, k_A , of the **multiple units** that are discussed in this section. The relation between k_i and k_A is conceptually described below to clarify the characteristics of **multiple units**.

Even when the subcriticality state is secured for each of the **single units** in a **multiple unit** according to the methods and principles described in Section 2.3.1, the **multiple unit** where two or more single units are gathered may achieve criticality. The reason is that when only one **single unit** is present, a neutron that has leaked from the unit cannot serve as a multiplication neutron. However, in **multiple units**, such a neutron may enter another **single unit**, where it can contribute to the multiplication of neutrons.

The phenomenon in which the effective multiplication factor, k_A , of a **multiple unit** becomes larger than the effective multiplication factor of a **single unit** through the mechanism described above is called the mutual interaction by neutrons. As confirmed in Section 2.3.1, $k_i < k_L$ will hold for any single unit i . Subcriticality of a **multiple unit** is secured by inhibiting this interaction within a certain limit. For the interaction between i and j , each a **single unit**, the probability of a neutron arriving at i from j is approximately proportional to the fractional solid angle $\omega_{ji} = \Omega_{ji}/4\pi$, where Ω_{ji} is the solid angle that unit i forms about a representative point of unit j . To limit the interaction, ω_{ji} can be reduced to a certain value by increasing the distance between i and j . Alternatively, the arriving

probability can be reduced by placing neutron shielding material in the path between the **single units**.

The following model provides insight into this mechanism. Suppose that the distance between **single units** is sufficiently large so the neutron flux distribution in each unit is unaffected by the presence of another **single unit**. Interaction between **single units** is so small that the neutron flux distribution in each unit is almost the same as for isolated **single units**. In such a case, the effective multiplication factor, k_i , and buckling, B_i^2 , can be defined for each of the **single units**. When the space between **single units** is assumed to be air, it is possible to prove that the following inequality can hold for the effective multiplication factor, k_A , of multiple units.

$$k_A < \text{Max}_{i=1,n} \frac{k'_i}{1 - \omega_i} \quad (2.7)$$

In this formula, ω_i is the result obtained by dividing the total of the solid angles that all **single units** around single unit i form with **single unit** i by 4π . The term k'_i is the effective multiplication factor of the **single unit** where air is assumed to surround the unit. This term is different from the effective multiplication factor, k_i , of a **single unit**, where water reflection is assumed. As is evident from this formula, to secure subcriticality of **multiple units**, the effective multiplication factor, k'_i , of each **single unit** and the solid-angle fraction, ω_i , must be controlled to small values.

2.4. Methods for Determining Subcriticality Conditions

Fig. 2.2 shows various levels of neutron multiplication factors used in criticality safety design. The basic principle of criticality safety control described in this Handbook is incorporating sufficient safety margins in setting evaluation conditions and modeling an evaluation object so that the calculation gives a conservative result. With respect to setting evaluation conditions, in particular, this Handbook requires setting the severest (most unfavorable) conditions that are technically conceivable to assure that required safety margins against many uncertainty elements be incorporated in this stage. The estimated maximum multiplication factor k_p is calculated under the severest (most unfavorable) conditions in terms of nuclear criticality. When calculating this multiplication factor, uncertainties associated with numerical calculations must be considered. When using a Monte Carlo method, 3σ (three times of standard deviation) shall be added to the calculated average neutron multiplication factor.

The maximum permissible multiplication factor k_a shall be set at a lower level than the estimated lower limit multiplication factor k_L . The difference between k_L and k_a , or Δk_s , is additional safety margins and can be set arbitrarily.

2.4.1. Methods for Determining Estimated Lower Limit Criticality Values

The **estimated lower limit criticality value** refers to the value determined as subcritical if the **criticality control factor** is less than that value. The estimated criticality value is that **criticality control factor** at which the system becomes critical under the severest conditions for criticality considering the fluctuations in the normal and abnormal states for the object system. There are two ways to find the **estimated criticality value**, a calculational method and a method using experimental data from a similar system. When a calculational method is employed, a sufficient safety margin including its accuracy must be considered. When experimental data are used, a sufficient safety margin found by evaluating the difference between the test system and the object system must be included. The value thus established is the **estimated criticality value**. Two methods are used to obtain the **estimated lower limit criticality value**. One method is to obtain the calculation accuracy from the results of many benchmark calculations using experimental data of the test system physically similar to the object system. Then, the multiplication factor (**estimated criticality lower limit multiplication factor**) at which the system may be judged as subcritical is obtained. The other method is to obtain the **estimated lower limit criticality value** by multiplying the estimated criticality value by a proper safety factor. The term **safety factor** here considers the safety margin only in the estimation error that may arise when the criticality value is estimated.

The relation between **estimated criticality lower limit multiplication factor** and safety factor is shown in Fig. 2.3. The safety factor shown in this diagram refers to the ratio of the values of those variables that correspond with the estimated criticality multiplication factor, to the values of the diameter of a circular cylinder, the thickness of a slab, or the volume or mass, where the multiplication factor is 1.0. This figure consists of an upper and lower diagram. The term C_k in the abscissa represents the shape and dimension reactivity factor, $\partial \ln k_i / \partial R$, and R_{crit} is the critical radius (or critical thickness. If the modified one-group theory equation is used, C_k and R_{crit} can be represented by the following equations for a sphere and a slab (for a circular cylinder, π is replaced by 2.405).

$$C_k = \frac{2}{\pi M} \cdot \frac{1}{k_\infty} (k_\infty - 1)^{\frac{3}{2}}$$

$$\left(\frac{\pi}{R_C + d_r} \right)^2 = \frac{k_\infty - 1}{M^2}$$
(2.8)

Consequently, if M^2 and d are considered as fixed, $1/C_k R_c$ becomes a function of k_∞ . This relation is shown in the lower diagram. This diagram assumes that $M^2 = 40 \text{ cm}^2$ and $d = 7 \text{ cm}$. Next, changes in the critical radius and changes in the neutron multiplication factor are expressed in the equation below.

$$\frac{\Delta R}{R_C} = \frac{1}{C_k \cdot R_C} \cdot \frac{\Delta k}{k}$$
(2.9)

Using this equation and the lower diagram, the safety factor with respect to the estimated criticality lower limit multiplication factor can be obtained as shown in the upper diagram Fig. 2.3.

Fig. 2.3 is best explained with an example. For the system where the infinite multiplication factor, k_{∞} , is 1.2, the safety factor corresponding to the **estimated criticality lower limit multiplication factor** of 0.95 for a circular cylinder, slab, or sphere is obtained. Transposing from the curve values at the ordinate value of 1.2 in the lower diagram to the **estimated criticality lower limit multiplication factor** of 0.95 in the upper diagram, the values of the safety factors are obtained. The estimated criticality lower limit multiplication factor of 0.95 corresponds to a safety factor of 0.85 for the diameter of a circular cylinder, 0.8 for the thickness of a slab, and 0.58 for a sphere.

As shown in Fig. 2.3, the relation between the **estimated criticality lower limit multiplication factor** and the safety factor varies greatly with the form of the system and the value of k_{∞} . Critical values in this Handbook are estimated using calculational codes verified by analyses of the results of many benchmark experiments. Therefore, the method of multiplying a quantity by a safety factor has not been used to find the **estimated lower limit criticality value**. Instead, the use of the **estimated criticality lower limit multiplication factor** has been adopted.

2.4.2. Method for Determining the Estimated Criticality Lower Limit Multiplication Factor

The **estimated criticality lower limit multiplication factor** has been obtained from the results of benchmark calculations using experimental criticality data from a system similar to the object system.

Table 2.3 shows the **estimated criticality lower limit multiplication factor** of a simple-form system with reflectors. This is a simple system that does not have neutron absorbers but has reflectors of sufficient thickness on its outer surfaces. Table 2.3 is based on the results of about 400 benchmark calculations for simple forms with reflectors (refer to ref(1) and data collection appendix C.) These calculations used the criticality safety assessment code system JACS that the Japan Atomic Energy Research Institute (JAERI) developed. The **estimated lower limit multiplication factors** of all groups are determined taking proper care so none of the benchmark calculational results will become less than the set values. The **estimated lower limit criticality value** can be obtained as the value corresponding to the **estimated criticality lower limit multiplication factor** for each group.

The **estimated criticality lower limit multiplication factors** of general-form systems with reflectors (all systems, including simple-form systems with reflectors) are shown in Table 2.4. This was prepared with the same method as was used for Table 2.3 and is based on the results of about 600 benchmark calculations for general-form systems with reflectors⁽²⁾.

When analytical criticality safety code systems other than JACS are used, **estimated criticality lower limit multiplication factors** can be obtained similarly from the results of benchmark

when using other well-verified code systems, the criticality safety of a **single unit** can be judged by verifying that the calculated effective multiplication factor satisfies the following expression.

$$k_{\text{eff}} \leq 0.95 \quad (2.10)$$

The method used to obtain the estimated criticality multiplication factor for confirming the criticality safety of a **multiple unit** shall be the same as that for a **single unit**. When the criticality safety of a **multiple unit** is to be confirmed using a simple calculation code such as MUTUAL, it is necessary to confirm in advance that the calculation results obtained using the simple calculation code have sufficient safety margins by comparison of the calculation results with the Monte Carlo code or with data from criticality experiments.

When a **criticality limit** is to be determined using the multiplication factor of an infinite system, the fact that there is no calculation error resulting from leakage of neutrons and that an infinite system is not really present must be considered in the safety margins. Consequently, when JACS or another adequately verified calculational code system is to be used, the **estimated criticality lower limit multiplication factors** of infinite systems shall be:

$$k_{\infty} = 0.98 \quad (2.11)$$

2.4.3. Methods for Determining Criticality Limits

Criticality limits need to be provided for **single units** with respect to the shape and dimensions of the equipment containing the nuclear fuel material, the concentration of nuclear fuel material in solution, the mass of the nuclear fuel material, the isotopic composition of the nuclear fuel material, and the shape and dimensions, concentration, and material quality of neutron absorbers. For **multiple units**, besides the configuration of contained **single units**, the configuration, and shape and dimensions of the neutron shielding material, or the combination of these, according to the requirements for criticality control, must be considered.

In determining the **criticality limits**, sufficient safety margins need to be provided for the physical and chemical properties of the nuclear fuel material handled, and the neutron absorption, moderation, and reflection conditions and effects. The fluctuations of the respective states also must be considered.

Criticality limits are provided to help operators make easy and correct judgment of the appropriateness of specific design and operating conditions of the equipment concerned. Consequently, equipment must not be allowed to be fabricated nor be allowed to operate in a way that will exceed these values in normal operation. Thus, **criticality limits** establish the limit values beyond which the **criticality control factors** must not be exceeded in normal operation. **Criticality control factors** are allowed to exceed the **criticality limits** due to a **single failure, etc.**, but even in that instance the **criticality limits** must be selected so that they do not exceed maximum permissible limits.

that instance the **criticality limits** must be selected so that they do not exceed maximum permissible limits.

2.5. Cautions for Each Method of Criticality Safety Control

Criticality safety control must be performed carefully according to the methodology described above. This section describes general cautions to be followed when criticality safety is to be secured by basic methods of criticality safety control including the shapes and dimensions of equipment, nuclear fuel material mass, nuclear fuel material concentration, neutron moderation, and neutron poison.

2.5.1. General Cautions for the Design of Equipment and Systems, and for the Control of Operations

- (1) No facility or equipment into which nuclear fuel material is not scheduled to flow may have any mechanism that could allow nuclear fuel material to flow into it.
- (2) The system design shall not allow migration of a significant quantity of nuclear fuel material from a safe form to an unsafe form to occur even if a single failure should occur.
- (3) The system design shall ensure that the leakage of nuclear fuel material from equipment or from confined locations will not cause criticality.
- (4) Sufficient operational control shall be performed so that the inflow quantity, outflow quantity, and concentration of nuclear fuel material can be maintained according to the design specifications related to criticality safety.
- (5) The system design shall ensure that no phase separation can occur where homogeneity is a precondition of criticality safety.

2.5.2. Cautions for Control by the Mass of Nuclear Fuel Material

- (1) When applying mass control, the mass control values of the nuclear fuel material must be determined for each work unit. When there is concern that the nuclear fuel material could reach the critical mass through the occurrence of a **single failure, etc.**, criticality control of the mass must be established, taking overloading into account.
- (2) The total quantity of nuclear fuel material handled by each work unit in all instances must be proved to be either equal to or less than a specified control value.
- (3) When excess accumulations of nuclear fuel material are anticipated, periodic inspections must be conducted for each work unit and, if necessary, these units must be cleaned to avoid such a phenomenon.

2.5.3. Cautions for Control by Equipment Shape and Dimensions

- (1) Assessments of criticality safety should allow for the possibility of a pan, or equivalent, being placed to retain nuclear fuel material solution leaking from or overflowing the containing equipment.
- (2) Measures shall be taken to prevent nuclear fuel material from being transferred into a container of unsafe form provided in an auxiliary system because of ventilation, discharge, reaction, heating, or cooling.
- (3) When a solution needs to be transferred from safe-form equipment to unsafe-form equipment, attention shall be directed to the following points. The safe-form equipment can be connected directly to unsafe-form equipment by a regular solution transferring circuit only under the situation where unsafe-form equipment can receive a solution of infinite medium free of criticality danger. When a solution of infinite medium having the danger of criticality is to be transferred, the transferring circuit should be kept closed until transfer is to occur. The circuit shall only be opened to allow transfer of the solution after confirming that the concentration of the nuclear fuel material in the safe-form equipment is sufficiently low and there is no danger of criticality. In this situation, the transfer circuit must be provided with a proper and reliable shut-off device.

2.5.4. Cautions for Control by the Concentration of Nuclear Fuel Material in Solution

- (1) Control shall be provided to ensure that no mechanism is present that can allow nuclear fuel arrive at a critical concentration. Alternatively, measures shall be provided to ensure that concentration control is performed by measurement and analysis, taking the time required for these tasks into consideration.
- (2) When nuclear fuel material in solution is homogeneous as a precondition, proper measures shall be taken to prevent an excessive increase of the nuclear fuel material concentration due to sedimentation, high polymerization, crystal deposition, unplanned extraction into solvent, or evaporation.

2.5.5. Cautions for Control by Neutron Moderation

Control by neutron moderation, in combination with other control methods, is usually used for dry nuclear fuel materials having low moisture contents. The water tightness of the containers must be assured when this control method is used.

2.5.6. Cautions for Control by the Use of Neutron Poison

Neutron poison is used for equipment having a volume larger than the safe form or when it is necessary to closely position an array of single units. In these situations, the presence of enough

neutron poison should be ensured. When a homogeneous neutron poison is to be used, its concentration shall be determined by redundant or diverse methods, and measures shall be taken to prevent dilution or sedimentation of the neutron poison. When heterogeneous neutron poison is to be used, the continuous presence of the coexisting neutron moderator with the material having a large cross section of neutron absorption should be assured after considering the danger of fire.

2.5.7. Criticality Control by Distance and Shielding Material

Criticality control for a multiple unit shall be performed using adequate distance between single units and employing shielding material.

- (1) Anchor bolts or other devices shall be used to securely fasten all units to ensure that a safe distance between units will be maintained even in case of earthquakes.
- (2) To avoid the danger of a movable container attaining criticality due to an increase in mutual interaction of neutrons as it approaches fixed equipment, a rigid structure for securing the isolation distance shall be provided, if necessary, around the movable container.
- (3) When considering facilities for which water submersion is not expected, measures must be taken against the possibility of submersion including rain, snow, operation of fire extinguishers, breakage of water pipes, etc.
- (4) When neutron shielding material is used, its performance should be guaranteed by considering the danger of fire, etc.

Reference for Chapter 2

- (1) H. Okuno and Y. Naito, Evaluation of Calculation Errors with the Nuclear Criticality Safety Analysis Code System JACS , JAERI-M 87- 057 (1987), (in Japanese)
- (2) M. Murazaki and H. Okuno, Base Data for Looking-up Tables of Calculation Errors in JACS Code System , JAERI-DATA/CODE 99-019 (1999), (in Japanese)

Table 2.1 Geometrical buckling of a bare system

Rectangular parallelepiped

$$: \left(\frac{\pi}{a + 2d} \right)^2 + \left(\frac{\pi}{b + 2d} \right)^2 + \left(\frac{\pi}{c + 2d} \right)^2$$

Circular cylinder

$$: \left(\frac{2.405}{R + d} \right)^2 + \left(\frac{\pi}{H + 2d} \right)^2$$

$$\text{Sphere} : \left(\frac{\pi}{R + d} \right)^2$$

where

a, b, c : Lengths of the three sides of the rectangular parallelepiped

R : Radius of the circular cylinder, radius of the sphere

H : Height of the circular cylinder

d : Extrapolation length

Table 2.2 Geometrical buckling of a system with a reflector

Rectangular parallelepiped

$$: \left(\frac{\pi}{a + 2d_r} \right)^2 + \left(\frac{\pi}{b + 2d_r} \right)^2 + \left(\frac{\pi}{c + 2d_r} \right)^2$$

Circular cylinder

$$: \left(\frac{2.405}{R + d_r} \right)^2 + \left(\frac{\pi}{H + 2d_r} \right)^2$$

$$\text{Sphere} : \left(\frac{\pi}{R + d_r} \right)^2$$

where

a, b, c : Lengths of the three sides of the rectangular parallelepiped

R : Radius of the circular cylinder, radius of the sphere

H : Height of the circular cylinder

d_r : Extrapolation length of a system with a reflector

Table 2.3 Evaluation of calculational errors for a simple-form system with reflectors^{*1}
(Revised Edition - 1987)

| Group name | Effective multiplication factor | | Benchmark calculational result (for information) | | |
|---------------------------------|--|--|--|--------------------|---|
| | Corresponding to estimated criticality value | Corresponding to estimated lower limit criticality value | Number of benchmarks | Standard deviation | Number of benchmarks whose effective multiplication factor is less than the estimated lower limit multiplication factor |
| Homogeneous U, low enrichment | 0.991 | 0.973 | 18 | 0.006 | 0 |
| Homogeneous U, high enrichment | 0.985 | 0.954 | 68 | 0.013 | 0 |
| -Pu | 1.008 | 0.980 | 71 | 0.011 | 0 |
| -MOX | 1.013 | 0.980 ^{*2} | 45 | 0.008 | 0 |
| -MIX ^{*3} | 1.010 | 0.980 | 10 | 0.008 | 0 |
| Heterogeneous U, low enrichment | 0.995 | 0.978 | 88 | 0.007 | 0 |
| -Pu | 1.004 | 0.964 | 9 | 0.010 | 0 |
| -MOX | 0.997 | 0.980 | 58 | 0.007 | 0 |

*1) Calculated value obtained by JACS Code System

*2) 0.992 according to statistical calculation

*3) MIX shall refer to $\text{UO}_2(\text{NO}_3)_2$ - $\text{Pu}(\text{NO}_3)_4$

Table 2.4 Evaluation of calculational errors for a general-form system with reflectors^{*1}
(Revised Edition - 1987)

| Group name | Effective multiplication factor | | Benchmark calculational result (for information) | | |
|---------------------------------|--|--|--|--------------------|---|
| | Corresponding to estimated criticality value | Corresponding to estimated lower limit criticality value | Number of benchmarks | Standard deviation | Number of benchmarks whose effective multiplication factor is less than the estimated lower limit multiplication factor |
| Homogeneous U, low enrichment | 0.989 | 0.958 | 92 | 0.013 | 0 |
| Homogeneous U, high enrichment | 0.989 | 0.954 ^{*2} | 85 | 0.014 | 0 |
| -Pu | 1.004 | 0.973 | 86 | 0.013 | 0 |
| -MOX | 1.018 | 0.980 ^{*3} | 94 | 0.009 | 0 |
| -MIX ^{*7} | 0.991 | 0.950 | 37 | 0.016 | 0 |
| Heterogeneous U, low enrichment | 0.996 | 0.978 ^{*4} | 120 | 0.007 | 0 |
| -Pu | 1.004 | 0.964 ^{*5} | 11 | 0.010 | 0 |
| -MOX | 0.998 | 0.980 ^{*6} | 64 | 0.007 | 0 |

*1) Calculated value obtained by JACS Code System

*2) 0.956 according to statistical calculation.

*3) 0.997 according to statistical calculation.

*4) 0.980 according to statistical calculation.

*5) 0.968 according to statistical calculation.

*6) 0.981 according to statistical calculation.

*7) MIX shall refer to $\text{UO}_2(\text{NO}_3)_2$ - $\text{Pu}(\text{NO}_3)_4$.

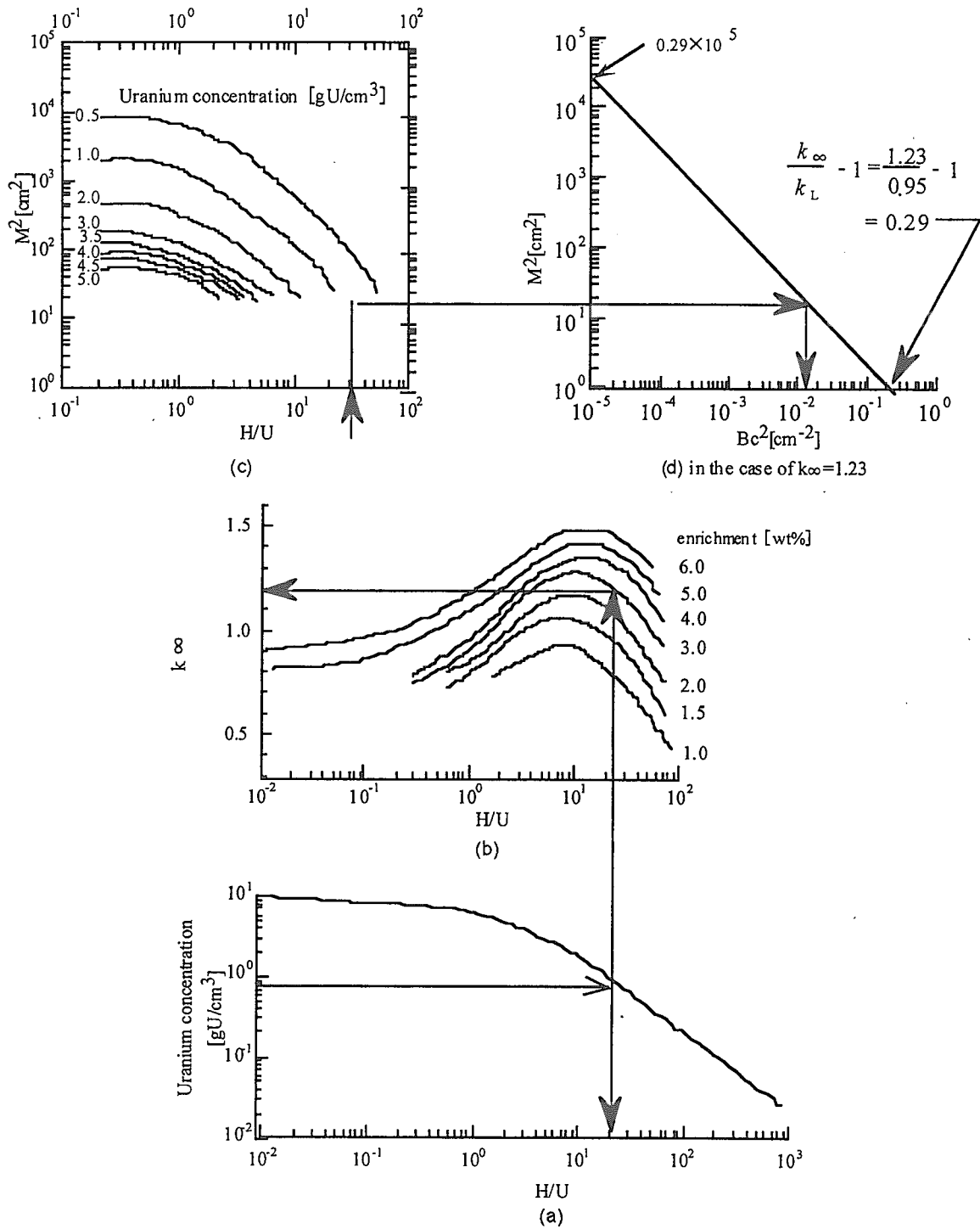


Fig 2.1 Critical buckling chart for homogeneous UO_2 - H_2O
(Example where $k_{\infty}=1.23$)

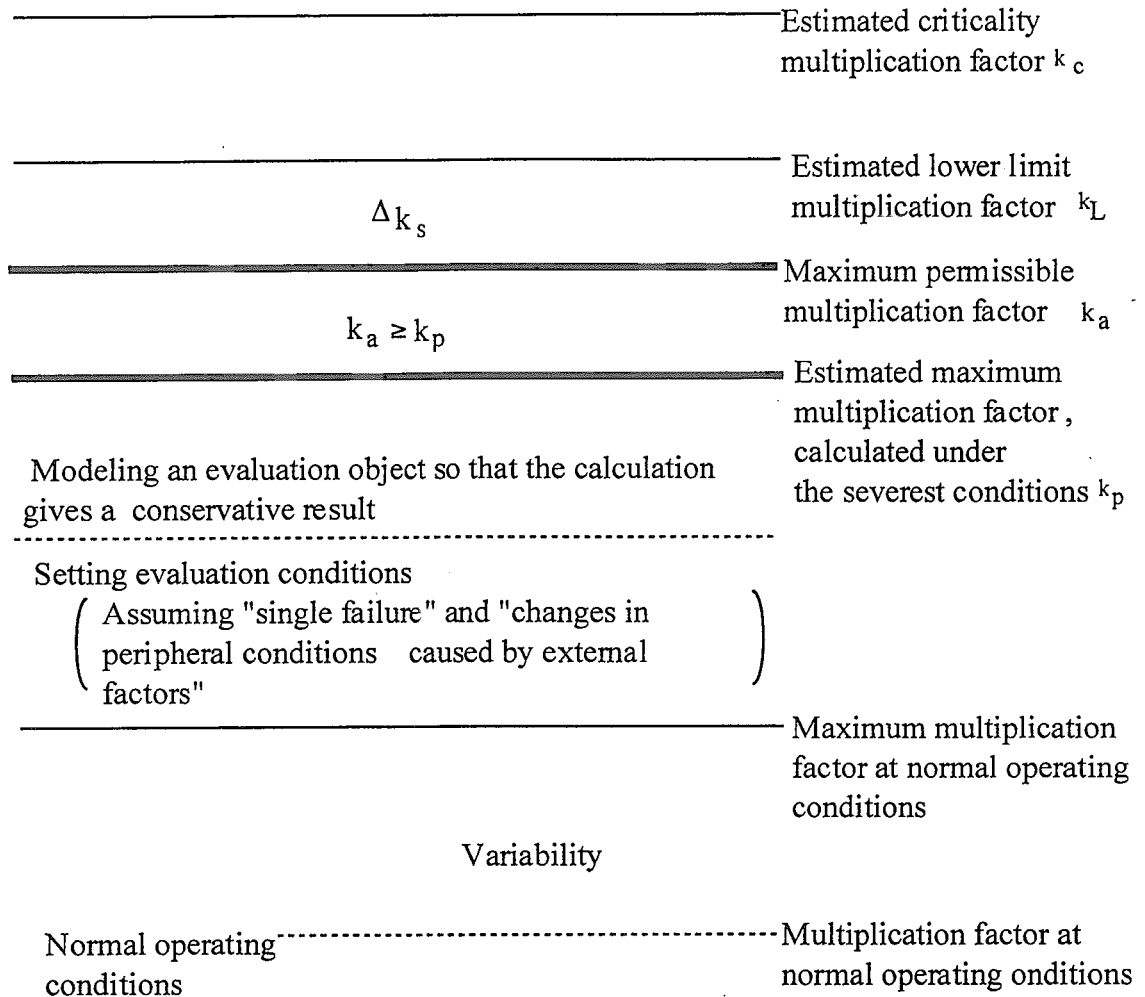


Fig 2.2 Various levels of neutron multiplication factors used in criticality safety design

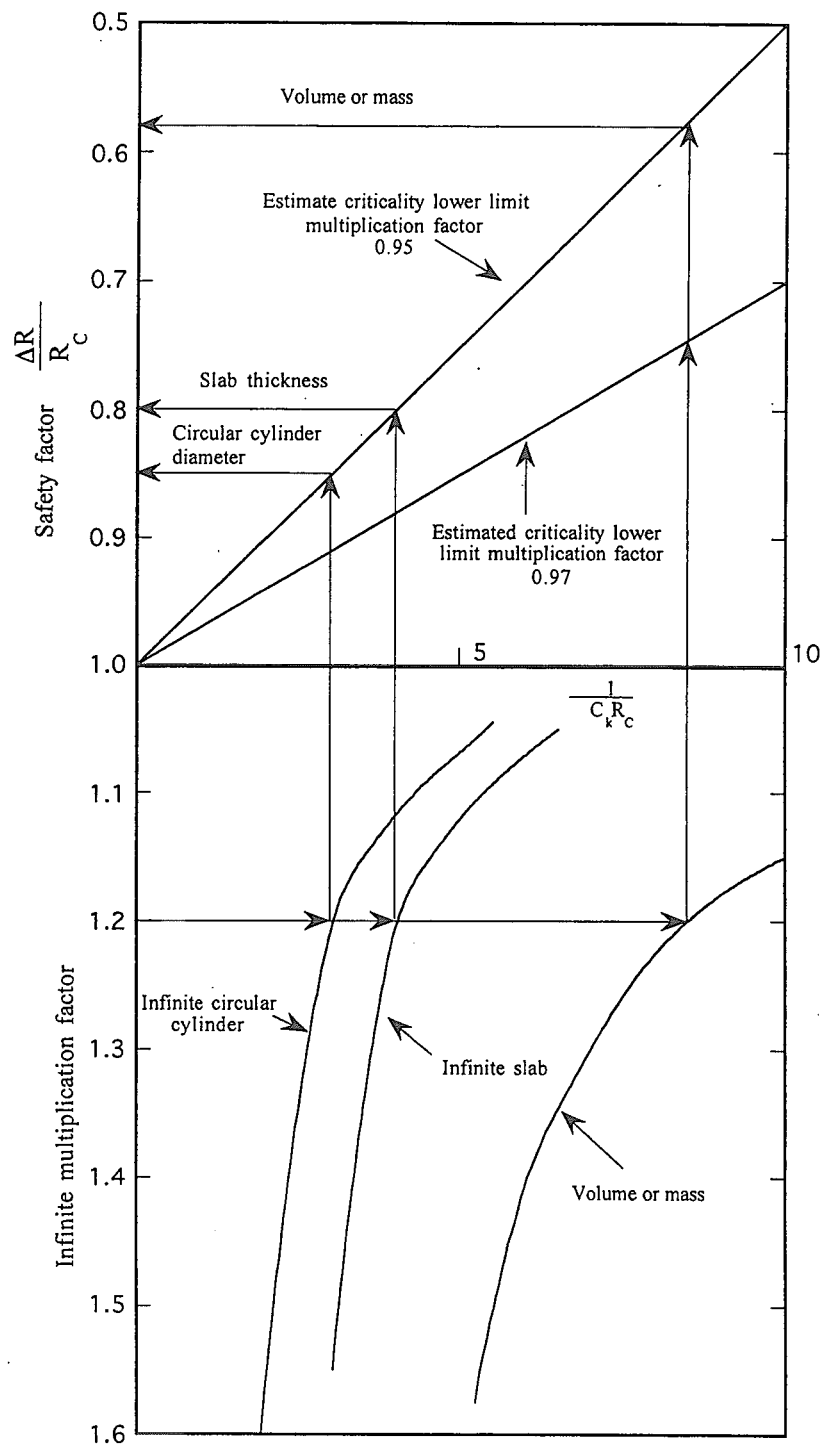


Fig 2.3 Relation between estimated criticality lower limit value and safety factor*

*Safety factor in this diagram refers to the ratio of the diameter of the circular cylinder, thickness of the slab, or volume or mass to those where the effective multiplication factor is 1.0,

3. SETTING OF ASSESSMENT CONDITIONS AND MODELING THE EVALUATION OBJECT

The severest evaluation conditions must be selected when evaluating the criticality safety of an object system considering not only the normal but also abnormal states. That is, the case with the largest neutron multiplication factor must be assumed by taking into account (1) the physical and chemical properties of the nuclear fuel material, (2) the absorption, moderation, and reflection conditions for neutrons, and (3) changes in the peripheral conditions caused by the occurrence of single failure, etc., or due to external factors. In addition, operating parameter variations of the system must be considered. Since the system assumed in this manner is not always a simple one, criticality calculations are often conducted by modeling (simplifying) the assumed system.

The following examples are cases where the neutron multiplication factor of the simplified model becomes larger than the original system:

- (1) Ignoring the wall thickness of a fuel container having a larger absorption effect than reflecting effect (simplifying geometry)
- (2) Setting the perfect reflection condition (albedo = 1) at the outside of a relatively thick finite reflector (simplifying boundary condition)
- (3) Treating a burnup fuel as a fresh fuel without containing neutron poison (simplifying nuclide composition)

In cases where the neutron multiplication factor of the simplified model becomes larger than the original system, the difference in the neutron multiplication factor of the simplified model from that of the original model can be considered as a safety margin for modeling.

In contrast, the following examples are cases where the neutron multiplication factor of the simplified model becomes smaller than the original system:

- (a) Ignoring small branch pipes (simplifying geometry)
- (b) Treating small particle size fuel as a homogeneous fuel (simplifying geometry)
- (c) Ignoring neutrons flowed through from an adjacent region a thick isolating material (simplifying boundary condition)

In the above cases, the difference in the neutron multiplication factor between the simplified model and the original model is called "cut off reactivity". When modeling a system, it is required to make the cut off reactivity sufficiently small and to secure a sufficient safety margin to account for the cut off reactivity.

In this chapter, the cautions are described in selecting the model evaluation conditions applied to modeling of the fuel, moderator, reflector, and absorber and calculation of the atomic number density.

3.1. Modeling of Fuel

In selecting evaluation conditions and modeling a fuel, besides addressing the physical and chemical properties of the fuel, it is necessary to consider the effects of homogeneity, heterogeneity, uniformity, and nonuniformity.

3.1.1. Nonuniform Effects in a Homogeneous System

When mixing or there is a chance of mixing a uranyl nitrate or a plutonium nitrate solution with a sufficiently large concentration differences, the large concentration differences in the solution may remain if a reliable forced mixing mechanism is not applied. However, even if an extremely large nonuniformity in the fuel concentration is created, the relative increase in the neutron multiplication factor of the nonuniform solutions in comparison to the uniform systems after mixing is limited to the following values.

| | |
|------------------------------------|---------------------|
| Aqueous uranyl nitrate solution | 1 [% $\Delta k/k$] |
| Aqueous plutonium nitrate solution | 4 [% $\Delta k/k$] |

These values are applicable to (1) an aqueous uranyl nitrate solution having a ^{235}U enrichment 5 wt% or less and a maximum concentration 1,330 gU/L with a sufficiently thick water reflector, and (2) an aqueous plutonium nitrate solution having a plutonium isotopic composition ratio of 5:1 (^{239}Pu : ^{240}Pu) or a similar value and a maximum concentration 1,000 gPu/L with a sufficiently thick water reflector.

Allowing for a nonuniform fuel concentration could potentially decreases the critical mass. In fact, Goertzel indicated that the critical mass reaches its minimum under a fuel concentration distribution giving a constant thermal neutron flux distribution (so called Goertzel distribution, hereafter called the G- distribution).⁽¹⁾ The G- distribution gives a higher concentration in the fuel center region and a lower concentration in the periphery region.

Using numerical calculation results, Katakura et al. showed the presence of a fuel concentration distribution yielding a smaller critical mass than the flat distribution for a low enrichment uranium fuel.⁽²⁾ Hirano et al. developed a numerical calculation technique under the diffusion approximation for obtaining an optimum fuel concentration distribution, based on fact that the optimum fuel concentration distribution could be obtained at a flat fuel importance distribution.⁽³⁾ They further evaluated the effect of nonuniformity at an actual plutonium handling facility.⁽⁴⁾

Critical experiments were performed simulating the nonuniform effect of homogeneous fuel by using a fuel rod array at the Tank-typed Critical Assembly, TCA, of JAERI. A nonuniform fuel distribution yielding a higher reactivity than the uniform distribution was not found (Yanagisawa et al.⁽⁵⁾). This result was probably due to the narrow range of water to fuel volume ratios examined. Yamane et al. conducted experiments at KUCA using high-enriched uranium fuel plates sandwiched by polyethylene plates, and confirmed that a fuel density distribution with a higher fuel density in the

center region gave a higher reactivity than the flat distribution.⁽⁶⁾

A series of numerical calculations were conducted to evaluate the nonuniform effect of homogeneous fuel, using a calculation code OPT-D for obtaining the optimum fuel concentration distribution based on the flat fuel importance principle.⁽⁷⁾ The behavior of the optimum fuel concentration distribution giving the maximum reactivity was characterized as follows in terms of the mean fuel concentration or the degree of moderation.

- Overmoderated fuel had an optimum fuel concentration of one with a higher concentration in the center,
- Undermoderated fuel had an optimum fuel concentration of one with a higher concentration in the vicinity of the reflector, and
- Medium-moderated fuel had an optimum fuel concentration of one with a high concentration both in the center and at the periphery.

Numerical calculations were performed for a slab fuel model containing an aqueous uranyl nitrate solution or an aqueous plutonium nitrate solution. To remove the heterogeneous effect of spatially oscillating fuel concentration, a constraint condition was set so as to allow only one minimum point in a symmetrical fuel concentration distribution relative to the fuel center. The calculation results of the nonuniform effect showed that the increase rate in the neutron multiplication factor for the 5 wt. % ^{235}U enriched uranyl nitrate aqueous solution was only about 1 % (Table 3.1), whereas that for the plutonium nitrate aqueous solution having a plutonium isotopic ratio of 5:1 (^{239}Pu : ^{240}Pu) was 3% to 4% (Table 3.2). Fig. 3.1 and 3.2 show examples of obtained optimum fuel concentration distributions. The fuel distribution of these examples corresponds to the medium-moderated fuel discussed above, i.e., there is a high fuel concentration near the reflector with remaining fuel located so as to produce a slightly higher concentration in the center region.

Using the same constraint condition above, the effect of nonuniformity was evaluated for a homogeneous 5 wt% ^{235}U -enriched $\text{UO}_2 - \text{H}_2\text{O}$ fuel in a 10 cm-thick slab with a full water reflector. Fig. 3.3 show the calculation results. The neutron multiplication factor reached its maximum around 2,500 gU/L under a uniform fuel distribution. In contrast, it reached its maximum at a mean concentration of 3,000 gU/L under a nonuniform fuel distribution, and the multiplication factor of the optimum distribution was about 4% larger than that of the uniform distribution.⁽⁸⁾

The nonuniformity effect was considered most significant with a slab fuel geometry compared with other convex type fuel geometries. In fact, the calculation results for homogeneous $\text{UO}_2\text{-H}_2\text{O}$ fuels showed that among spherical (diameter 34 cm), circular cylindrical (diameter 22 cm), and slab (thickness 10 cm) geometries giving the same neutron multiplication factor, the rate of increase in the neutron multiplication factor due to the nonuniformity effect under the optimum moderation (1.8 gU/cm³) were 0.5%, 1.7%, 2.9%, respectively.⁽⁸⁾

It was revealed that the diffusion approximation based optimum fuel concentration distribution calculation code, OPT-D⁽⁷⁾, was applicable to a system with a reflector, whereas it was not applicable to a system without a reflector due to larger calculation errors. Thus, another fuel concentration

distribution calculation code, OPT-SN, was developed, which solves neutron transport equations by the discrete-ordinates method, and the nonuniformity effect of a system without a reflector was calculated using this code. The optimum fuel concentration distribution and the neutron multiplication factor were calculated for a 5 wt% ^{235}U -enriched uranium dioxide-water (no voids) fuel contained in a 10 cm thick plate without a reflector. The mean fuel concentration was varied in the range from 700 to 4,000 gU/L. Fig. 3.4 shows the neutron multiplication factor under the optimum fuel concentration distribution in comparison to that under the uniform distribution.⁽⁸⁾ This figure shows that the optimum moderating condition of this fuel system falls within the fuel concentration range of 1,500 to 2,000 gU/L, and the nonuniformity effect in the neutron multiplication is 1% $\Delta k/k$ or less in this range, whereas outside of this range it is several % $\Delta k/k$.

The nonuniformity effect is significant when the fuel solution concentration varies by several hundred gU/L as shown in Fig. 3.1. Therefore, it is not necessary to consider the nonuniformity effect when modeling a tank just containing a uranyl nitrate solution or a plutonium nitrate solution, since the concentration in the tank under normal conditions is virtually constant.

3.1.2. Nonuniform Effects in a Heterogeneous System

A nonuniform state in a heterogeneous system occurs when fuel elements are irregularly arrayed in a moderator or when fuel elements having density differences are regularly arrayed. Basically, the considerations are the same as for the nonuniform effects in a homogeneous system.⁽⁹⁾ However, experiments⁽¹⁰⁾⁽¹¹⁾ and theory⁽⁶⁾ confirm that the reactivity of fuel elements piled randomly is lower than the reactivity of fuel elements arrayed regularly. Consequently, criticality safety should be assessed by modeling the fuel on a regular lattice array at the optimum pitch for criticality.

3.1.3. Particle Size of Fuel to Be Regarded as Homogeneous

Heterogeneous fuel systems without the existence of moderator among fuel particles can be modeled as homogeneous systems, irrespective of fuel particle sizes. Powder, slurry, or sludge fuel systems (heterogeneous systems) with water (moderator) present among fuel particles cannot be regarded as homogeneous, since the neutron moderation and diffusion behavior of these heterogeneous fuel systems are different from those of corresponding homogeneous systems. As the fuel element particle size approaches zero, however, there exists a range of particle sizes where these heterogeneous systems can be regarded as homogeneous from the neutron behavior viewpoint.

In the criticality safety evaluation of a mixed system of uranium dioxide and water (uranium enrichment 10 wt% or less, mean uranium concentration from 1 gU/cm³ to 3 gU/cm³), the heterogeneous system can be regarded as homogeneous provided that the fuel particle size is less than the size in mm given by Equation (3.1).

$$a = \frac{\varepsilon_g}{\varepsilon_G} \quad (3.1)$$

where ε_g is the maximum allowable percent rate increase of the infinite multiplication factor when regarding a heterogeneous system as a homogeneous system (i.e. if the rate increase is more than ε_g , the heterogeneous system cannot be regarded as homogeneous) and ε_G is $\Delta k/k$ expressed in % in Fig. 3.5.

When evaluating the criticality safety of a mixed oxide fuel of uranium and plutonium (hereafter called MOX fuel) immersed in water, the same equation can be applied. It should be noted that the appropriate ε_G value is ten times the % $\Delta k/k$ given by the solid line in Fig. 3.6. The equation is applicable to a plutonium enrichment (oxide mass ratio) of no more than 10 wt%, an isotopic composition of 71:17:12 (^{239}Pu : ^{240}Pu : ^{241}Pu) (metal mass ratio), the isotopic composition of natural uranium, and a mean MOX concentration range of 0.5 to 1.1 g ($\text{UO}_2 + \text{PuO}_2$)/cm³.

Powder, slurry, and sludge fuels are often processed in nuclear fuel facilities. Slurry is a fluid mud-like mixture in which fine solid fuel particles are suspended or dispersed in the solution. For example, during uranium conversion, slurry is formed as uranium precipitates when an alkaline material is added to uranyl nitrate water solution. A material with less fluidity and a high solid fuel content, for example filtered slurry, is called cake or sludge.

From the criticality safety evaluation viewpoint, when the increase rate of the multiplication factor of a heterogeneous system relative to its corresponding homogeneous system is small (e.g. 0.3% $\Delta k/k$ or less), the heterogeneous system may be regarded as homogeneous, even if the reactivity of the heterogeneous system is higher than that of the homogeneous system. To evaluate the effect of fuel heterogeneity, the infinite multiplication factor of a small cell containing sphere particles of a low-enriched uranium dioxide fuel immersed in water was calculated. The water to fuel volume ratio was set as the optimum moderation for the homogeneous system was achieved. The increase rates of the infinite multiplication factor (% $\Delta k/k$) relative to corresponding homogeneous systems are given in Fig. 3.7.⁽¹²⁾ The increase rate (% $\Delta k/k$) of uranium enrichment 3 wt% is almost identical to that of 5 wt%. At higher ^{235}U enrichment of 10 wt% and 20 wt%, the increase rate (% $\Delta k/k$) becomes smaller, and at 40 wt% the increase rate (% $\Delta k/k$) decreases with increasing fuel sphere diameter. The primary reason for this behavior is the resonance escape probability (p). Fig. 3.8 shows change rates in the infinite multiplication factor and its four factors in the 5 wt% ^{235}U enriched fuel particle range from 0 to 1 mm in diameter.⁽¹²⁾ The figure demonstrates that the change rate of the infinite multiplication factor ($\Delta k/k$) is virtually dependent on $\Delta p/p$, and that these two values increase almost proportionally to the fuel particle diameter.

Calculations were performed for a 5 wt% ^{235}U -enriched system with various fuel volume ratios. Fig. 3.5 shows the change rate in the infinite multiplication factor k relative to the homogeneous system as a function of mean uranium concentration for a fuel system containing 1 mm diameter fuel particles.⁽¹²⁾ This figure demonstrates an almost linear change in $\Delta k/k$ as a function of mean uranium concentration between 1 and 3 gU/cm³. Fig. 3.5 also includes data for 3 and 10 wt% uranium

enrichment. The function obtained for 5 wt% enrichment is virtually independent of the enrichment level.

According to Fig. 3.7 and 3.8, $\Delta k/k$ is proportional to the fuel particle diameter for particle diameters less than or equal to 1 mm. The fuel particle diameter, a , in mm that can be regarded as homogeneous can be obtained from Equation (3.1), where ϵ_g [%] (e.g. 0.3 %) is the rate of the infinite multiplication factor increase that can be ignored, and ϵ_G [%] is $\Delta k/k$ value corresponding to the mean uranium concentration (or water to fuel volume ratio) of the object system (given by Fig. 3.5). For example, consider a uranium dioxide-water heterogeneous system with a 5 wt% enrichment and a mean uranium concentration of 2 gU/cm³. Assume that the increase rate of the infinite multiplication factor that can be ignored is 0.3 %. Inserting $\epsilon_g = 0.3$ and $\epsilon_G = 1.28$ into equation 3.1, the fuel particle diameter that can be regarded as homogeneous becomes 0.23.

$$a = \frac{0.3}{1.28} = 0.23 \text{ [mm]} \quad (3.2)$$

Thus, a fuel particle diameter of 200 μm , which is no larger than 0.23 mm, can be regarded as homogeneous.

Similar cell calculations were conducted for a MOX fuel and the result for a fuel particle diameter of 0.1 mm is given in Fig. 3.6. The figure shows that the rate of increase in the infinite multiplication factor (homogeneous vs. heterogeneous) increases with the plutonium enrichment. The solid line is the case for a plutonium enrichment of 10 wt%. The range of MOX fuel concentration evaluated for the experiment is in the vicinity of optimum moderation of the homogeneous system. In this fuel concentration range, supposing that the increase rate of the infinite multiplication factor that can be ignored is 0.3 %, the MOX fuel particle size that can be ignored as homogeneous is calculated to be 80 μm or less.

Actual fuel particle diameters of uranium dioxide powder vary with manufacturing and measurement methods. It is reported that among three measuring methods (gas absorption, permeability method, air sedimentation method) air sedimentation method gives the largest particle diameter, 5 to 50 μm .⁽¹³⁾ The fuel particle diameters of MOX fuel powder processed by Power Reactor and Nuclear Fuel Development Corporation were reported to be relatively small, ranging from 1.7 to 2.0 μm .⁽¹⁴⁾

3.1.4. Heterogeneous Effects of Low-enriched Uranium System

In a heterogeneous system of low enriched fuel, care must be taken because modeling as a homogeneous system may underestimate the effective multiplication factor. In a low enriched fuel, the effect of neutron resonance absorption by ²³⁸U is significant. In the heterogeneous system, however, water existing between fuels moderates neutrons and the probability of avoiding such resonance increases. In a uniformly mixed system of fuel material and moderator, considering only the density of the mixture, such heterogeneous effects are lost and the neutron multiplication factor may be

underestimated. This underestimating tendency in case of uranium fuel depends on ^{235}U enrichment and dimension of fuel (The resonance absorption of ^{238}U decreases as enrichment increases. Refer to Fig. 3.7.) Therefore, in case of the heterogeneous system using a low enriched uranium fuel, modeling should take account of actual heterogeneous rather than using a simple volume averaged model.

3.1.5. Modeling Fuel Solutions of Heterogeneous Systems

When a heterogeneous fuel is present in a fuel solution, such as occurs in a dissolver tank of a reprocessing facility, there are many parameters, including the fuel solution concentration and fuel rod diameters, which should be considered for the criticality safety assessment. There are reports⁽¹⁵⁾⁽¹⁶⁾⁽¹⁷⁾ (also refer to section 5.3.3) of a model that shows higher reactivity where the fuel solution acting as a moderator is replaced by water and the optimum moderation condition is assumed. The reason may be that the resonance escape probability of neutrons will increase when a fuel solution acting as a moderator is replaced by water. The calculational examples in these references show that the model replacing the fuel solution with water, assuming the optimum moderation condition, leads to an overestimation of the effective multiplication factor in a system of low-enriched uranium fuel. Consequently, this is thought to be a conservative assessment model (one that is pessimistic).

3.1.6. Modeling of Complex Geometric Shapes

When it is difficult to make an analysis because of the presence of a complex fuel array or shapes of equipment, a simplified model is used for the analysis. However care must be exercised not to underestimate the reactivity by making a model that overstates the fuel area.

Often many fuel rods are arrayed in water with lattice spacing that varies from one blocked region to another, as in a spent fuel storage pool. In this situation, it is possible to determine an effective multiplication factor by calculating the whole system with a simplified block-homogenized cell model. This model can be obtained by cell calculations where a heterogeneous cell, which is constructed from the fuel rod and moderator, is replaced with a homogeneous cell for each region having an equal lattice-spacing. When setting up the cell model, consideration should be given to ensure the reactivity is not underestimated.

3.1.7. Modeling of Pipes

Complicated fuel handling units containing pipes and valves are common in reprocessing plants. Performing numerical calculations using the actual complicated geometry leads to many difficulties, e.g. input data preparation becomes complex and laborious and the criticality conditions described in this handbook can no longer directly be applied to the geometry. To cope with these difficulties, criticality calculations may be performed using a simplified geometry by configuring each unit using

multiple spheres, circular cylinders, rectangular parallelepipeds, slabs or combinations of these shapes. For example, the neutron multiplication factor k for a system including a vessel filled with fuel solution (hereafter called a main vessel) with thin pipes (hereafter called branch pipes) intersected may be evaluated by the following equations⁽¹⁸⁾.

$$k = \frac{k_0}{(1-\delta)} \quad (3.3)$$

$$\delta = \sum_{i=1}^n \delta_i \quad (3.4)$$

$$\delta_i = \frac{1}{1 - \frac{k_i}{k_0}} \left(\frac{L}{L+A} \right)_0 \left(\frac{L}{L+A} \right)_i \omega_{0i} \omega_{i0} \quad (3.5)$$

where n is the number of branch pipes, and 0 is the subscript for main vessel and i is the subscript for branch pipes.

k_i :neutron multiplication factor of unit i ($i=0, 1, \dots, n$),

$\left(\frac{L}{L+A} \right)_i$:leakage ratio from unit i ($i=0, 1, \dots, n$),

ω_{0i} :the fractional solid angle when looking at unit i from the center of unit 0 ($i=1, \dots, n$)

ω_{i0} :the maximum fractional solid angle when looking at unit 0 from unit i ($i=1, \dots, n$)

In particular, when branch pipes are connected perpendicularly to the main vessel side surface, the reactivity effect of the branch pipes, irrespective of their length, can be neglected, provided that the diameters of the branch pipes and the main vessel are so related that they are below the curves shown in Fig. 3.9. However, the relative error in the neutron multiplication factor associated with neglecting the branch pipes must be less than $0.3\% \Delta k/k$ (Here, $0.3\% \Delta k/k$ was selected as the relative neutron multiplication factor that can be neglected). The above equations are applicable in the range where the neutron interactions among the branch pipes are negligibly small relative to the neutron interaction between the main vessel and the branch pipes. These equations, however, cannot be applied to a system surrounded by reflectors.

In a nuclear fuel plant, there are many cases in which small fuel elements are situated in the vicinity of a large fuel unit. A typical example is a fuel solution tank intersected by small pipes. It is generally difficult to accurately calculate the neutron multiplication factor of such a complicated geometry. To cope with the complicated geometry, a Monte Carlo code may be used, but the input data creation is complicated, and a high calculation accuracy is required for analyzing the effect of branch pipes. Thus, the computation time becomes much longer. When the reactivity increment of branch pipes is very small, the branch pipes can be neglected. When the reactivity increment of the branch pipes is not negligible, the reactivity increment of the branch pipes shall be calculated by a

simple calculation method and added to the reactivity of the main vessel.

Equations (3.3) through (3.5) were obtained based on the neutron balance calculations. These equations are basically the same equations applied to the criticality safety assessment code MUTUAL for multiple unit fuel systems, except that they consider the deviation of the representative neutron source point from the center of each pipe. The results based on these equations were compared with the output of the Monte Carlo code KENO IV for three-dimensional fuel systems of 5 wt% ^{235}U ⁽¹⁸⁾ enriched uranyl nitrate solution. The KENO-IV geometry consisted of a main vessel and four branch pipes, both of which were modeled as cuboid. The wall thickness of the main vessel and branch pipes was neglected. On each center of the four side-surfaces of the cuboid (length of each side: 32 cm; height: 1m), one thin cuboid (length of each side: variable; length: 50 cm) was perpendicularly attached. The calculation results based on Equation (3.5) are shown in Table 3.3 and its comparison with KENO-IV results are shown in Fig. 3.10. It was found that Equation (3.5), giving a higher multiplication factor than KENO-IV, could be fully used as an equation giving a conservative result.

Based on these equations above, the diameter and the number of branch pipes that can be neglected were calculated and graphically shown in Fig. 3.9.⁽¹⁸⁾ The object system in this case contained a cylindrical main vessel with cylindrical branch pipes perpendicularly connected to the main vessel. The parameters used were $\omega_{i0}=0.5$, $\omega_{i0} = (d_i/d_0)^2/4$, neutron migration area in solution $M^2=40 \text{ cm}^2$, and an extrapolation distance $=3 \text{ cm}$. Here, d_0 and d_i is the diameter of the main vessel and the branch pipes, respectively.

3.1.8. Temperature Effects of Fuel Solution

For uranium fuel solution temperatures that are higher than normal, normal temperatures may be used in the evaluation safely. However, reactivity may increase with an increase in temperature for systems containing plutonium.

An increase in temperature generally decreases the density of fuel material in a system and increases the leakage of neutrons. Furthermore, an increase in temperature raises the neutron temperature, which results in a decrease in the ratio of nuclear fission to neutron absorption. Even in the various processes that are accompanied by temperature increases, there generally will be no criticality safety problem for uranium fuels if assessment is made at normal temperature because reactivity will normally decrease with an increase in temperature.

However, reactivity may increase with a temperature rise in a system containing plutonium because a temperature increase may unsafely affect the system's nuclear properties.⁽¹⁹⁾ Thus, for plutonium systems, modeling should consider any elevation in temperature.

At JAERI, a plutonium nitrate solution including gadolinium in a circular-cylindrical system 39 cm in diameter \times 100 cm in height with a sufficiently thick water reflector was examined. Temperature coefficients were calculated as a function of the fuel concentration for some gadolinium concentrations.⁽²⁰⁾ The results are shown in Fig. 3.11. In this example, plutonium nitrate solution without the gadolinium poison, the temperature reactivity coefficient does not become positive until

the concentration decreases to about 15 gPu/L. In contrast, the temperature coefficient becomes positive at a higher plutonium concentration for plutonium nitrate solution containing gadolinium. This tendency is more significant at higher gadolinium concentrations. At a gadolinium concentration of 0.25 g/L, the temperature coefficient is still positive at a plutonium concentration as high as 200 gPu/L.

3.1.9. Density and Moisture Content in Oxide Powder Fuel

In selecting the density and moisture content of fuel for a calculational model used to assess criticality safety of uranium oxide powder fuel, the maximum values that can be conceived in the actual operation should be considered after sufficiently studying the oxide powder manufacturing process and the extent of moisture control for the facility and equipment.

The critical mass will decrease with an increase in density, if this increase in density is not accompanied by compensating changes in moisture content or changes in material composition. Fig. 3.12 shows a calculation example.⁽²¹⁾ When the changes in density are accompanied by changes in moisture content, however, the fuel will have the maximum reactivity when it has optimum moderation. The moisture content in ordinary oxide powder fuel provides undermoderated conditions compared with optimum moderation. Reactivity increases when the moisture content is increased. Similarly, reactivity increases when density is increased. Fig. 3.13 through 3.15 show calculation examples for a sphere, circular cylinder, and slab geometry, respectively.⁽²¹⁾

In a reported example⁽²²⁾ where the criticality conditions of UO_2 powder were calculated using the density and moderation ratio as parameters, the critical mass decreased with an increase in density and the hydrogen to uranium (H/U) ratio, as shown in Fig. 3.16. In this figure, the powder density is fixed and the changes in the moderation ratio are caused by the entry of water into the voids in the powder. Consequently, the place where the H/U ratio is a maximum is where the powder fuel and water are homogeneously mixed and in a void-free state.

It is reported that the density of oxide powder fuel extends over a wide range; that is, the density of UO_3 is 2.5-4.3 g UO_3/cm^3 ,⁽²²⁾ the tap density of UO_2 powder is 1.3-5.7 g UO_2/cm^3 , and the bulk density is 0.6-4.0 g UO_2/cm^3 .⁽²³⁾

There are calculation examples in which the powder density of PuO_2 changes from 1.25-4.0 g PuO_2/cm^3 .⁽²⁴⁾⁽²⁵⁾ These references provide useful information. The moisture content when the fuel is in air is a few percent at most. These references also report examples where the maximum moisture content is set at 5% in safety analyses.⁽²⁴⁾⁽²⁵⁾

Therefore, unless unexpected introduction of water is possible, the maximum density and moisture content that are conceivable may be used in modeling ordinary fuel powder.

3.1.10. Chemical forms, etc., of Fuel

In a process where a chemical reaction takes place, the nuclear fuel and other materials undergo

changes in chemical and physical properties. In a system where materials of different chemical and physical forms are together, the safety assessment should be made with the combination of material forms that would result in the highest reactivity conceivable.

As a specific example, in the denitration process of a reprocessing facility, the chemical and physical forms of the fuel material change from an aqueous solution of uranyl nitrate to uranium oxide powder. The moisture content also changes due to evaporation, etc.

In the conversion process of a uranium processing facility, there is an example where the chemical composition of ADU (ammonium diuranate), produced as an intermediate product, varied with pH as follows.⁽¹⁷⁾

- i) $3\text{UO}_3 \cdot \text{NH}_3 \cdot 5\text{H}_2\text{O}$
- ii) $2\text{UO}_3 \cdot \text{NH}_3 \cdot 3\text{H}_2\text{O}$
- iii) $3\text{UO}_3 \cdot 2\text{NH}_3 \cdot 4\text{H}_2\text{O}$

Reactivity increases as the nitrogen to uranium (N/U) value decreases, so the criticality for ADU is calculated with the item i) chemical composition in "Data Collection of Nuclear Criticality Safety Handbook."

3.1.11. Fuel Treatment Adopting Burnup Credit

A criticality safety evaluation may be conducted taking account of the fuel burnup. In the safety evaluation, however, it must be independently confirmed by a certain method that the actual fuel burnup exceeds the burnup assumed in the evaluation. For setting of the nuclide composition of the fuel burnup, refer to section 3.5.2 "Changes in nuclide composition resulting from burnup." When setting the nuclide composition by a fuel burnup calculation, it must be assumed that the fuel was "burned" at a high power density and was subsequently cooled using the shortest cooling time allowed. Fig. 3.17 shows an example calculation of the effect of the cooling time on the reactivity.⁽²⁷⁾ In general, the reactivity decreases as the cooling time increases due to the transmutation of ^{241}Pu to ^{241}Am and the accumulation of fission products (hereafter called FP_s) which have a large neutron absorption cross section. A notable exception to this rule is the initial peak period associated with short-lived FP nuclide decay. Calculation results are given in reference (28), using initial enrichment, burnup, cooling time, etc. as parameters.

When a fuel contains burnable poisons such as gadolinium, the reactivity worth increases as the fuel burns up. In this case, the criticality evaluation may be conducted by assuming the maximum reactivity reached in the burnup period. Further depletion related reactivity decreases may also be considered in the evaluation. Fig. 3.18 depicts the relation between burnup and the infinite multiplication factor (hereafter called k_∞).⁽²⁹⁾ Here, a BWR fuel assembly was burned up under a moderator void coefficient (volumetric percentage) of 40%. In this example, k_∞ of the burnup of the BWR fuel assembly increases up to 8 GWd/t due to the depletion effect of gadolinium, and decreases

thereafter. When a fuel assembly contains gadolinium, the safety evaluation may be conducted using a nuclide composition corresponding to a burnup giving the maximum k_{∞} . When the fuel further depletes beyond a burnup giving the maximum k_{∞} , the nuclide composition corresponding to the further depletion may be used for the criticality safety evaluation so long as the actual burnup can be verified.

For PWR spent fuel with a uranium enrichment of no more than 5 wt% and an assumed burnup of no more than 20 GWd/t, it can be assumed in the criticality safety evaluation that the fuel depletes uniformly in the longitudinal direction. When assuming a fuel with a burnup of 20 GWd/t or more, the effect of burnup distribution needed to be considered. When ignoring the formation of FPs, however, the burnup distribution effect becomes smaller, and thus this burnup distribution effect needs to be considered at burnup values higher than 20 GWd/t. Fig. 3.19 shows the neutron multiplication factor for a PWR fuel assembly of an infinite array accounting for the burnup distribution in the axial direction in comparison to the neutron multiplication factor ignoring the burnup distribution.⁽²⁹⁾ Above a burnup of 30 GWd/t, the neutron multiplication factor taking account of the burnup distribution exceeds the neutron multiplication factor without taking account of the burnup distribution. In this example, the FP effects are considered. When the FP effects are ignored in a criticality safety evaluation, the reactivity worth from the burnup distribution is small. In addition, this example assumed a vertically symmetrical axial burnup distribution. If this assumption is not valid, from the beginning of the fuel depletion, the neutron multiplication factor accounting for the burnup distribution may exceed the neutron multiplication factor ignoring the burnup distribution.

Voids in moderator strongly affect the nuclide composition of BWR spent fuel. To obtain a conservative evaluation result, a criticality safety evaluation can be conducted by setting a high moderator void coefficient and by assuming an averaged burnup uniformly distributed in the entire fuel length. However, the enrichment distribution of a BWR fuel assembly is more complicated than that of PWR, and the burnup conditions in reactor are affected by moderator voids and control rods. Therefore, these effects need to be carefully analyzed individually.

In the chopping and dissolution processes of a reprocessing plant, fuel is handled as chopped pieces. As the result, only fuel end pieces with lower burnup may be processed together. Care must be taken in selecting a burnup value to account for this case. In extraction and down stream processes, since the isotopic composition of fuel can be measured, subcriticality can be verified by simply confirming that the actual isotopic composition satisfies criticality limit conditions.

3.2. Modeling of Moderator

3.2.1. Water Density between Multiple Units

The density of the water moderator that may be present between multiple units should be the severest conceivable for reactivity.

When many single units are stored, such as fuel assemblies in dry or wet storage, the density of

water present between units can vary from 0 to 1.0 g/cm³ and there are some cases that show a higher reactivity with low density water. This point should be heeded in making a criticality safety assessment. However, when the possibility of low water density is thought to be negligibly small, this may be omitted from the items considered.

Even when actuation of a fire-extinguishing sprinkler in a dry storage installation is considered, water density in all ranges need not be considered in preparing an assessment model. For example, there are only a few water density ranges that are conceivable under various conditions.^{(30) ~ (37)} According to those reports, the conceivable ranges are:

| | |
|-------------------------|-----------------------------|
| Moist air | 0 ~ 0.006 g/cm ³ |
| Fire extinguishing foam | 0 ~ 0.004 g/cm ³ |
| Water from a sprinkler | 0 ~ 1.000 g/cm ³ |
| Mist | 0 ~ 0.004 g/cm ³ |
| Rainfall | 0 ~ 0.030 g/cm ³ |
| Snowfall | 0 ~ 0.100 g/cm ³ |

For example, in a spent fuel water pool in a storage installation, if the water-circulation cooling system has stopped functioning due to a power failure or other cause, the pool water temperature will rise due to the decay heat from the spent fuel. The temperature increase will decrease water density. As a result, reactivity may increase. In this situation, an abnormal rise in pool water temperature that causes the abnormal lowering of water density may be prevented by switching to an emergency power source or by actuation of a spare cooling and water supply system. When these backup systems are available and the probability of abnormal changes in water density is judged to be negligibly small, the temperature rise of the pool water may be eliminated from consideration in assessing criticality safety.

In a dry storage installation, the reactivity of the system may increase due to the introduction of a water moderator between the fuel assemblies due to causes that include water spray from the actuation of a fire extinguishing system, abnormally high tide, or inundation. For these unusual occurrences, if the maximum quantity or extent of the water moderator possible can be determined from records of natural disasters or extremes, or if the installation includes systems that preclude the introduction of water, criticality safety may be assessed within the conceivable ranges.

3.2.2. Modeling of Air

Nitrogen, oxygen, and a small amount of moisture are contained in air, altogether having a density of about 1.3×10^{-3} g/cm³. The influence of these trace components is negligibly small. Therefore, when assessing the criticality safety of a nuclear fuel handling installation, air may be modeled as a vacuum.

3.2.3. Modeling Neutron Shielding Materials

When a material having a hydrogen content higher than water, such as polyethylene, paraffin, or acrylic, is used as a neutron shield, it may be replaced by water in a criticality safety model.

Normally, polyethylene, acrylic, water, and concrete are used as shielding materials for neutrons. In TID-7016 Rev.2⁽³⁸⁾ and others, water is used in the assessment as a neutron shielding material in the inner container of an annular tank. Water is used most commonly as a neutron shield, but other neutron shielding materials having a high hydrogen content are sometimes used. A model for one of these materials may be prepared by replacing the material with water for assessment of criticality safety. This is permissible because the material has a neutron shielding capacity greater than water, as shown in Fig. 3.20.

3.2.4. Modeling Moderator in Fuel

When it is possible that moderating materials other than water may be contained in fuel, modeling should include the severest reactivity conditions for the moderators.

There are many cases in which water is used as a moderator in a nuclear fuel material criticality evaluation. A material that contains a high percentage of hydrogen, including polyethylene, paraffin, and Plexiglas works well as a neutron moderator.⁽²⁵⁾ Consequently, criticality safety should be assessed for the conditions where the moderator will cause maximum reactivity considering the existence of these materials in the fuel.

In the criticality safety evaluation of nuclear fuel handling facilities, it is required to adopt sufficiently conservative assumptions when modeling an object system. When evaluating the criticality safety of fuel reprocessing plant extraction processes that use organic solvents, the organic solvents are often modeled as water. It is necessary to study if this assumption is sufficiently conservative.⁽⁴⁰⁾

3.2.5. Free Nitric Acid in Solutions of Nitrates

The lowest value that is conceivable should be used as the free nitric acid concentration in an aqueous solution of uranium nitrate or plutonium nitrate.

The atomic nucleus of the nitrogen contained in the nitric acid has a relatively large neutron absorption cross section. It therefore has a negative contribution toward reactivity. Consequently, for assessing criticality safety, the concentration of the free nitric acid should be set at zero or at the lowest conceivable value. The concentration should be assumed to be zero when the concentration of the free nitric acid in the nitrate solution is unknown or when a nonzero value cannot technically be assured.

Data for the changes in critical mass of an aqueous solution of plutonium nitrate as a function of the concentration of free nitric acid are included in the American ARH-600 Handbook.⁽⁴¹⁾ One example of these data is shown in Fig. 3.21. As is evident from this figure, the critical mass has

minimum values for the full range of plutonium concentration when the concentration of free nitric acid is zero (0 M).

3.3. Modeling of Reflector

3.3.1. General Modeling of Reflectors

Reflection conditions may be selected by considering realistic circumstances. In criticality safety evaluations, the following conditions are commonly used among various reflection conditions.

- (i) Full reflection by concrete: the case where a concrete wall with enough thickness is located near the object is considered.
- (ii) Full reflection by water: the case where submersion in water and other less reactive conditions are considered.
- (iii) Conditions without reflection: the case where mutual interaction is considered.
- (iv) Reflection by a water reflector 2.5 cm thick: the case where reflections from structural materials and human bodies near the object are considered.

In a criticality safety evaluation for a nuclear fuel installation, unrealistic models such as complete water submergence, etc., are often used for a conservative or “pessimistic” evaluation. Abnormal changes in the conditions for reflection result from submersion in water due to inundation or water spray from a fire extinguisher and careless introduction of reflecting materials near the fuel (wood, plastic, human bodies). However, the probability of the occurrence of such abnormalities can be reduced to a minimum by proper selection of site and layout of equipment, by chemical fire extinguishing materials instead of water, and by strict enforcement of operational controls. When adequate countermeasures are taken against the occurrence of the above abnormalities, the use of unrealistic models, such as complete water submersion, is not necessary. Instead, realistic conditions may be used.

3.3.2. Sufficient Reflector Thickness⁽⁴²⁾

Normally, criticality safety assessment of a single unit is done by choosing a model where the single unit is surrounded by a sufficient reflector. A water reflector thickness of about 20 cm is often used for sufficient reflector thickness. This value for a sufficient reflector thickness is derived below.

The ratio of effective multiplication factor that will change with an increase of reflector thickness, R , is represented as reflection factor (RF) as follows:

$$RF \equiv |(k(R) - k_s) / k_s| \quad (3.6)$$

where

k_s : Effective multiplication factor of a single unit enclosed in an infinitely thick reflector.

$k(R)$: Effective multiplication factor where the reflector thickness is R and a vacuum is outside the reflector.

RF can be represented by the following equation according to the one-group diffusion approximation.

$$RF(R) = Ae^{-2k_r R}$$

$$\text{where, } k_r^2 = B_{\perp}^2 + \frac{1}{M_r^2} \quad (3.7)$$

In an infinite slab system, since B_{\perp}^2 (which represents axial buckling) is 0, RF decreases exponentially with a gradient inversely proportional to the square root of the neutron migration length (M_r) of the reflector. If a sufficient reflector thickness is assumed where RF is less than $10^{-3}\Delta k/k$, from Eq. (3.6) and (3.7) the following equation is obtained:

$$\begin{aligned} RF(R) &= RF(0) \cdot e^{-2k_r R} \\ &= (1 - k(0)/k_s) e^{-2k_r R} \leq e^{-2k_r R} \leq 10^{-3} \end{aligned} \quad (3.8)$$

According to ANL.5800,⁽⁴³⁾ the neutron migration length in water is $M_r = 5.93$ cm. From Eq. (3.8), the following equation is obtained:

$$R \geq -2.97 \cdot \ln(10^{-3}) \cong 20 \text{ (cm)} \quad (3.9)$$

Consequently, for water, a thickness of 20 cm or more will provide a sufficient reflector thickness. A sufficient reflector thickness will be provided for general reflectors, if a thickness of $3.5 M_r$ is secured, where M_r represents neutron migration length in the reflector. Calculated values of RF, where the water reflector thickness is used as a parameter with respect to various fuel systems, are compared in Fig. 3.22.

3.3.3. Reflection Effects of a Concrete Wall

In the criticality safety assessment for a single unit, it is normally acceptable to use a model where the single unit is surrounded with a water reflector having a sufficient thickness. However, when modeling a concrete wall with an air gap between the concrete wall and the nuclear fuel container, a conservative result may not be obtained in this way. In this situation, criticality safety should be assessed by adopting a model where nuclear fuel + space + concrete are included.

An example in which the effective albedo is calculated using the diameter of a circular cylinder,

^{235}U enrichment, and uranium concentration as parameters is given below. The ratio of the inside length of the concrete vessel to the inside diameter of the circular cylinder (L/D) is taken as the abscissa for an infinite circular cylinder system of homogeneous $\text{U-H}_2\text{O}$ fuel surrounded by the concrete vessel.⁽⁴⁴⁾ Effective albedo refers to the value at which the same effective multiplication factor can be obtained as the case where a real system is analyzed as a result of a calculation that assumes this albedo on the surface of the fuel body. The effective albedo decreases with an increase in the L/D ratio. The L/D ratio at which the effective albedo becomes equal to the albedo resulting from a sufficient water reflector is shown in Fig. 3.23. This value varies with the kind of fuel body. CASE No. 1 is for the system in which the leakage of neutrons is very large and the neutron energy spectrum is hard. Very few examples are expected to exceed the reflection effect of CASE No. 1. If the L/D ratio is 2 or more and the safety margin is slightly larger than for the result of CASE No. 1, the critical conditions that assume a sufficient water reflector can be used.

3.3.4. Reflection Effect of Container Wall with Vacuum Exterior

When the container wall is surrounded by a vacuum in an assessment model, the container wall will act as a reflector and must be considered. When the container wall is SUS (stainless steel) and the wall is less than 6 cm thick, the assessment may be done with a model where a water reflector having sufficient thickness surrounds the exterior of fuel area instead of the container.

For example, the results⁽⁴⁵⁾ of a calculation of effective multiplication factor using uranium concentration and the container (SUS-304) wall thickness as parameters with respect to an infinite circular cylinder system with a vacuum exterior for homogeneous $\text{UO}_2\text{-H}_2\text{O}$ fuel are shown in Fig. 3.24. As is evident from this figure, when the container is surrounded by a vacuum, the container wall functions as a reflector. Thus, the reactivity of this system increases with an increase in the wall thickness. Comparison of the effective multiplication factor where the fuel area is enclosed with a water reflector having sufficient thickness shows that the water reflector having sufficient thickness has a larger effective multiplication factor, if the thickness of SUS-304 is less than 6 cm.

3.3.5. Reflection Effect of Container Wall with External Water Reflector

When the exterior of the container wall is exposed to water-reflecting conditions, the container wall may function as a reflector more effectively than water when the thickness exceeds a certain value. In such a case, criticality safety must not be assessed with a model where a water reflector only surrounds the exterior of fuel area and the container wall is disregarded.

The criticality data commonly described in the criticality handbooks are calculated with a model where a water reflector surrounds the fuel without taking the container wall into consideration. When the container wall is thin, the wall will function as a neutron absorber and an assessment with a model having the container wall replaced by water wall will give an accurate reactivity result. In this situation, the criticality data described in the handbook may be used as they are. On the other hand,

when the thickness of container wall exceeds a certain value, the wall may function as a more effective reflector than water. Consequently, if assessment is done with a model in which the container wall is disregarded and is replaced by water, the reactivity may be assessed lower than really exists. In this situation, the use of the data described in the handbook is inappropriate. The wall thickness limit value varies with fuel form, fuel composition, fuel concentration and container wall material. When criticality data are used in the criticality safety assessment or when the container wall is neglected in preparing an analytical model, the suitability of this approach should be fully studied.

At JAERI, the effective multiplication factor was calculated using wall thickness (SUS-304) and uranium concentration in fuel as parameters in an infinite circular-cylinder system and an infinite slab system of homogeneous $\text{UO}_2 - \text{H}_2\text{O}$ fuel with a 30 cm thick water reflector.⁽⁴⁵⁾ As is evident from the calculational results shown in Fig. 3.25 and 3.26, the container wall serves as an absorber up to a thickness of about 2 cm. When the thickness increases beyond that, the container wall starts acting as a reflector. Depending on conditions, when the container wall reaches a thickness 5 cm or more, reactivity becomes higher than when the container wall thickness is zero.

3.3.6. Reflection Effect of Container Wall where Exterior is Surrounded by Cd + H₂O

When the exterior of the fuel container wall is surrounded by Cd + H₂O, the container wall will function as a reflector. In preparing an analytical model for the criticality safety assessment, therefore, the container wall should be considered.

When storing fuel solutions in a nuclear fuel installation, a neutron absorber such as cadmium plate is often attached to the container surface to increase the storage capacity. In this situation, the container wall functions as a reflector. If water reflecting conditions for the exterior are modeled, the container wall should be considered in preparing the model.

A calculation example is shown in reference 46. In this example, a submersion state is modeled with a system having a cadmium plate 1 mm thick attached to the surface of the fuel container wall on the inside of an annular tank containing homogeneous Pu-H₂O fuel. As is evident from Fig. 3.27, the ring thickness at which the fuel becomes critical decreases with an increase in the thickness of the inner container wall of the annular tank. This shows that the container wall is serving as a reflector. This occurs because most of the neutrons that are thermalized in the water reflector are absorbed by the cadmium plate before they can return to the thermal area. Thus, the absorption effect of the container wall becomes less than its reflection effect.

3.3.7. Criticality Safety Evaluation without Assuming Flooding

When evaluating the criticality safety of a system where there is no chance of water submersion in contingencies such as floods and where fire extinguishers using water are prohibited, it is not necessary to assume the submersion in water. When evaluating the criticality safety of a single unit

system with no possibility of submersion in water, it is not necessary to assume a water reflector of sufficient thickness. However, the reflection effect of structural materials surrounding the unit must be considered. The design of structural materials surrounding a unit is often not determined at the basic design stage, or when the design is determined, it is often a very complicated one. To account for this, the neutron reflection effect by surrounding materials is approximated by a simple model, or an actual design is simplified to a simple model by ignoring branch pipes.

To account for the neutron reflection effect of surrounding materials and the reactivity corresponding to the reactivity worth of branch pipes, a water reflector having a 2.5-cm thickness is chosen. Since this neutron reflection effect of surrounding structural materials does not include that of surrounding walls, the reflection effect of surrounding walls must be considered separately.

When evaluating the criticality safety of multiple units where there is no chance of water submersion, it is not necessary to assume the presence of water between units, but it is necessary to confirm that the system does not become critical due to neutron interaction among multiple units. This confirmation of subcriticality is given by first calculating the effective multiplication factor of the multiple units ignoring branch pipes and surrounding structural materials except walls and then by checking that subcriticality is achieved even after adding an appropriate amount of reactivity taking account of the reflection from structural materials and the reactivity of branch pipes.

Reference (47) confirms that using a water reflector of 2.5 cm thickness in place of surrounding structural materials is a conservative assumption. The results of criticality calculations assuming models similar to the actual systems are summarized in the following quote. "It was revealed that in both cases of single unit and multiple units the neutron reflection effect of surrounding structural materials is smaller than that of a water reflector of 2.5 cm thickness, except for a special case where a cadmium plate was installed at the bottom of unit."

3.4. Modeling of Absorber

Both soluble (solution) and insoluble (solid) neutron absorbers are used in criticality control. For a soluble material, a calculation model is prepared on the assumption that absorber and nuclear fuel are homogeneously mixed. For criticality control, it is important that this mixture remains homogeneous at all times. For analysis, assuming a pessimistic concentration of absorber may be sufficient. When considering a solid neutron absorber, assuming the mixture as homogeneous will be dangerous because the absorption effect will be overestimated. For a solid neutron absorber, therefore, a calculation model should be prepared that closely models the real shape and arrangement of the neutron absorber. In particular, great care should be taken in modeling Raschig rings used as neutron absorbers. The cautions for modeling Raschig rings in a calculation model are described below.

A Raschig ring is a solid neutron poison often loaded in a fuel solution storage tank at a reprocessing installation to ensure subcriticality. Of the several calculation models conceivable for assessment of criticality safety of a system like this, the simplest model is a homogeneously mixed model. For this model, the neutron absorption effect of Raschig rings may be over-estimated and

therefore the effective multiplication factor of the system may be under-estimated compared with the actual system.

Fig. 3.28 shows the analysis of experimental data of plutonium nitrate solution systems containing Raschig rings.⁽⁴⁸⁾ The following 4 models are used for this analysis.

H : Homogeneous model

V : Vertical-tube model

M : Mutually perpendicular ring model

R : Random geometry model

An examination of this diagram shows that where the plutonium concentration is high, the calculated values of effective multiplication factor are over-estimated with all models. It also shows that with the H (homogeneous) model, the effective multiplication factor is remarkably under-estimated compared with other models when the plutonium concentration lowers.

As described above, in a system where Raschig rings are used as a neutron poison, great care should be taken when assessment is made with a model where fuel and poison are homogenized. It is desirable to consider heterogeneity in the assessment.

3.5. Calculation of Atomic Number Density

To make a criticality safety assessment for an instrument or for equipment, it is necessary to obtain the atomic number density of the nuclide contained in the nuclear fuel material and the structural material in each region that is modeled. To provide an accurate analysis, the values of atomic number density must be obtained as accurately as possible. However, there is usually a lack of data or limitation due to other reasons, so assumptions using theories or calculations need to be made. In this section, for the convenience of analyses, examples of theoretical formulas and data frequently used are described.

3.5.1. Theoretical Density of Fuel Compounds and Density Formula of Fuel Aqueous Solutions

Table 3.4 lists theoretical densities of elemental uranium and plutonium and compounds of these elements frequently used for assessment of criticality safety of nuclear fuel handling installations. The value of ρ_{UO_2} in the table is used for high content ^{238}U uranium with a ^{235}U enrichment less than about 3 wt%. Theoretical density values of UO_2 having different enrichments are shown in Table 3.5. The value of ρ_{UO_2} in Table 3.4 is used for PuO_2 having a high enrichment of ^{239}Pu (higher than about 90 wt%). Calculation examples of theoretical density values of PuO_2 having different enrichments of ^{239}Pu are shown in Table 3.6. The theoretical density of UO_2 - PuO_2 mixed oxide is calculated by the following formula.

$$\rho = \frac{4M}{(a_0)^3 \cdot N_A} \quad (3.10)$$

where

ρ : Theoretical density [g/cm³]

M: Molecular weight

A: Avogadro's number

a_0 : Lattice constant of UO₂-PuO₂

The lattice constant (a_0) of UO₂-PuO₂ is obtained from the following equation,

$$a_0 = 5.4700 - 7.40 \times 10^{-4} \cdot \varepsilon_{\text{PuO}_2} \quad (3.11)$$

where $\varepsilon_{\text{PuO}_2}$ represents the ratio of PuO₂ in mixed oxide in percentage of atomic ratio, that is, it represents PuO₂/(UO₂ + PuO₂).

Calculational equations for atomic number density in solution systems are shown below.

- (1) Moeken's equation⁽⁵³⁾ described below is used for calculating the density of an aqueous solution with uranyl nitrate. Based on the density at 25°C as the standard, the density ρ_t [g/cm³] at temperature t °C can be represented as follows.

$$\begin{aligned} \rho_t &= 1.0125 \times \rho_{25} + 0.000145 \times t - 0.0005 \times \rho_{25} \times t - 0.0036 \\ \rho_{25} &= 1.0171 + 1.2944 \times 10^{-3} \times C_U + 0.0289 \times C_{\text{HN}} \end{aligned} \quad (3.12)$$

- (2) Maimoni's equation⁽³⁶⁾ is used for calculating the density of an aqueous solution with plutonium nitrate. Temperature T [°C] is obtained by converting solution temperature t [°C] according to the relationship $T = t - 25$. Density ρ_t is represented by the following equation.

$$\begin{aligned} \rho_t &= 0.99708 + 1.65625 \times 10^{-3} \times C_{\text{Pu}} + 3.2959 \times 10^{-2} \times C_{\text{HN}} \\ &\quad - 5.9915 \times 10^{-4} \times T - 4.8706 \times 10^{-5} \times C_{\text{Pu}} \times C_{\text{HN}} \\ &\quad - 1.4217 \times 10^{-6} \times C_{\text{Pu}} \times T - 3.418 \times 10^{-8} \times C_{\text{Pu}}^2 \end{aligned} \quad (3.13)$$

- (3) The following density equation derived from Eqs. (3.12) and (3.13) is used for calculating the density of aqueous solution with uranyl nitrate and plutonium nitrate.

$$\begin{aligned} \rho_{\text{U+Pu}} &= R_U \times \rho_{\text{U(U+Pu)}} + R_{\text{Pu}} \times \rho_{\text{Pu(U+Pu)}} \\ &\quad - R_U \times C_{\text{Pu}} (1.50 \times 10^{-3} \times C_N^{-1} - 3.05 \times 10^{-4}) \end{aligned} \quad (3.14)$$

where,

$$R_U = \frac{C_U}{C_U + C_{\text{Pu}}}, R_{\text{Pu}} = \frac{C_{\text{Pu}}}{C_{\text{Pu}} + C_U}, C_N = 4 \times C_{\text{PN}} + 2 \times C_{\text{UN}} + C_{\text{HN}}$$

In Eq. (3.14), $\rho_{U(U+Pu)}$ represents the density calculated with Eq. (3.12) where all uranium and plutonium in the solution are regarded as uranium. Similarly, $\rho_{Pu(U+Pu)}$ is the density obtained with Eq. (3.13) where all uranium and plutonium are regarded as plutonium.

When the volume of the solution is 1 L, the relation between density, ρ , and the weights of solutes is represented by the following equation.

$$1000\rho = C_U + C_{Pu} + C_{HN} \times M_H + (2 \times C_U \times M_U^{-1} + 4 \times C_{Pu} \times M_{Pu}^{-1} + C_{HN}) \times M_{NO3} + 2 \times C_U \times M_U^{-1} \times M_O + C_{H2O} \times M_{H2O} \quad (3.15)$$

where, concentration C_t [g/L] in Equation (3.15) is at t [°C]. The relation between reactivity and concentration at 25°C is given by the equation below.

$$C_t / C_{25} = \rho_t / \rho_{25} \quad (3.16)$$

where,

- C_t : concentration of a component [g/L] at t [°C],
- C_{25} : concentration of a component [g/L] at 25 °C,
- ρ_t : density [g/cm³] at t [°C] calculated from Equation (3.17), and
- ρ_{25} : density [g/cm³] at 25 °C calculated from Equation (3.17).

The symbols used in Eqs. (3.12) through (3.15) are defined as follows:

- C_U : Concentration of uranium [g/L],
- C_{UN} : Molarity of uranium [mol/L],
- C_{Pu} : Concentration of plutonium [g/L],
- C_{PN} : Molarity of plutonium [mol/L],
- C_{HN} : Molarity of nitric acid [mol/L],
- M_O : Atomic weight of oxygen [g/mol],
- C_{H2O} : Molarity of water [mol/L],
- M_U : Atomic weight of uranium [g/mol],
- M_H : Atomic weight of hydrogen [g/mol],
- M_{Pu} : Atomic weight of plutonium [g/mol],
- M_{NO3} : Molecular weight of nitrate ion [g/mol], and
- M_{H2O} : Molecular weight of water [g/mol].

C_U , C_{Pu} , and C_{HN} used in Eqs. (3.12), (3.13), and (3.14) represent the concentrations at 25 °C.

The following equation⁽⁵⁵⁾ can be adopted for aqueous uranium (VI)-nitrate solution, aqueous plutonium (IV)-nitrate solution, and aqueous uranium (VI)-plutonium (IV)-nitrate solution:

$$\rho = 0.99833 + 1.6903 \times 10^{-3} \times C_{Pu25} + 1.4276 \times 10^{-3} \times C_{U25}$$

$$\begin{aligned}
& +3.9956 \times 10^{-2} \times C_{\text{HN25}} \\
& -8.696 \times 10^{-8} \times C_{\text{Pu25}}^2 \\
& -1.087 \times 10^{-7} \times C_{\text{U25}}^2 \\
& -8.513 \times 10^{-4} \times C_{\text{HN25}}^2 \\
& -5.442 \times 10^{-6} \times T^2 \\
& -4.4889 \times 10^{-5} \times C_{\text{Pu25}} \times C_{\text{NH25}} \\
& -1.310 \times 10^{-6} \times C_{\text{Pu25}} \times T \\
& -1.564 \times 10^{-5} \times C_{\text{U25}} \times C_{\text{HN25}} \\
& -9.487 \times 10^{-7} \times C_{\text{U25}} \times T \\
& -8.684 \times 10^{-5} \times C_{\text{HN25}} \times T
\end{aligned} \tag{3.17}$$

where

ρ : solution density [g/cm³] at T[°C],
 C_{Pu25} : plutonium concentration at 25 °C[g/L],
 C_{U25} : uranium concentration at 25 °C[g/L],
 C_{HN25} : nitrate solution at 25 °C[mol/L] and
 T : temperature[°C]

Equation (3.17) is applicable to $C_{\text{U25}} < 530$ g/L, $C_{\text{Pu25}} < 480$ g/L, $C_{\text{Pu25}} + C_{\text{U25}} < 350$ g/L, $C_{\text{HN25}} < 7$ mol/L, and a temperature range between 10 and 60°C (standard deviation: 0.0032 g/cm³).

- (4) The density of an aqueous solution of uranyl fluoride is calculated from the following equation.⁽⁵⁶⁾

$$\frac{1}{\rho} = \frac{1}{\rho_0} - 0.9120F + 0.0567F^2 \tag{3.18}$$

where

ρ : Solution density [g/cm³],
 ρ_0 : Water density [g/cm³], and
 F : Weight ratio of UO₂F₂ in solution [-].

3.5.2. Changes in Nuclide Composition Resulting from Burnup

Fresh fuel contains more fissile material than burned fuel, but does not include fission products (FP_s) which have a neutron absorption effect. Therefore, when the presence of burnable poisons such as gadolinium is neglected, fresh fuel has higher reactivity. Consequently, in a criticality safety assessment model, if burned fuel is replaced by fresh fuel, a severer (conservative) result in reactivity can be obtained. However, when the evaluation of burnup is valid from the criticality safety evaluation viewpoint, the criticality safety evaluation may be conducted taking account of changes in nuclide composition resulting from burnup.

3.5.2.1. Composition of Uranium and Plutonium Nuclides

Table 3.7 quotes a set of data from "the Nuclear Criticality Safety Handbook - Data Collection," which shows assembly averaged composition changes of uranium and plutonium nuclides that can be used in a criticality safety evaluation model of fuel depleted in a pressurized light water reactor (PWR). Fig. 3.29 shows the same data graphically. Intermediate values between one burnup level and another in Table 3.7 were linearly interpolated.⁽⁵⁷⁾ Fig. 3.30 graphically shows the equation of Pu/U given in the footnote of Table 3.7.

The data in Table 3.7 and the Pu/U equation in the footnote were compared with the actual measurement data of many PWR and BWR burnup fuels (Fig. 3.31 to 3.36).⁽⁵⁷⁾ The trend of compositional changes shown in these graphs is considered to be applicable to BWR fuel assemblies as well as PWR fuel assemblies having similar specifications to those assemblies measured in Fig. 3.31 to 3.36, in an initial ^{235}U enrichment range of 2wt% to 4wt%, and a burnup range of 0 to 50 GWd/t. However, the burnup distribution effect on reactivity such as the fuel end effect in the axial direction and should be considered separately.

3.5.2.2. FP Nuclides that can be Considered in Criticality Safety Evaluation

When evaluating the criticality safety of a storage, transportation, dissolution, or clarification process for spent fuel from a light water reactor, the presence of FP nuclides shown in Table 3.8 may be considered in the evaluation.⁽⁵⁷⁾ The amount of neutron absorption due to FP nuclides in spent fuel pulled from a PWR was calculated using the ORIGEN2 Code⁽⁵⁹⁾ and listed in descending order of neutron absorption in Table 3.9.^{(57),(60)} However, it is not desirable to consider all the nuclides in the table in a criticality safety evaluation from the viewpoint of the calculation accuracy of FP formation and the complication of the criticality analysis. Ten or so nuclides were selected for a criticality safety evaluation. The FP nuclides in the following categories were excluded from the evaluation:

(1) Nuclide with small neutron absorption

FP nuclides whose contribution to the total neutron absorption is small are excluded. As a guideline, FP nuclides to be excluded are selected in such a manner that the sum of absorption of selected nuclides amounts to no more than 30% of the total neutron absorption of all FP nuclides.

(2) Nuclides with short half-lives

FP nuclides that reduce their masses significantly by decaying after a spent fuel is pulled out of a reactor (FP nuclides having half-lives of less than 30,000 years) are excluded. Those nuclides with half-lives of more than 30,000 years must be considered in the evaluation, since more than 99.9 % of those nuclides present when the fuel is pulled out of a reactor are still present 40 years later, and the reactivity increase due to the decay is negligibly small. For spent fuels that have been cooled outside a reactor for several years,

this half-life criterion is applicable to only Pm-147 (half-life of 2.6 years) among the top ten nuclides of neutron absorption. The impact of ignoring the presence of Pm-147, however, is small.

(3) Gaseous or volatile nuclides

Gaseous FP nuclides such as Kr and Xe, and volatile FP nuclides such as Br and I are excluded. These nuclides (elements) are released from fuel pellets of a fuel rod and are distributed in the gas plenum (or released to outside if the fuel rod is damaged), and it is not likely that these nuclides stay in the fuel.

(4) Semi volatile nuclides

Nuclides such as Rb, Te, Se, Cd, Sb, and Ag from semi volatile elements or semi volatile compounds. These nuclides (elements) are excluded because although they are deposited on the inside surface of cladding, they may be released from the inside surface due to the impact given by fuel handling or other reasons.

The FP nuclides mentioned in (1) through (4) above are excluded when performing a criticality safety evaluation of a transportation and storage of spent fuel. When performing a criticality safety evaluation of a dissolution, clarification, and their down stream processes, the following additional FP nuclides should be excluded from the evaluation.

(5) Nuclide remained as undissolved residues (hulls)

Mo, Tc, Ru, Rh, and Pd are FP nuclides that may be constituents of undissolved residues (hulls) in the dissolution process. These nuclides (elements) have been found in undissolved residues in the form of hexagonal Mo-(Tc+Ru)-(Rh+Pd) following the dissolution of spent fuel with nitric acid solution. These nuclides are excluded from the criticality safety evaluation because they may possibly be separated from fuel dissolving solution in the dissolution process.

As the result of excluding these FP nuclides above, remaining FP nuclides that can be used for the criticality safety evaluation are denoted by a symbol "o" in Table 3.8. Those nuclides denoted by a symbol "x" are ones that can be included in the criticality safety evaluation of transportation and storage of spent fuel but should not be used for that of the dissolution, clarification, and their down stream processes at a reprocessing plant. Fig. 3.37 shows the amount of contribution (%) of each nuclide to the total neutron absorption of all FP nuclides, using the cooling time as a parameter. The data in the table are applicable to PWR fuel assemblies arrayed in water.

3.5.3. Nuclide Reactivity Worth⁽⁶¹⁾

There are many varieties of nuclides in spent fuel. However, a model in which the fuel

composition is replaced by that of simple nuclide composition needs to be adopted if analyses are to be simplified and if the criticality data that are already calculated are to be used. In this case, the fuel composition should be replaced by one having a high reactivity value.

When a neutron is absorbed into a fissile nuclide and fission takes place at a probability of σ_f/σ_a , a quantity ν of neutrons are generated. Here, σ_f represents the fission cross section and σ_a represents the absorption cross section. Consequently, when one neutron is absorbed into a fissile nuclide, on the average $\nu \cdot \sigma_f/\sigma_a$ neutrons are produced. This value is generally called the regeneration factor and is represented by η . Since this value varies greatly with neutron energy, an average η for a certain nuclide, i , is defined by the following equation. This value is used as an approximation for the degree of contribution of nuclide i towards the reactivity of the system.

$$\eta_i = \frac{\int_{\text{all energy}} \nu_i(E) \cdot \sigma_{fi}(E) \cdot \phi(E) dE}{\int_{\text{all energy}} \sigma_{ai}(E) \cdot \phi(E) dE} \quad (3.19)$$

Values of η for each of the nuclides calculated by the JACS Code System are shown in Fig. 3.38 along with the order of their sizes. These values were calculated against varied H/ ^{235}U atomic ratios in a homogeneous (^{235}U - H_2O) system to represent their dependency on the neutron energy spectra. The effective cross sections were calculated using background cross section, σ_0 , as a parameter. When nuclide j in the assessment system is to be replaced by another nuclide i having a higher reactivity, the following equation must be satisfied.

$$\frac{N_j}{N_i} \leq \frac{\eta_i}{\eta_j} \cdot \frac{\bar{\sigma}_{ai}}{\bar{\sigma}_{aj}} \cdot \left\{ 1 - \left(\frac{\eta_j}{\eta_i} - 1 \right) \cdot \frac{\bar{\sigma}_{aj}}{\sigma_{a0}} \right\} \quad (3.20)$$

where

- N_j : Atomic number density of nuclide j ,
- N_i : Atomic number density of nuclide i ,
- $\bar{\sigma}_{aj}$: Microscopic absorption cross section of nuclide j averaged with space and energy,
- $\bar{\sigma}_{ai}$: Microscopic absorption cross section of nuclide i averaged with space and energy, and
- σ_{a0} : Cross section of neutron elimination from the system, per one nuclide j to which attention is paid. It is represented by the following equation.

$$\sigma_{a0} = \frac{1}{N_j} \left\{ \sum_{i \neq j} N_i \bar{\sigma}_{ai} + L / (\bar{\phi} \cdot V) \right\} \quad (3.21)$$

where

- L : Amount of leakage of neutrons from the system,
- $\bar{\phi}$: Average neutron flux, and
- V : Volume.

Equation (3.20) is a function of the value of η shown in Fig. 3.38 and also that of σ_{ai} and σ_{aj} . The average microscopic absorption cross section can be obtained from the following equation.

$$\sigma_a = g\sigma_{22}\sqrt{\frac{\pi}{4} \cdot \frac{T_0}{T}} \cdot \frac{\phi_{th}}{\phi_{tot}} + RI \frac{\phi_{epi}}{\phi_{tot}} + \sigma_{fs} \cdot \frac{\phi_{fast}}{\phi_{tot}} \quad (3.22)$$

where

- g : Ratio of reactivity by cross section extrapolated according to the $1/v$ law from the value of σ_{22} to the reactivity by true cross section,
- σ_{22} : Absorption cross section to neutron of 2200 m/sec,
- T_0 : 293.15K ,
- T : Moderator temperature [K],
- RI : Effective resonance integral,
- σ_{fs} : Average cross section where fission spectrum is used as weight,
- ϕ_{th} : Neutron flux of thermal group (< 0.5 eV) ,
- ϕ_{epi} : Neutron flux of epithermal group,
- ϕ_{fast} : Neutron flux of fast group , and
- ϕ_{tot} : $\phi_{th} + \phi_{epi} + \phi_{fast}$.

Equation (3.20) was used to show qualitatively how many ^{235}U atoms become equivalent in reactivity with one object nuclide. The result is shown in Fig. 3.39. The calculation assumed absorption cross sections to 2200 m/sec absorption as σ_{ai} and σ_{aj} . For η_i and η_j , the values corresponding to the cases of $H/^{235}\text{U} = 10^3$ and $\sigma_0 = 10^2$ were used.

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Table 3.1 The comparison and increase rate of the neutron multiplication factors under the optimum vs. uniform distribution of aqueous uranyl nitrate solution (^{235}U enrichment: 5 wt%, nitric acid molarity : 0) in a 18 cm thick slab with a full water reflector (A constraint condition was set so as to allow only one minimum point in a symmetrical fuel concentration distribution relative to the fuel center.)

| Mean concentration [gU/L] | Neutron multiplication factor $k_{\text{eff}}[-]$ | | Increase rate of neutron multiplication factor [% $\Delta k/k$] | Optimum distribution |
|------------------------------|--|----------------------|---|----------------------|
| | Uniform distribution | Optimum distribution | | |
| 300 | 0.6799 | 0.6826 | 0.40 | --- |
| 500 | 0.8156 | 0.8190 | 0.42 | --- |
| 700 | 0.8803 | 0.8856 | 0.60 | --- |
| 900 | 0.9092 | 0.9186 | 1.03 | Fig 3.1 |
| 1,100 | 0.9169 | 0.9255 | 0.94 | --- |

Table 3.2 The comparison and increase rate of the neutron multiplication factors under the optimum vs. uniform distribution of aqueous plutonium nitrate solution (^{239}Pu : ^{240}Pu = 5:1, nitric acid molarity: 0) in a 8 cm thick slab with a full water reflector (A constraint condition was set so as to allow only one minimum point in a symmetrical fuel concentration distribution relative to the fuel center.)

| Mean concentration [gPu/L] | Neutron multiplication factor $k_{\text{eff}}[-]$ | | Increase rate of neutron multiplication factor [% $\Delta k/k$] | Optimum distribution |
|-------------------------------|--|----------------------|---|----------------------|
| | Uniform distribution | Optimum distribution | | |
| 70 | 0.8662 | 0.8961 | 3.45 | --- |
| 100 | 0.8925 | 0.9242 | 3.55 | --- |
| 200 | 0.9089 | 0.9443 | 3.89 | --- |
| 300 | 0.9044 | 0.9408 | 4.02 | Fig 3.2 |
| 500 | 0.8923 | 0.9204 | 3.15 | --- |

Table 3.3 Example calculations of reactivity effect of branch pipes based on the simplified equation (Equation 3.5 of the main text)

| Thickness of a branch pipe [cm] | k_i | $\left(\frac{L}{L+A} \right)_i$ | ω_{0i} | ω_{i0} | ω_i |
|---------------------------------|---------|----------------------------------|-----------------------|---------------|-----------------------|
| 2 | 0.00379 | 0.997 | 1.24×10^{-3} | 0.5 | 2.38×10^{-4} |
| 5 | 0.0164 | 0.984 | 7.53×10^{-3} | 0.5 | 1.45×10^{-3} |
| 10 | 0.121 | 0.897 | 2.76×10^{-2} | 0.5 | 5.59×10^{-3} |
| 15 | 0.299 | 0.758 | 5.45×10^{-2} | 0.5 | 1.27×10^{-2} |

Table 3.4 Theoretical densities of uranium and plutonium compounds (Units: g/cm³)

| | |
|---|------------------------|
| ρ_{UO_2} | 10.96 ⁽⁴⁹⁾ |
| ρ_{UO_3} | 7.29 ⁽⁴⁹⁾ |
| ρ_{U} | 19.05 ⁽⁴⁹⁾ |
| ρ_{UF_6} | 5.09 ⁽⁴⁹⁾ |
| $\rho_{\text{U}_3\text{O}_8}$ | 8.5 ⁽⁵⁰⁾ |
| ρ_{ADU}^{*1} | 4.83 ⁽⁵¹⁾ |
| ρ_{Pu} | 19.816 ⁽⁴⁹⁾ |
| ρ_{PuO_2} | 11.46 ⁽⁴⁹⁾ |
| $\rho_{\text{H}_2\text{O}}^{*2} (20^\circ\text{C})$ | 0.9982 ⁽⁵²⁾ |

*1 $3\text{UO}_3 \cdot \text{NH}_3 \cdot 5\text{H}_2\text{O}$ (Amonium Diuranate)

*2 Water density (for information)

Table 3.5 **Densities of UO_2 where ^{235}U enrichment varies**

| ^{235}U enrichment (wt%) | UO_2 density (g/cm^3) |
|--------------------------------------|--|
| Natural | 10.96 |
| 20 | 10.93 |
| 40 | 10.91 |
| 60 | 10.89 |
| 80 | 10.86 |
| 100 | 10.84 |

Table 3.6 **Densities of PuO_2 where $(^{239}\text{Pu}+^{241}\text{Pu})/\text{Pu}$ is used as a parameter**

| $(^{239}\text{Pu}+^{241}\text{Pu})/\text{Pu}$ [wt%] | 93 | 90 | 80 | 75 | 50 |
|---|-------|--------|-------|-------|--------|
| PuO_2 density [g/cm^3] | 11.46 | 11.46 | 11.47 | 11.48 | 11.49 |
| (Pu isotopic composition, wt%) | | | | | |
| ^{238}Pu | 0.0 | 0.02 | 0.09 | 0.494 | 0.101 |
| ^{239}Pu | 93.0 | 90.484 | 77.68 | 68.18 | 38.408 |
| ^{240}Pu | 7.0 | 8.573 | 18.50 | 22.07 | 45.624 |
| ^{241}Pu | 0.0 | 0.853 | 3.22 | 7.268 | 10.702 |
| ^{242}Pu | 0.0 | 0.07 | 0.51 | 2.037 | 5.165 |

Table 3.7 Example of evaluation model for (assembly-averaged) composition changes in uranium and plutonium nuclides resulting from burnup^{*1}

| Burnup [GWd/t] | $\frac{{}^{235}\text{U}}{({}^{235}\text{U})_{\text{initial}}}$ | $\frac{{}^{239}\text{Pu}}{\text{Pu}}$ | $\frac{{}^{240}\text{Pu}}{\text{Pu}}$ | $\frac{{}^{241}\text{Pu}}{\text{Pu}}$ | $\frac{{}^{242}\text{Pu}}{\text{Pu}}$ |
|-------------------|--|---------------------------------------|---------------------------------------|---------------------------------------|---------------------------------------|
| 0 | 1 | 1 | 0 | 0 | 0 |
| 5 | 0.88 | 0.95 | 0.05 | 0 | 0 |
| 15 | 0.70 | 0.80 | 0.15 | 0.05 | 0 |
| 30 | 0.40 | 0.60 | 0.25 | 0.15 | 0 |
| 50 | 0.20 | 0.50 | 0.25 | 0.15 | 0.10 |

*1 "Nuclear Criticality Safety Handbook -Data Collection-" shows approximation equation for Pu/U as follows:

$$\frac{\text{Pu}}{\text{U}} \times 100 = \frac{aE}{b+E},$$

where E is burnup [GWd/t] and the coefficients a and b are given in the table shown right.

A value of ${}^{238}\text{U}/\text{U}$ at each burnup, which is not shown in Table 3.7, is derived by considering that a nuclear fission produces energy of 200 MeV and the number of (U+Pu) decreases accordingly.

| Enrichment [wt%] | a | b |
|---------------------|-------|-------|
| 2.0 | 1.711 | 22.69 |
| 3.0 | 1.975 | 36.85 |
| 3.5 | 2.119 | 45.49 |
| 4.0 | 2.273 | 55.47 |

Table 3.8 FP nuclides that can be included in criticality calculations

| Nuclide | Storage and transportation | Dissolution and clarification | Remarks |
|---------|----------------------------|-------------------------------|--|
| Sm-149 | ○ | ○ | |
| Rh-103 | ○ | × | One of the main undissolved residue elements in dissolved fuel solution |
| Nd-143 | ○ | ○ | |
| Cs-133 | ○ | ○ | |
| Tc-99 | ○ | × | One of the main undissolved residue elements in dissolved fuel solution |
| Sm-152 | ○ | ○ | |
| Gd-155 | ○ | ○ | A half-life of the parent nuclide Eu-155 is short (4.68 years)* ¹ |
| Nd-145 | ○ | ○ | |
| Sm-147 | ○ | ○ | A half-life of the parent nuclide Pm-155 is short (2.62 years)* ¹ |
| Mo-95 | ○ | × | One of the main undissolved residue elements in dissolved fuel solution |
| Eu-153 | ○ | ○ | |
| Sm-150 | ○ | ○ | |
| Total | 12 | 9 | |

*1 When the shortest cooling time can be specified, the amount of nuclides right after the cooling time can be used. If not, use the amount of nuclides right after being pulled from the reactor.

Table 3.9 Top 30 nuclides in neutron absorption contribution of FP_s in a PWR spent fuel (30 GWd/t)

(Unit: %)

| Order | Pulled out of reactor | | 0.5 years | | 4 years | | 10 years | | 30 years | |
|-------|-----------------------|-------|-----------|-------|---------|-------|----------|-------|----------|-------|
| 1 | Xe135 | 18.78 | Sm149 | 12.94 | Sm149 | 12.96 | Sm149 | 12.80 | Sm149 | 12.58 |
| 2 | Rh103 | 9.02 | Rh103 | 12.09 | Rh103 | 12.16 | Rh103 | 12.01 | Rh103 | 11.80 |
| 3 | Nd143 | 8.53 | Nd143 | 10.64 | Nd143 | 10.66 | Nd143 | 10.53 | Gd155 | 11.60 |
| 4 | Sm149 | 7.02 | Cs133 | 7.17 | Cs133 | 7.18 | Gd155 | 9.04 | Nd143 | 10.34 |
| 5 | Pm149 | 6.13 | Xe131 | 7.07 | Xe131 | 7.08 | Cs133 | 7.09 | Cs133 | 6.97 |
| 6 | Cs133 | 5.90 | Pm147 | 6.81 | Gd155 | 5.25 | Xe131 | 6.99 | Xe131 | 6.87 |
| 7 | Xe131 | 5.77 | Tc99 | 5.17 | Tc99 | 5.18 | Tc99 | 5.11 | Tc99 | 5.02 |
| 8 | Tc99 | 4.28 | Sm151 | 4.71 | Sm151 | 4.59 | Sm152 | 4.49 | Sm152 | 4.42 |
| 9 | Sm151 | 3.87 | Sm152 | 4.54 | Sm152 | 4.55 | Sm151 | 4.33 | Sm151 | 3.65 |
| 10 | Sm152 | 3.78 | Eu153 | 3.01 | Eu153 | 3.02 | Eu153 | 2.98 | Eu153 | 2.93 |
| 11 | Nd145 | 2.49 | Nd145 | 2.99 | Nd145 | 3.00 | Nd145 | 2.96 | Nd145 | 2.91 |
| 12 | Eu153 | 2.48 | Mo99 | 2.31 | Pm147 | 2.70 | Sm147 | 2.44 | Sm147 | 2.53 |
| 13 | Sm150 | 1.70 | Sm150 | 2.04 | Mo95 | 2.40 | Mo95 | 2.37 | Mo95 | 2.32 |
| 14 | Pm148m | 1.65 | Ag109 | 1.63 | Sm150 | 2.04 | Sm150 | 2.02 | Sm150 | 1.98 |
| 15 | Mo95 | 1.64 | Eu154 | 1.58 | Sm147 | 1.93 | Ag109 | 1.61 | Ag109 | 1.58 |
| 16 | Eu155 | 1.39 | Eu155 | 1.56 | Ag109 | 1.63 | Ru101 | 1.54 | Ru101 | 1.51 |
| 17 | Eu154 | 1.37 | Ru101 | 1.55 | Ru101 | 1.56 | Pr141 | 0.95 | Eu151 | 1.39 |
| 18 | Ag109 | 1.35 | Pr141 | 0.96 | Eu154 | 1.20 | Pd105 | 0.79 | Pr141 | 0.94 |
| 19 | Ru101 | 1.29 | Cs134 | 0.93 | Pr141 | 0.97 | Eu154 | 0.73 | Pd105 | 0.77 |
| 20 | Cs134 | 0.92 | Gd155 | 0.90 | Eu155 | 0.95 | Kr83 | 0.73 | Kr83 | 0.71 |
| 21 | Rh105 | 0.87 | Sm147 | 0.89 | Pd105 | 0.80 | La139 | 0.58 | La139 | 0.57 |
| 22 | Pm147 | 0.84 | Pd105 | 0.80 | Kr83 | 0.74 | Zr93 | 0.57 | Zr93 | 0.56 |
| 23 | Pr141 | 0.76 | Kr83 | 0.73 | La139 | 0.59 | Pm147 | 0.55 | Pd108 | 0.52 |
| 24 | Pd105 | 0.66 | La139 | 0.58 | Zr93 | 0.58 | Pd108 | 0.53 | Cs135 | 0.42 |
| 25 | Kr83 | 0.61 | Zr93 | 0.58 | Pd108 | 0.54 | Eu151 | 0.51 | I129 | 0.42 |
| 26 | Sm147 | 0.54 | Pd108 | 0.54 | Cs135 | 0.43 | Cs135 | 0.43 | Gd157 | 0.41 |
| 27 | La139 | 0.49 | Cs135 | 0.43 | I129 | 0.43 | I129 | 0.43 | Cd113 | 0.38 |
| 28 | Zr93 | 0.48 | I129 | 0.43 | Gd157 | 0.43 | Gd157 | 0.42 | Mo97 | 0.38 |
| 29 | Pd108 | 0.45 | Gd157 | 0.43 | Cd113 | 0.40 | Eu155 | 0.41 | Pd107 | 0.30 |
| 30 | Cs135 | 0.36 | Cd113 | 0.40 | Mo97 | 0.39 | Cd113 | 0.39 | Nd144 | 0.26 |

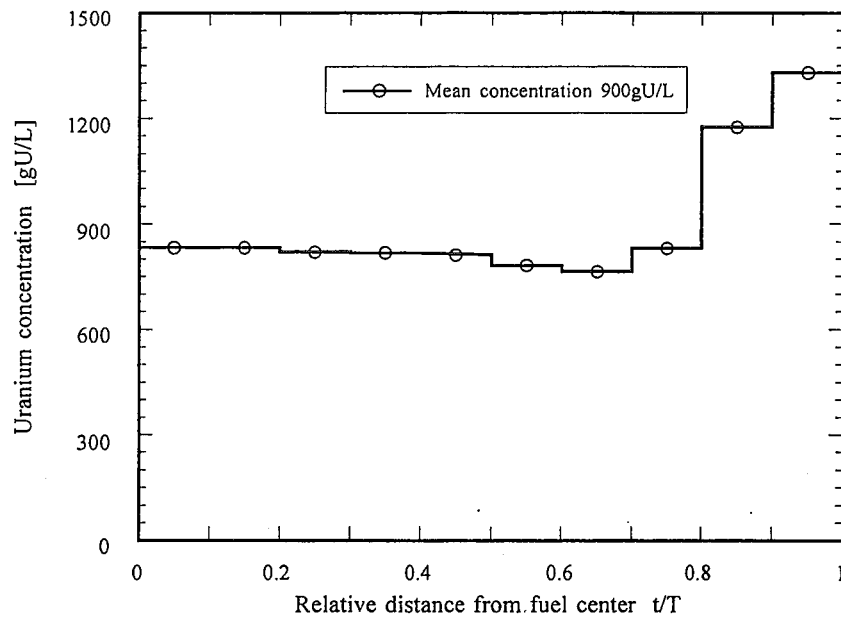


Fig 3.1 The optimum distribution of aqueous uranyl nitrate solution in a slab with a full water reflector
 (^{235}U enrichment : 5wt%, mean uranium concentration : 900gU/L, nitric acid molarity:0)
 (Only one minimum point in a symmetrical concentration distribution relative to the fuel center)

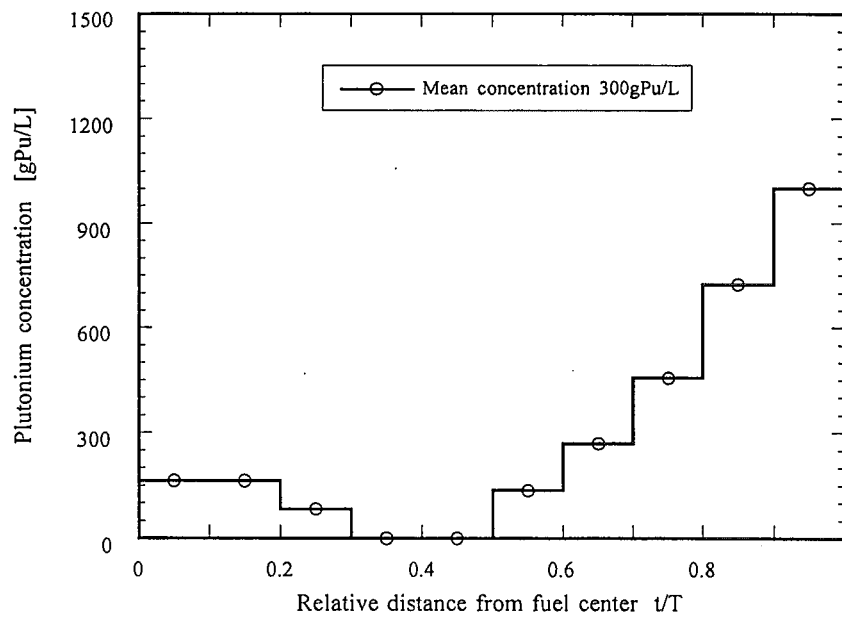


Fig 3.2 The optimum distribution of plutonium nitrate solution in a slab with a full water reflector
 ($^{239}\text{Pu} : ^{240}\text{Pu} = 5:1$, mean minimum concentration:300gU/L, nitric acid molarity:0)
 (Only one minimum point in a symmetrical fuel concentration distribution relative to the fuel center.)

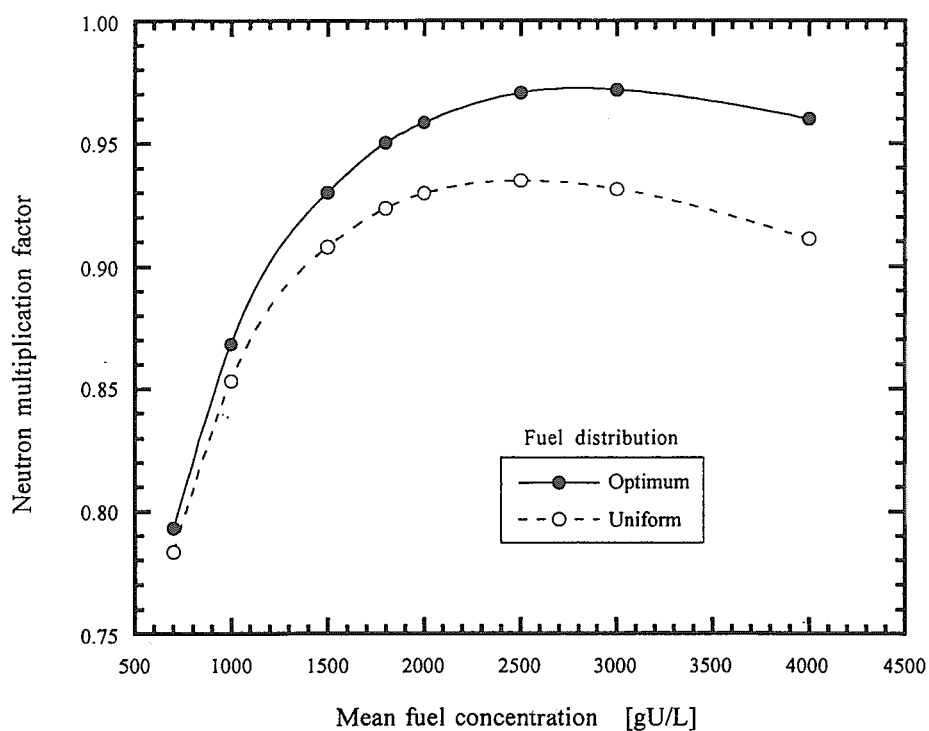


Fig 3.3 The neutron multiplication factors under optimum and uniform distributions (1)
(a 10-cm thick slab of homogeneous 5wt% ^{235}U -enriched $\text{UO}_2\text{-H}_2\text{O}$ with
a full water reflector; calculations were made with the OPT-SN program.)

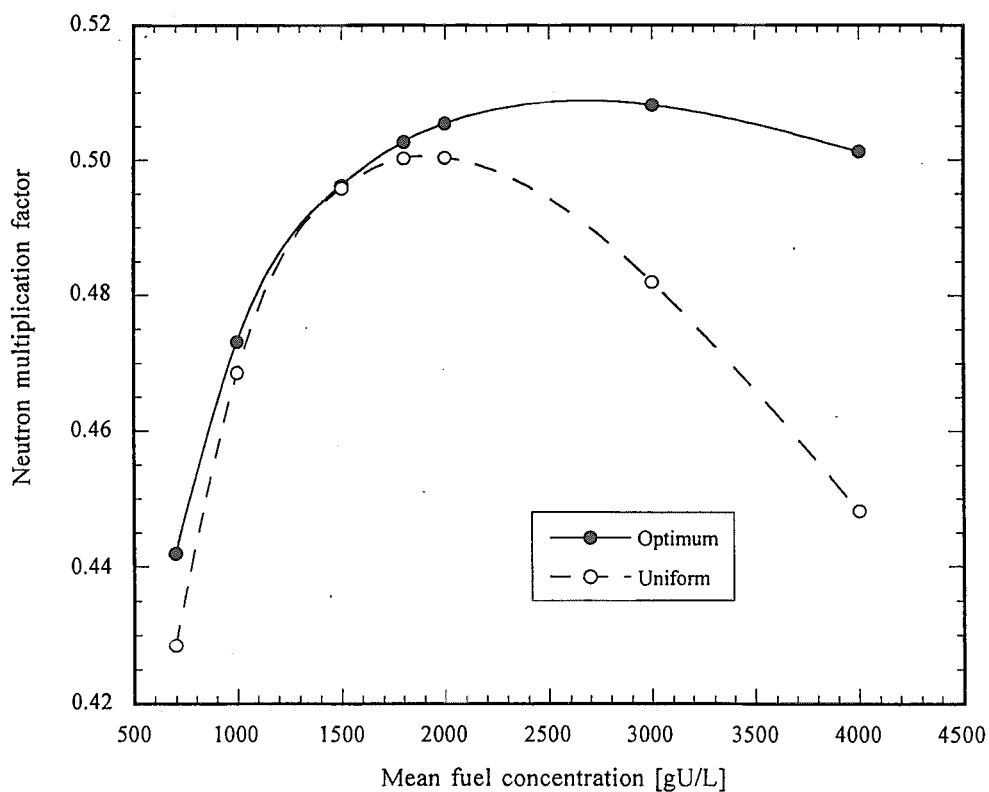


Fig 3.4 The neutron multiplication factors under optimum and uniform distributions (2)
(a 10-cm thick slab of homogeneous 5wt% ^{235}U -enriched $\text{UO}_2\text{-H}_2\text{O}$ without a full
water reflector; calculations were made with the OPT-SN program.)

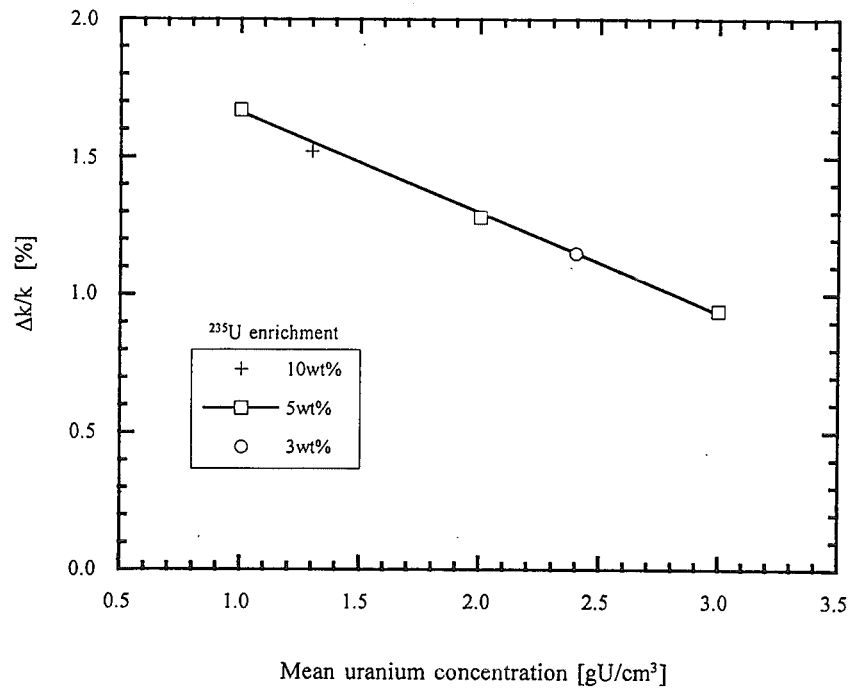


Fig 3.5 Heterogeneous effect of homogeneous low ^{235}U -enriched (10 wt% or less) $\text{UO}_2\text{-H}_2\text{O}$ systems (%increase in the infinite multiplication factor (heterogeneous vs.homogeneous) of a fuel systems containing 0.1 mm diameter fuel particles)

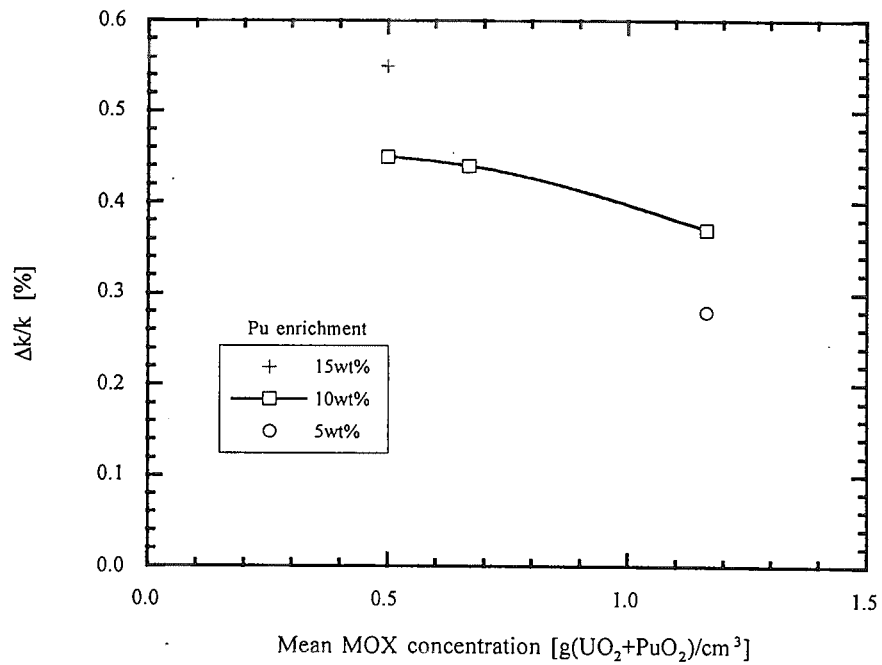


Fig 3.6 Heterogeneous effect of heterogeneous low Pu-enriched (15 wt% or less) $\text{MOX-H}_2\text{O}$ systems (%increase in the infinite multiplication factor (heterogeneous vs. homogeneous) of fuel systems containing 0.1 mm diameter fuel particles)

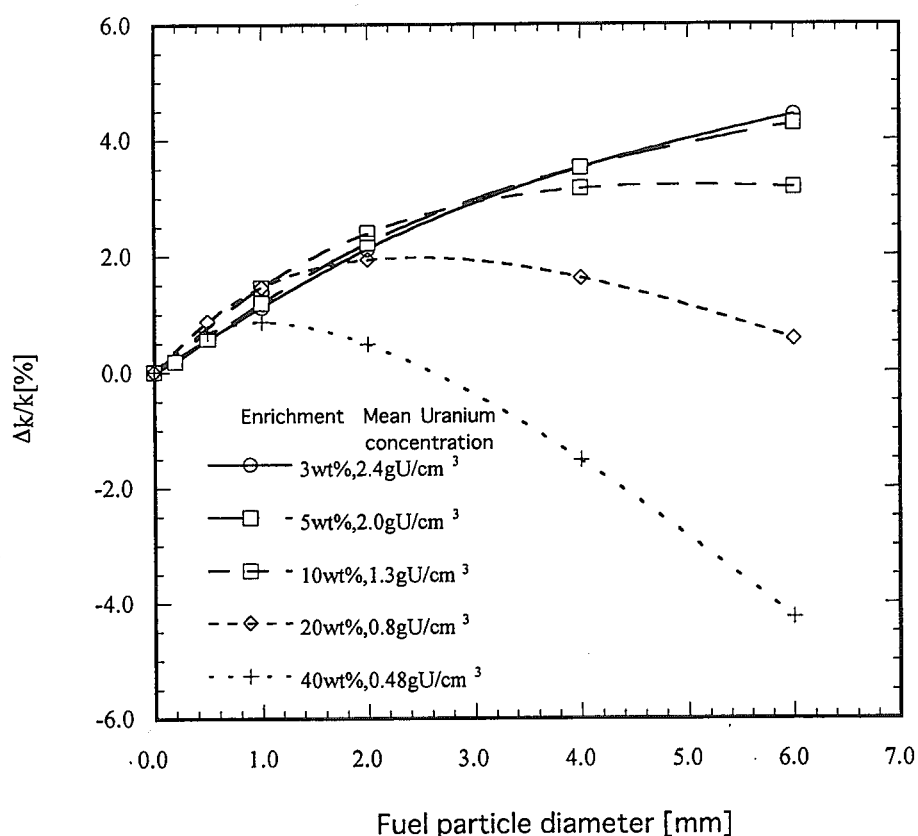


Fig 3.7 The increase rate of the infinite multiplication factor ($\% \Delta k/k$) from corresponding homogeneous systems (An array system of low-enriched uranium dioxide spherical fuel immersed in water. The water to fuel volumetric ratio was set so that this ratio gives the optimum moderation for the homogeneous system.)

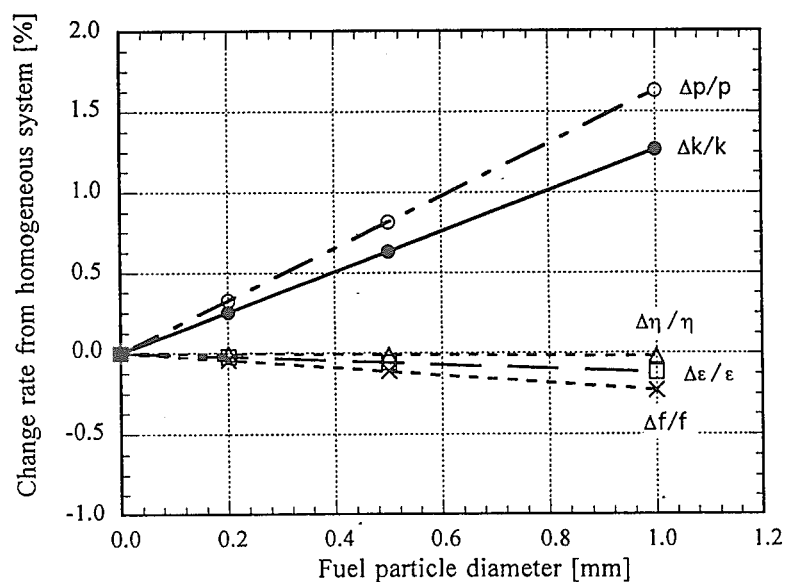


Fig 3.8 The change rates of the infinite multiplication factor and its four factors (homogeneous vs. heterogeneous) (5wt% ^{235}U enriched UO_2 fuel particles immersed in water, mean uranium concentration: 2gU/cm^3)

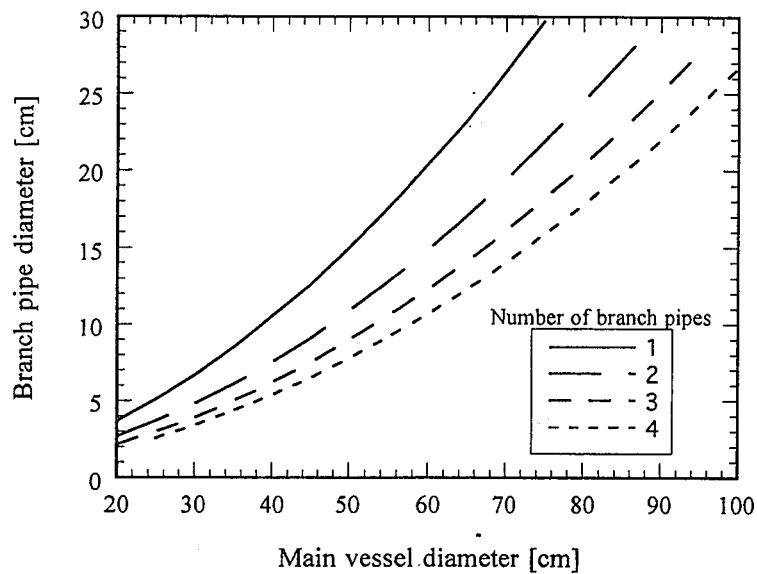


Fig 3.9 The number of branch pipes and diameters that can be neglected
Branch pipes are perpendicularly connected with the side surface
of a main vessel

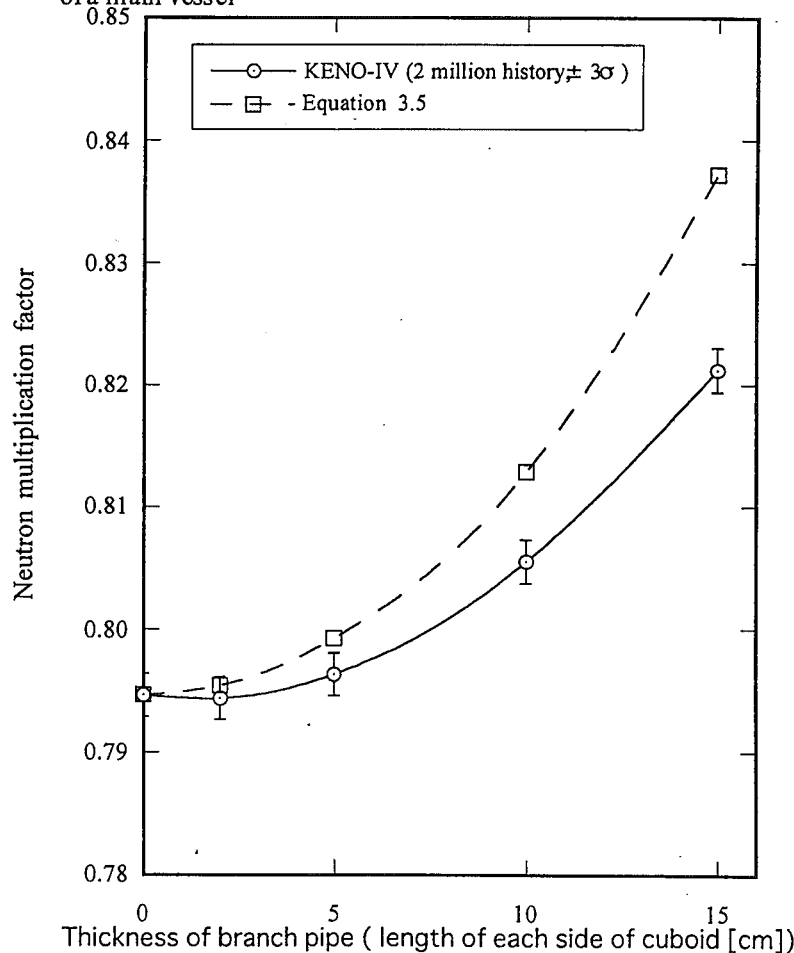


Fig 3.10 Calculated neutron multiplication factors for a quadrangular prism system
containing a uranyl nitrate solution (5wt% ^{235}U -enrichment, uranium
concentration 1000gU/L) On the center of each surface of the cuboid
(length of each side: 32cm, height: 1m), one cuboid (length: 50cm) was
perpendicularly attached.

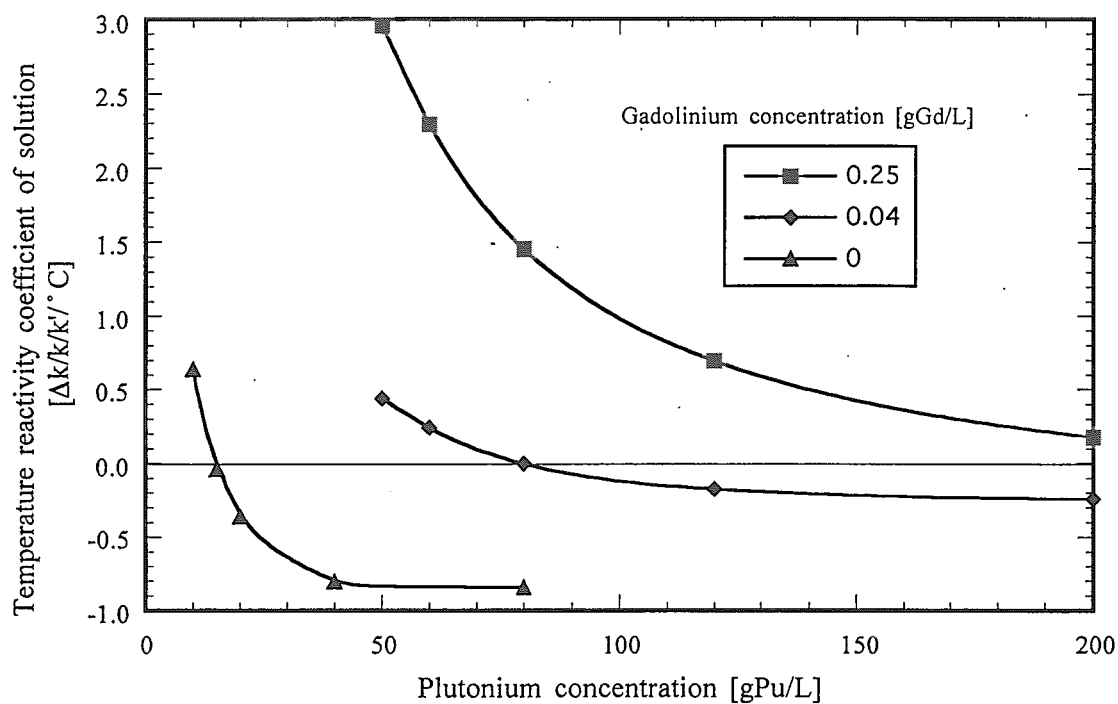


Fig 3.11 Temperature reactivity coefficients of plutonium nitrate solution including gadolinium (Pu isotope weight ratio $^{238}\text{Pu}:^{239}\text{Pu}:^{240}\text{Pu}:^{241}\text{Pu}:^{242}\text{Pu}=1.2:64.3:24.9:5.1:4.4$, nitric acid molarity 3N, a circular-cylindrical of 39cm indiameter and 100cm in height with a sufficiently thick water reflector)

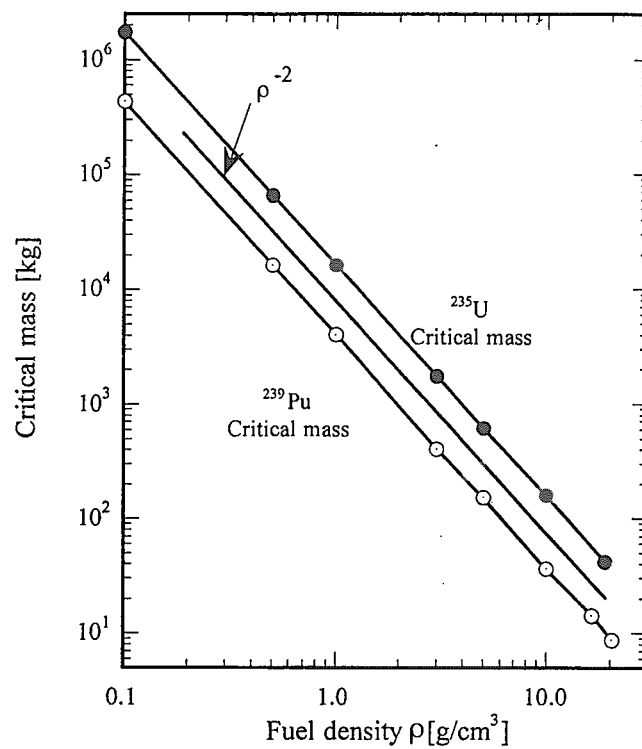


Fig 3.12 Critical mass and fuel density of ^{235}U and ^{239}Pu

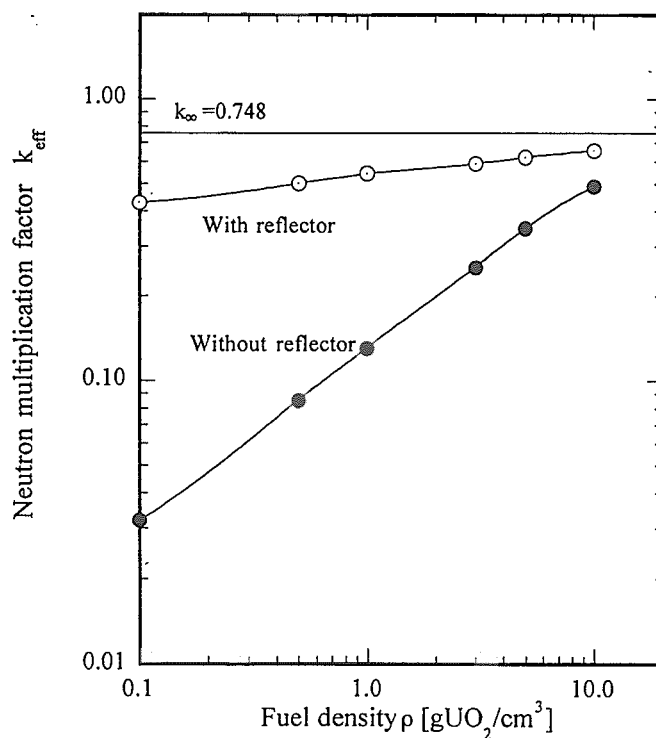


Fig 3.13 Critical mass and fuel density (for a sphere)
 Calculations were made for UO_2 powder fuel
 (^{235}U enrichment 4 wt%, H/U ratio 0, fuel mass 2145 kg UO_2)

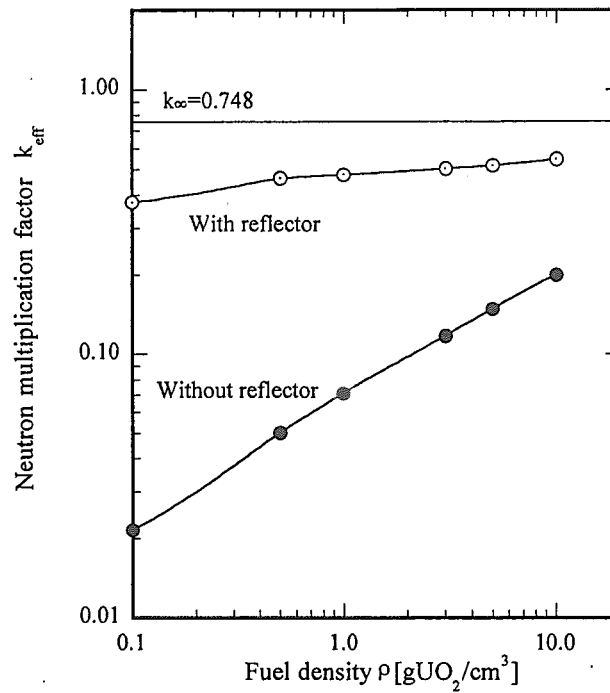


Fig 3.14 Critical mass and fuel density (for a circular cylinder, 10m high)
Calculations were made for UO_2 powder fuel
(^{235}U enrichment 4wt%, H/U ratio O, fuel mass 2145 kg UO_2)

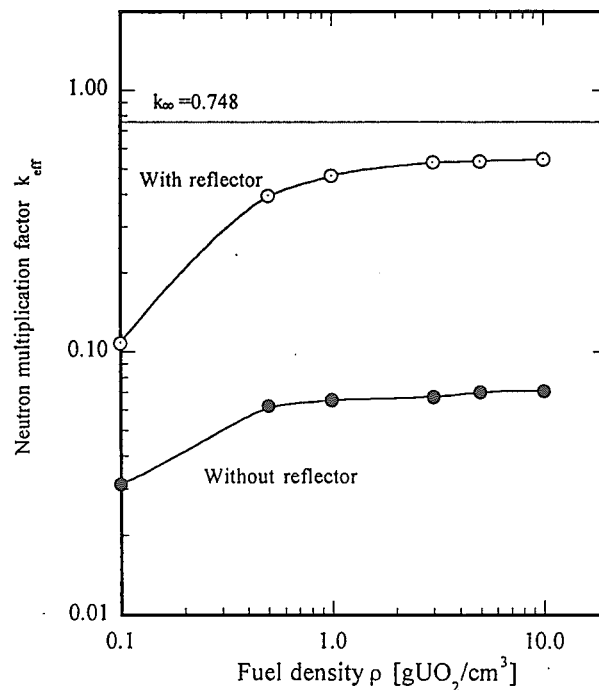


Fig 3.15 Critical mass and fuel density (for a slab, 3m side)
Calculations were made for UO_2 powder fuel
(^{235}U enrichment 4wt%, H/U ratio O, fuel mass 2145kg UO_2)

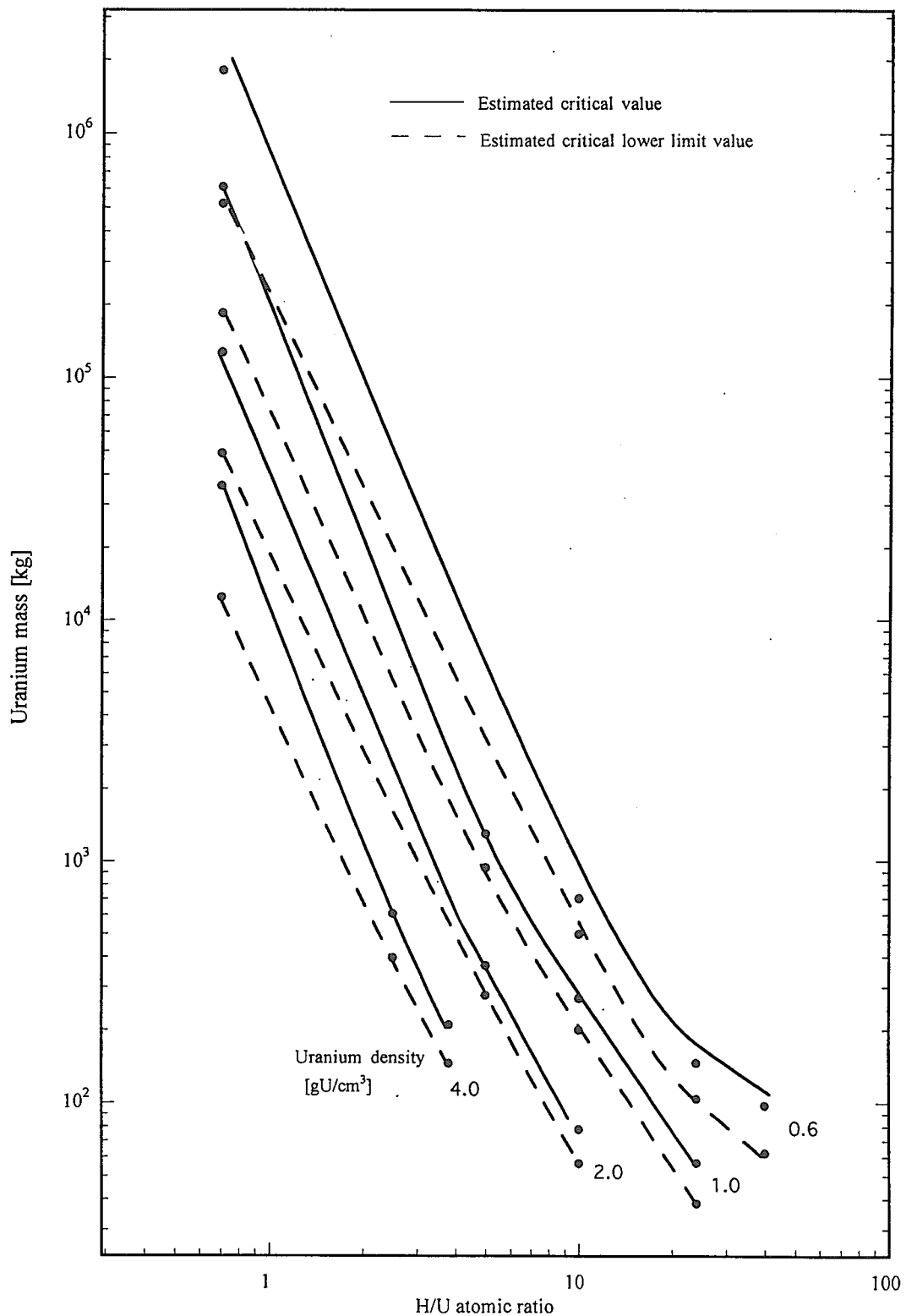


Fig 3.16 Estimated criticality value and estimated criticality lower limit value of uranium mass as a function of H/U atomic ratio in UO₂ powder fuel (²³⁵U enrichment 4wt% with a water reflector)

The values in the figure represent uranium density. The H/U atomic ratio is varied with the remaining constant.

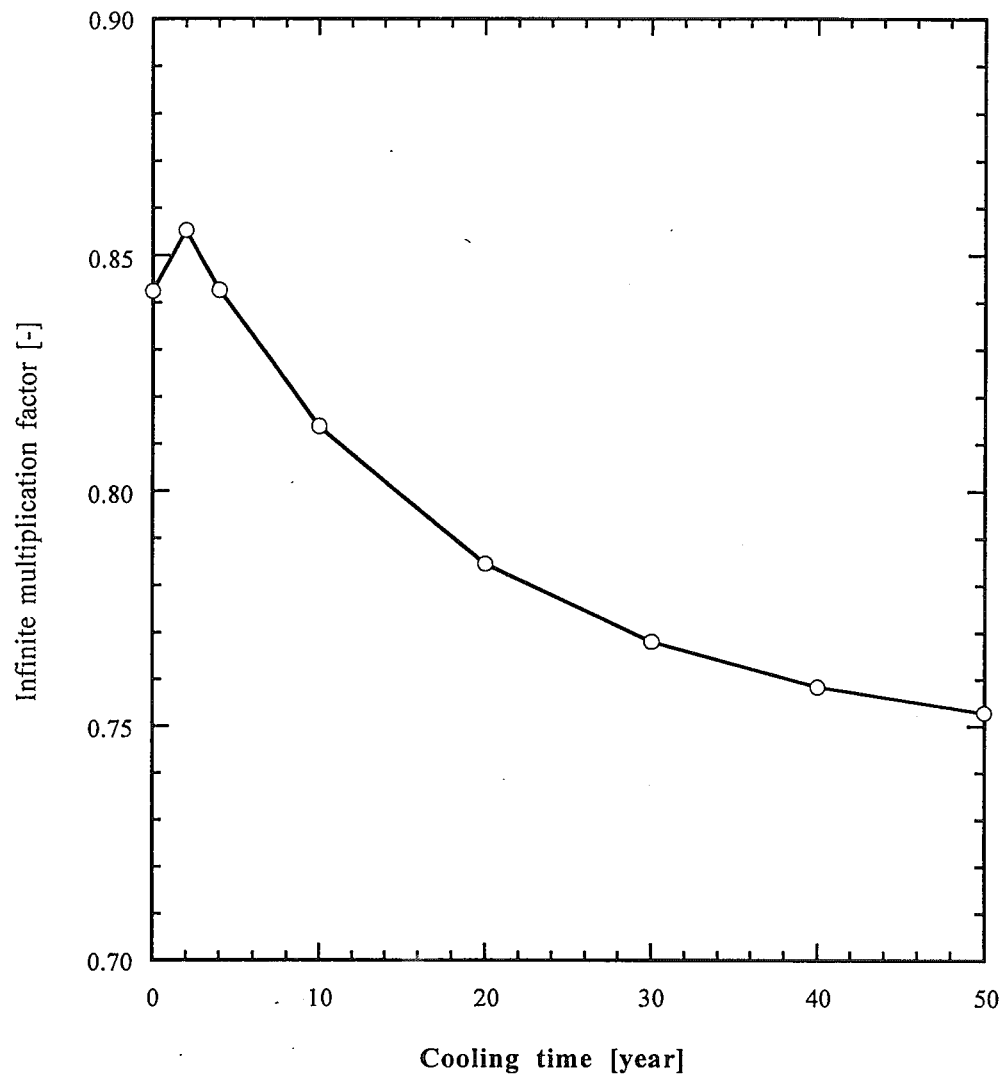


Fig 3.17 Effect of the cooling time on the infinite multiplication factor
(initial enrichment 4.1wt%, an infinite array of PWR fuel rods)

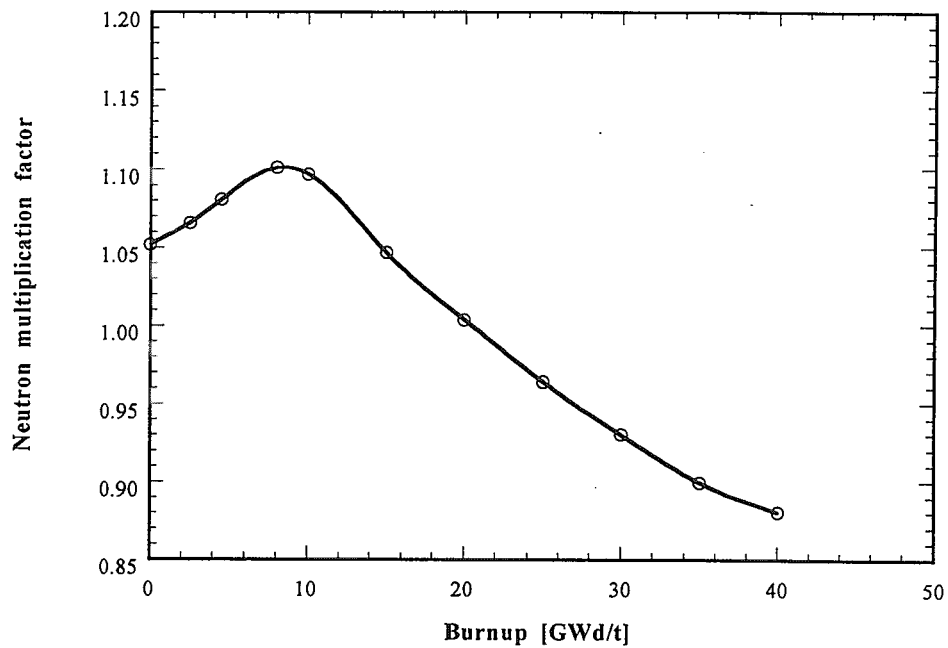


Fig 3.18 Changes in the infinite multiplication factor of an infinite array of BWR fuel assemblies as a function of burnup (moderator void ratio 40%)

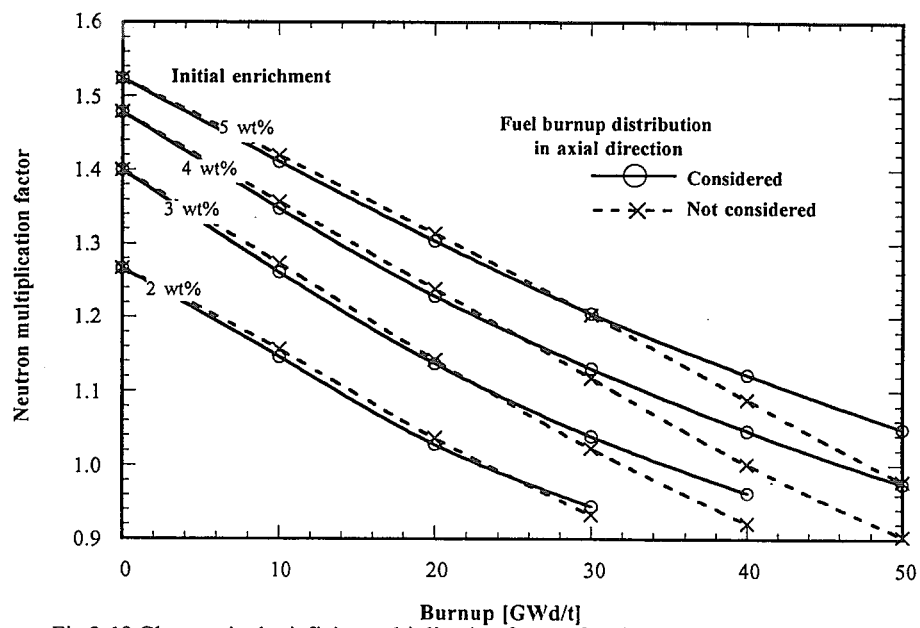


Fig 3.19 Changes in the infinite multiplication factor of an infinite array of BWR fuel assemblies as a function of burnup

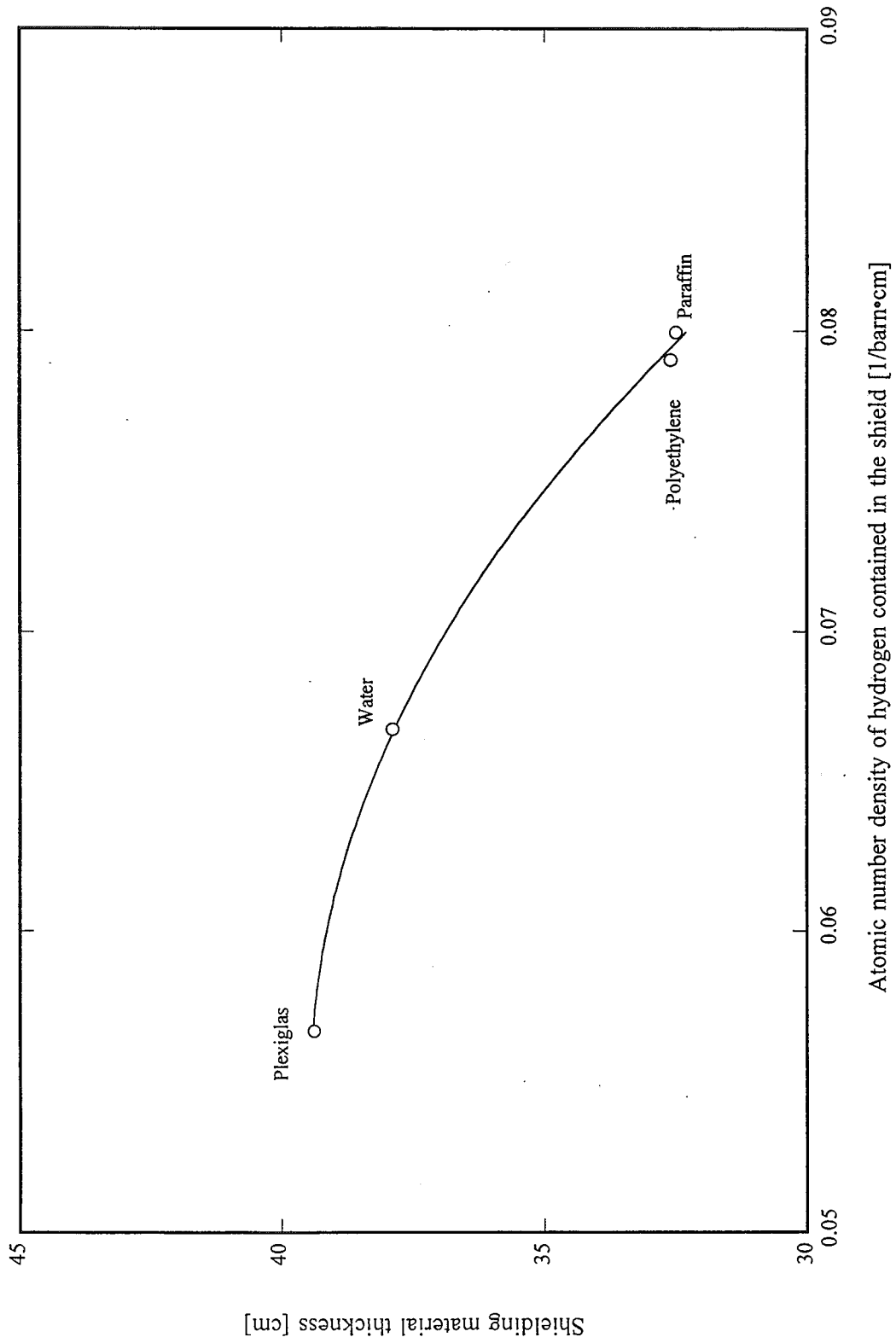


Fig 3.20 Thickness of neutron shielding materials having the same shielding effect as hydrogen contained in shielding

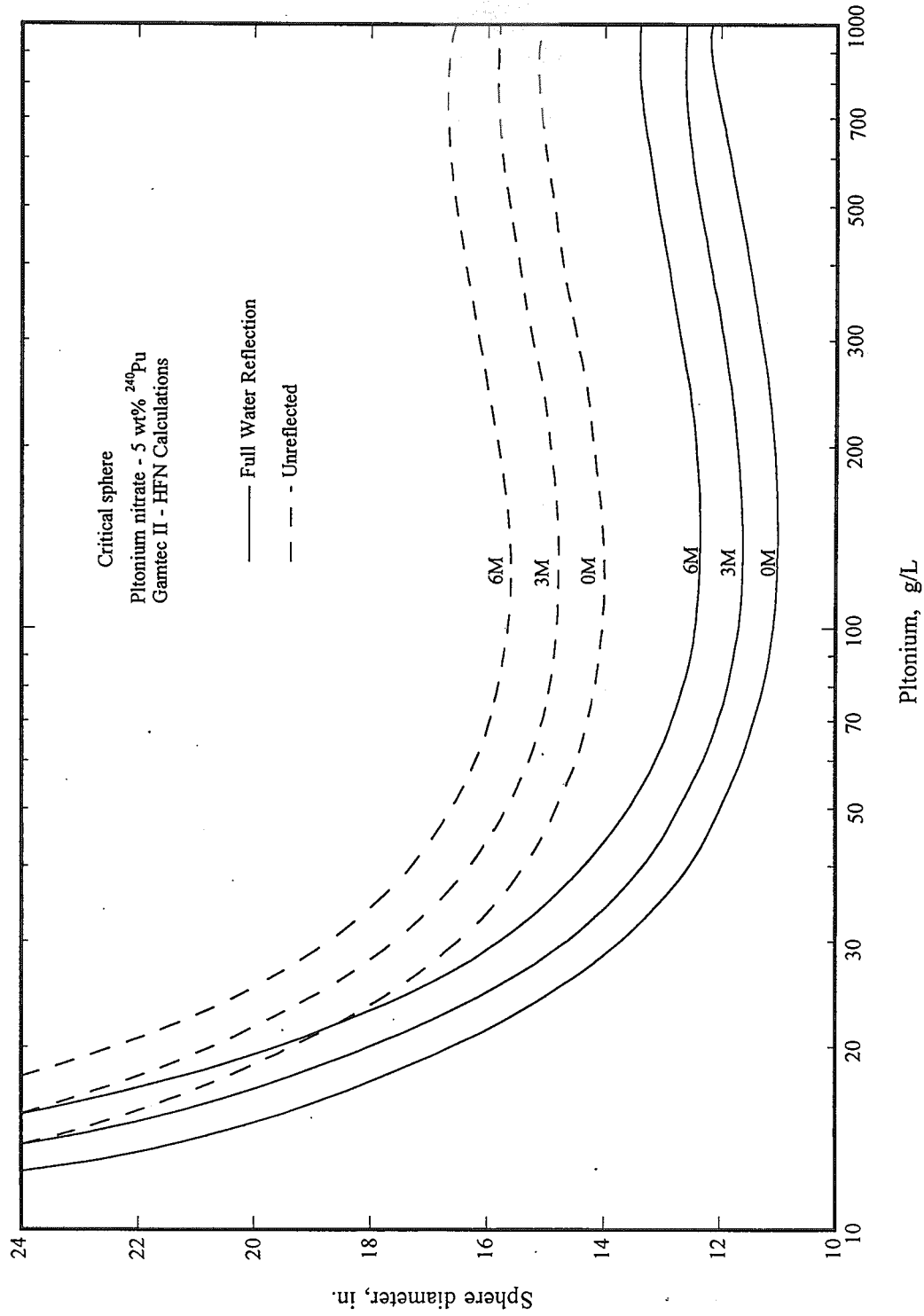


Fig 3.21 Critical sphere diameter of plutonium nitrate solution (4)
(The values shown in the figure represent the concentration of nitric acid.)

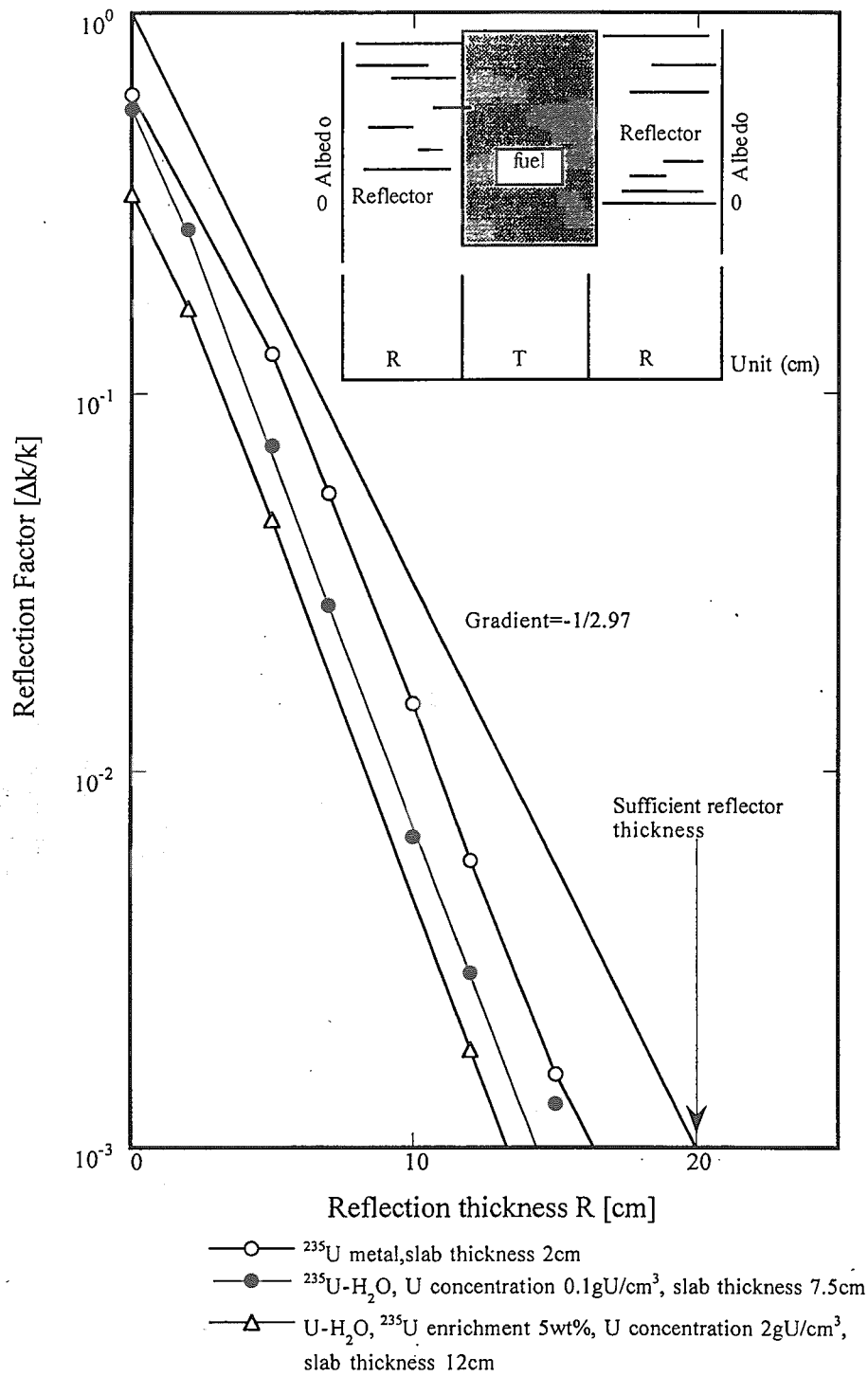
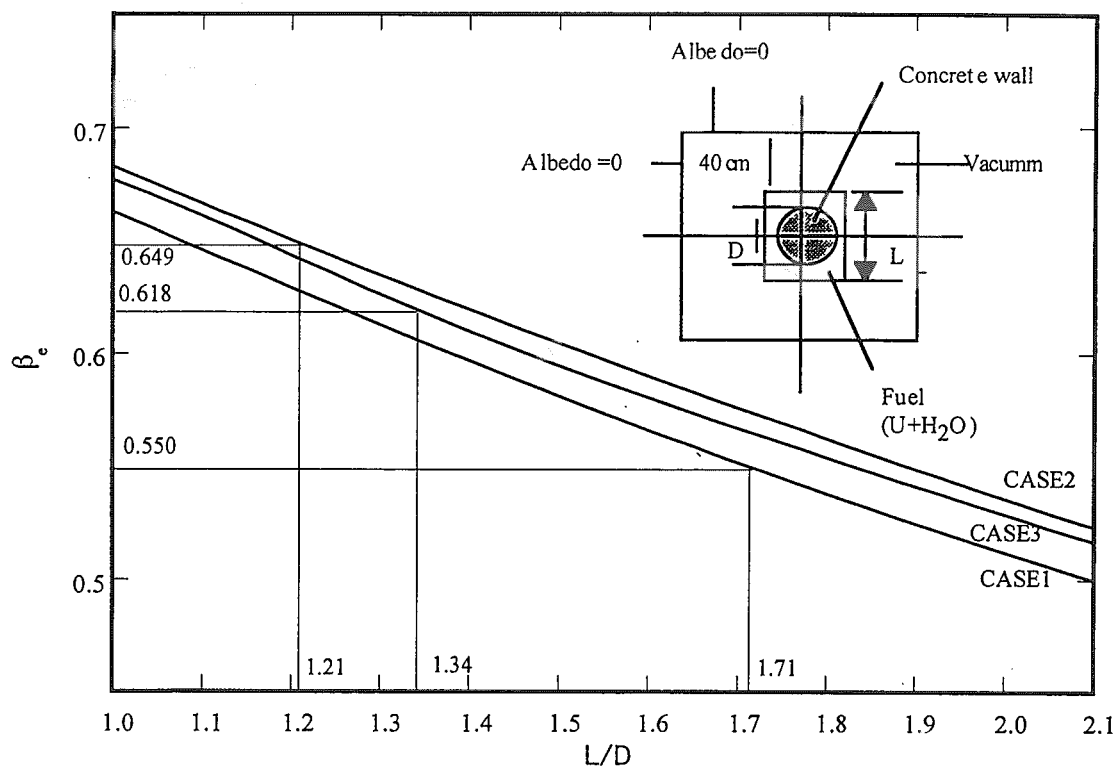


Fig 3.22 Reflection factor as a function of reflector thickness in an infinite slab system



| Case No. | Diameter of infinitely long circular cylinder | ^{235}U enrichment | Uranium concentration |
|----------|---|-----------------------------|-----------------------|
| | [cm] | [w/o] | [gU/cm ³] |
| Case 1 | 8.1 | 93.5 | 18.0 |
| Case 2 | 17.6 | 100.0 | 0.1 |
| Case 3 | 24.0 | 5.0 | 4.5 |

Fig 3.23 Relation between L/D of ordinary concrete reflector and effective albedo, β_e (The diameter and composition of the fuel for each CASE are shown in the accompanying table. The composition of ordinary concrete are those described in Table 2.6 of the "Nuclear Criticality Safety Data Collection." The values of 0.550, 0.618, and 0.649 in the diagram are the albedo values of a water reflector corresponding to each case.)

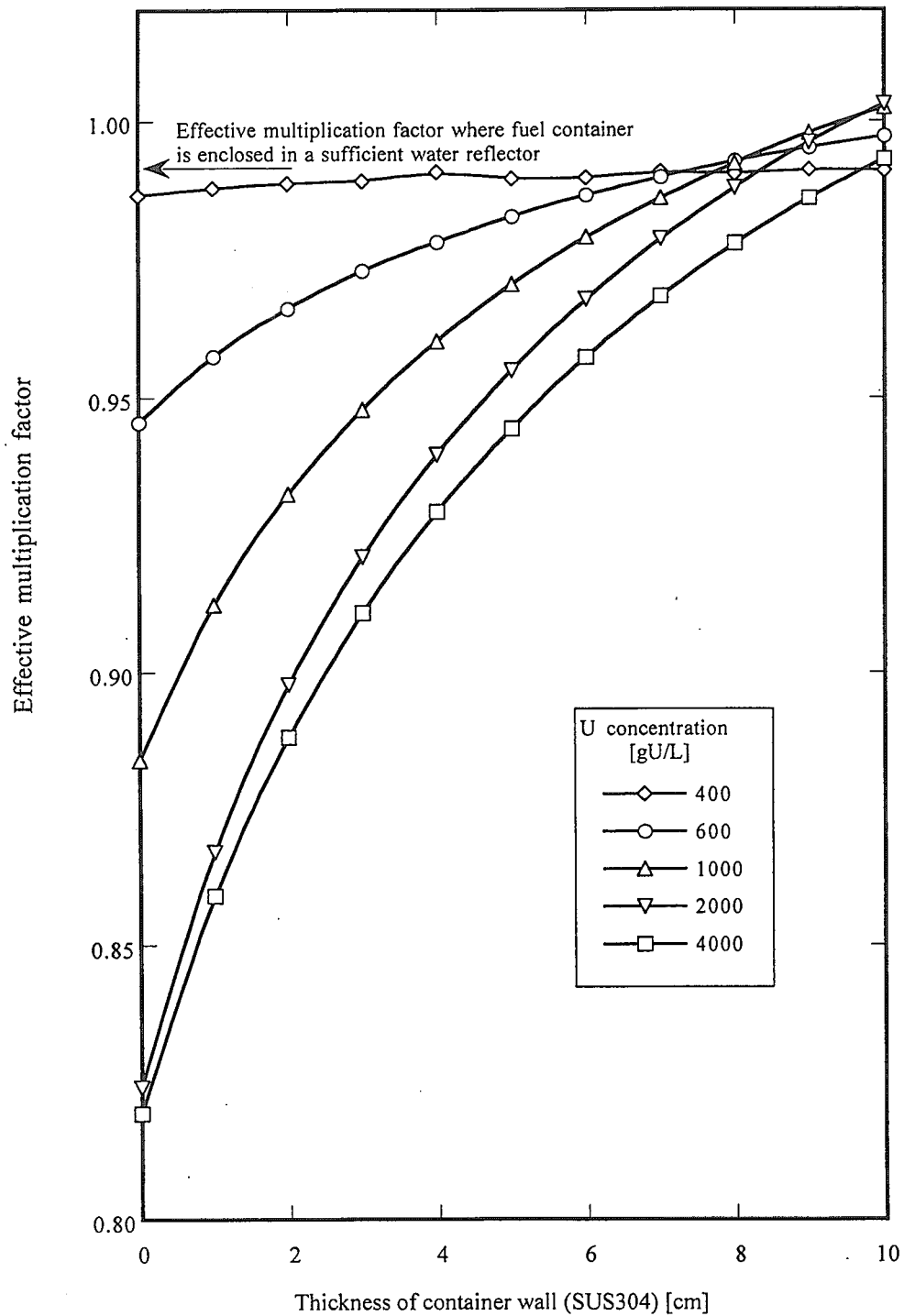


Fig 3.24 Changes in effective multiplication factor due to changes in thickness of a container wall of homogeneous $\text{UO}_2 - \text{H}_2\text{O}$ fuel system
(infinite circular cylinder, enrichment 4wt%, container surrounded by a vacuum)

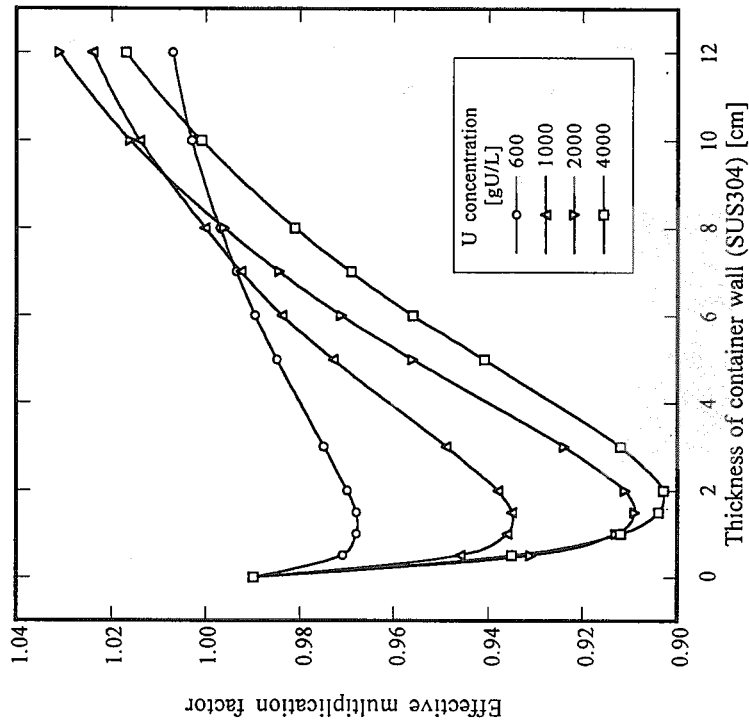


Fig 3.26 Changes in effective multiplication factor due to changes in thickness of container wall (SUS-304) of homogeneous UO_2 - H_2O fuel system (infinite slab, enrichment 4 wt%, container enclosed in a 30 cm thick water reflector)

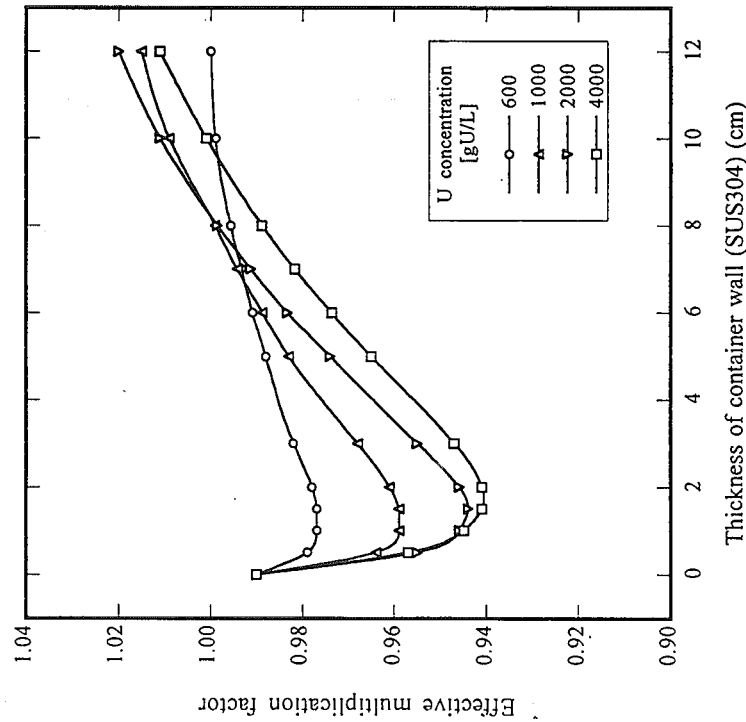


Fig 3.25 Changes in effective multiplication factor due to changes in thickness of container wall (SUS-304) of homogeneous UO_2 - H_2O fuel system (infinite circular cylinder, enrichment 4 wt%, container enclosed in a 30 cm thick water reflector)

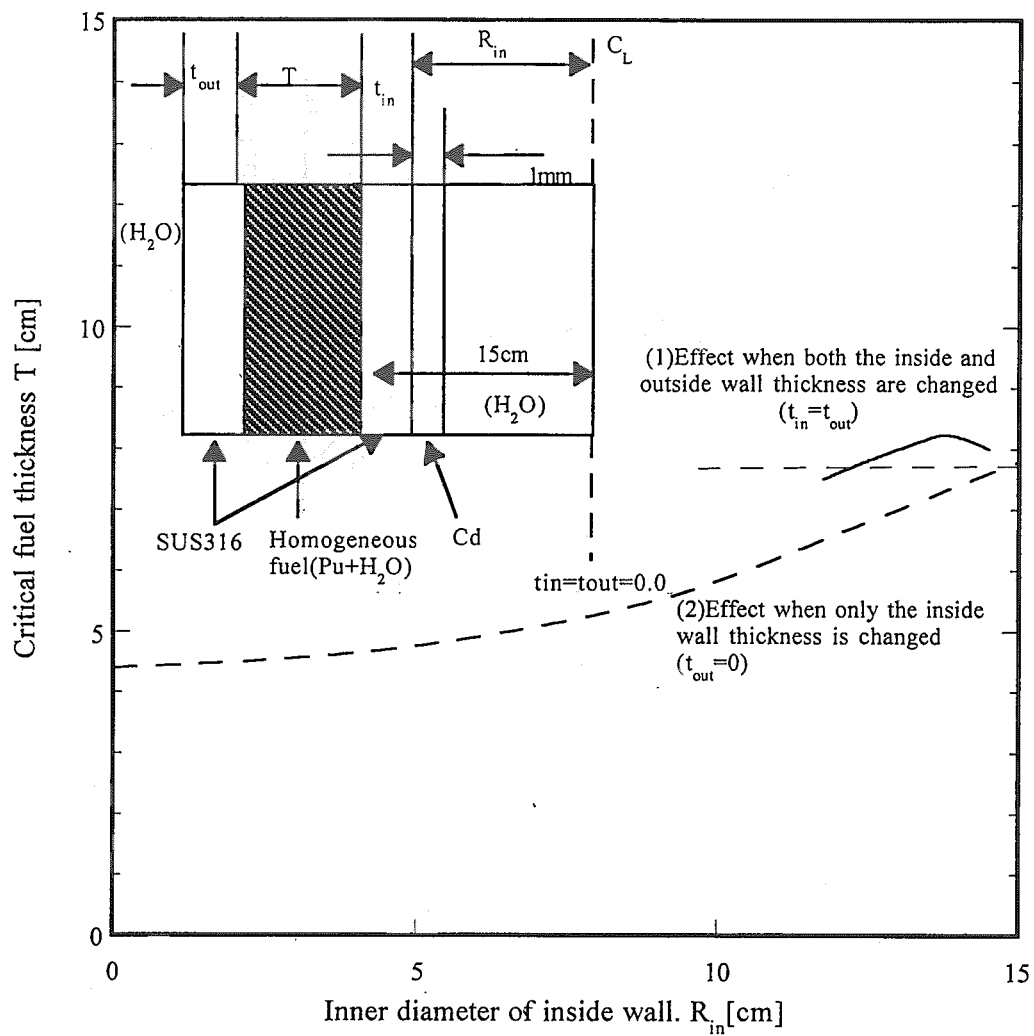


Fig 3.27 Thickness of critical fuel to the changes in thickness of SUS316 container wall of annular tank containing homogeneous Pu-H₂O

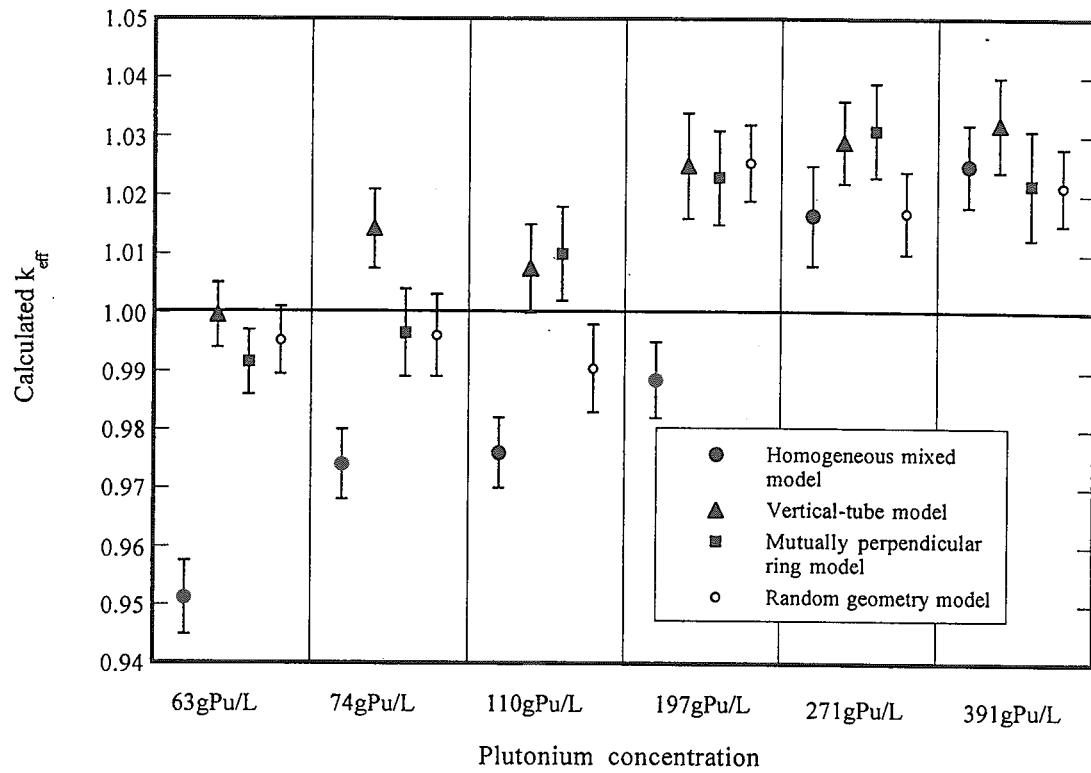


Fig 3.28 Calculated values of k_{eff} resulting from different models of Raschig rings (48)

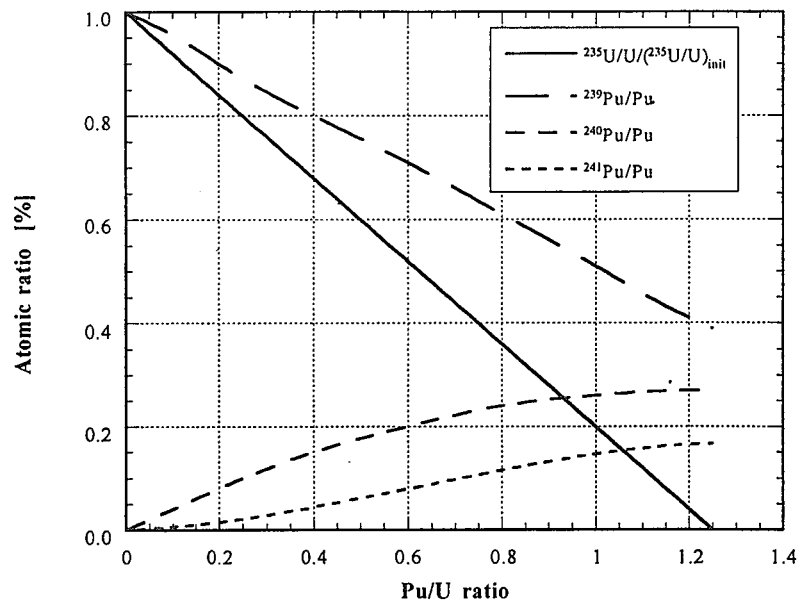


Fig 3.29 Fuel depletion and changes in uranium and plutonium composition

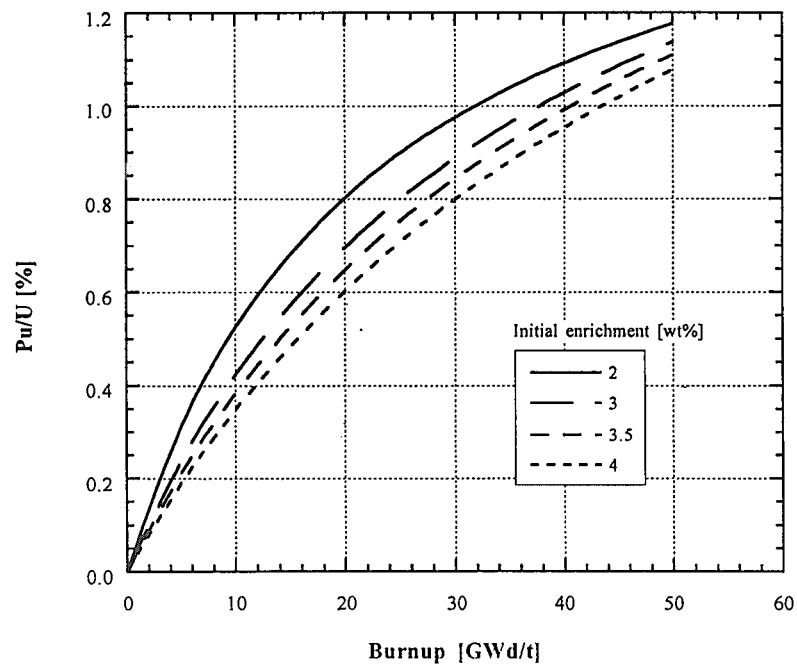
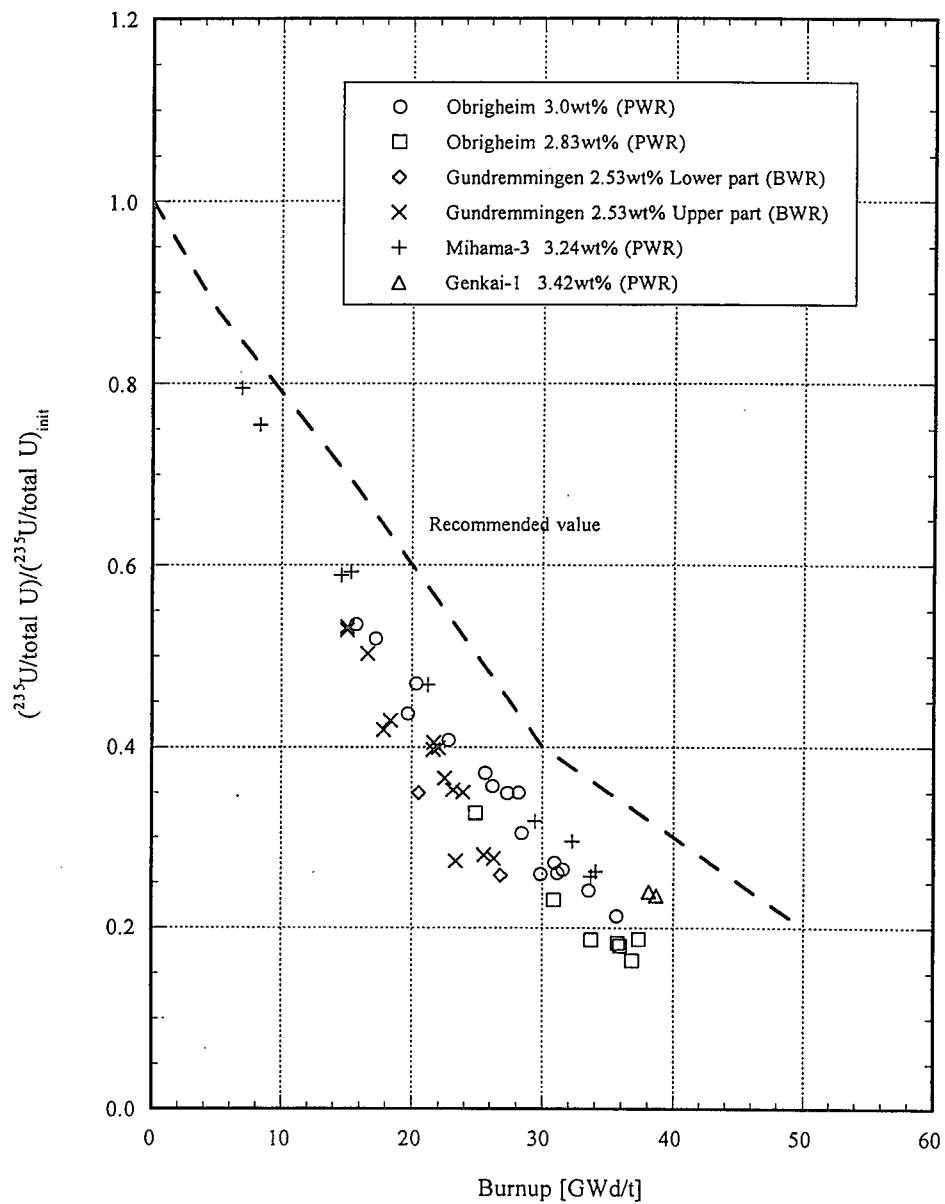


Fig 3.30 Pu/U ratio [%] as a function of burnup for various initial enrichment fuels

Fig 3.31 Fuel depletion and change in $(^{235}\text{U}/\text{total U})/(^{235}\text{U}/\text{total U})_{\text{init}}$

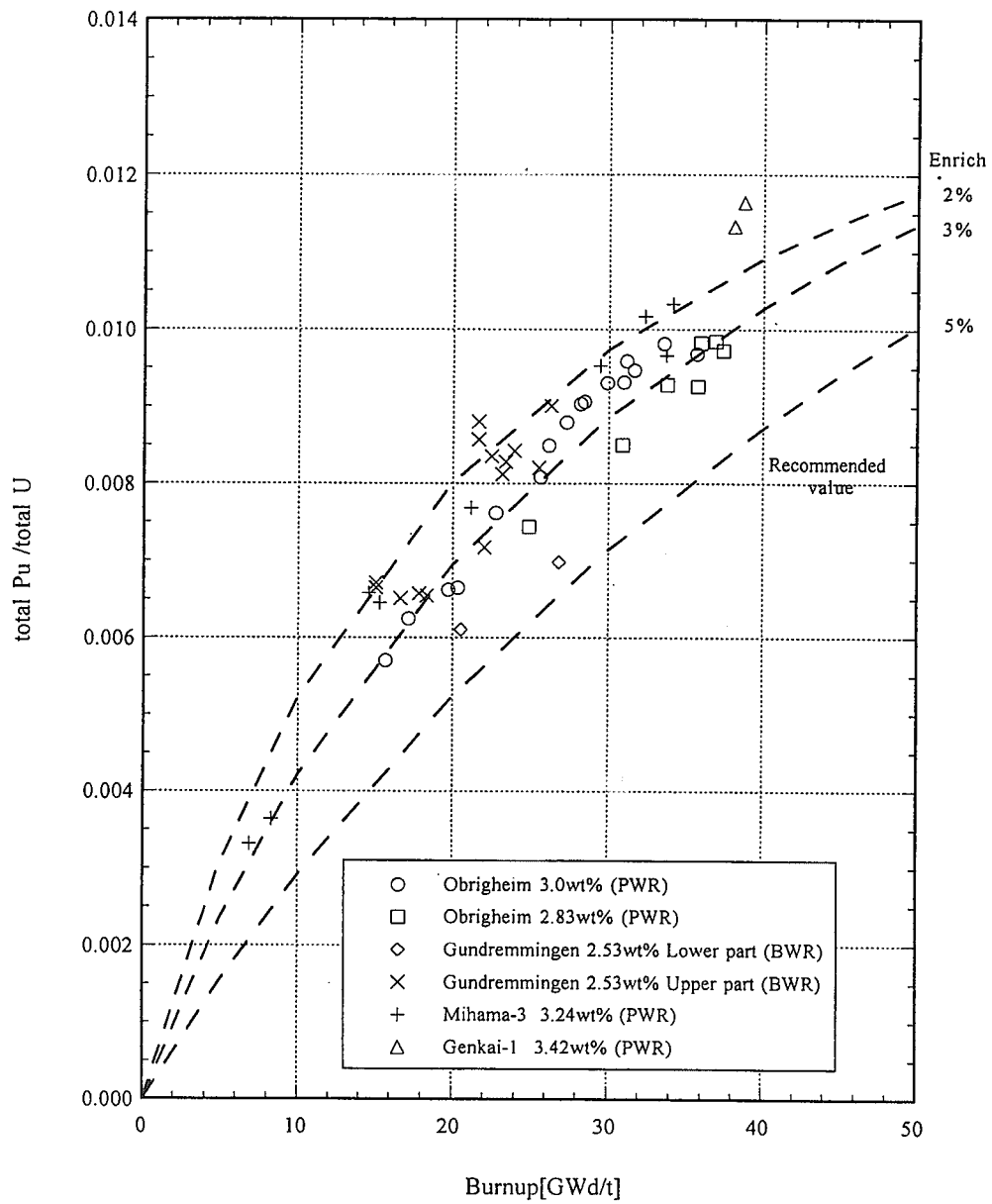
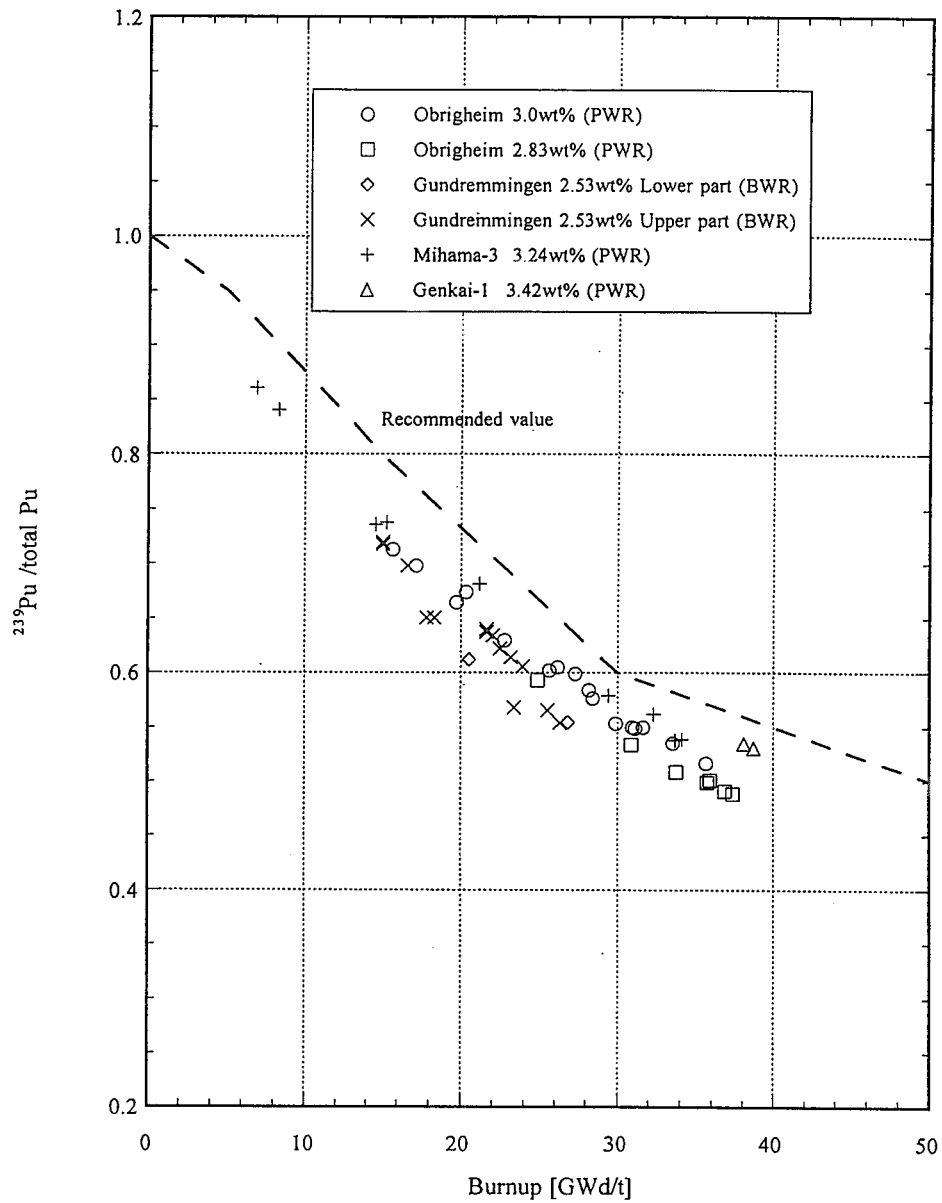


Fig 3.32 Fuel depletion and change in total Pu/total U

Fig 3.33 Fuel depletion and change in $^{239}\text{Pu}/\text{total Pu}$

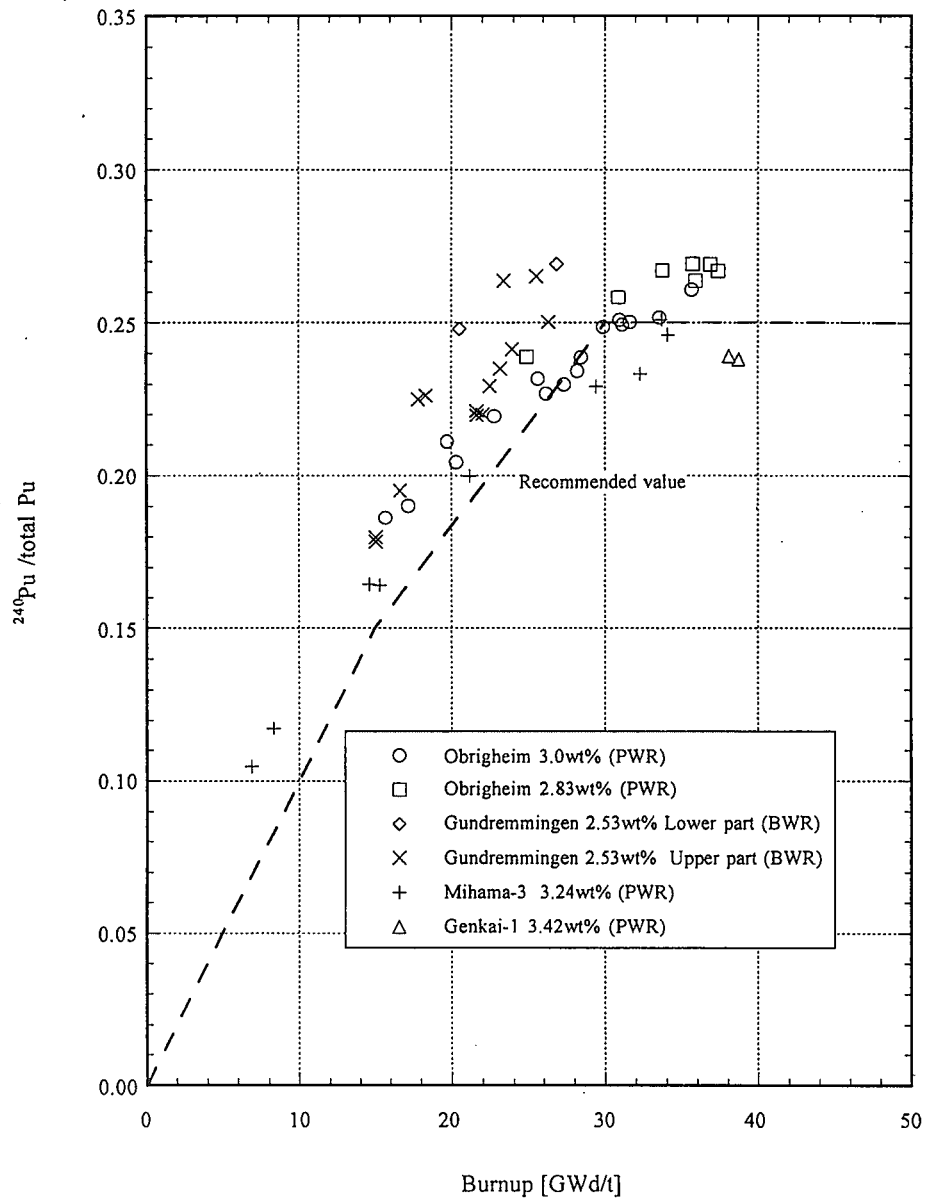
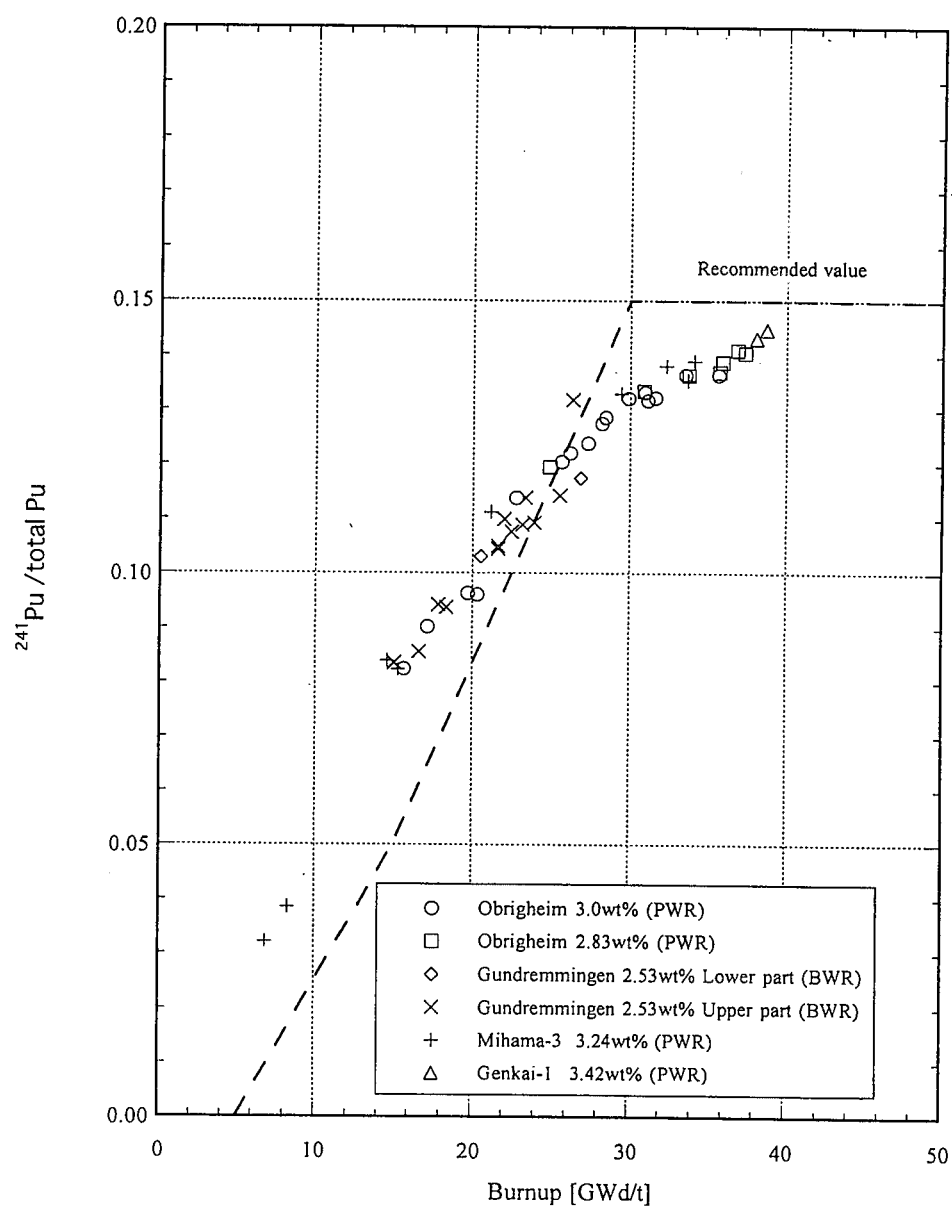
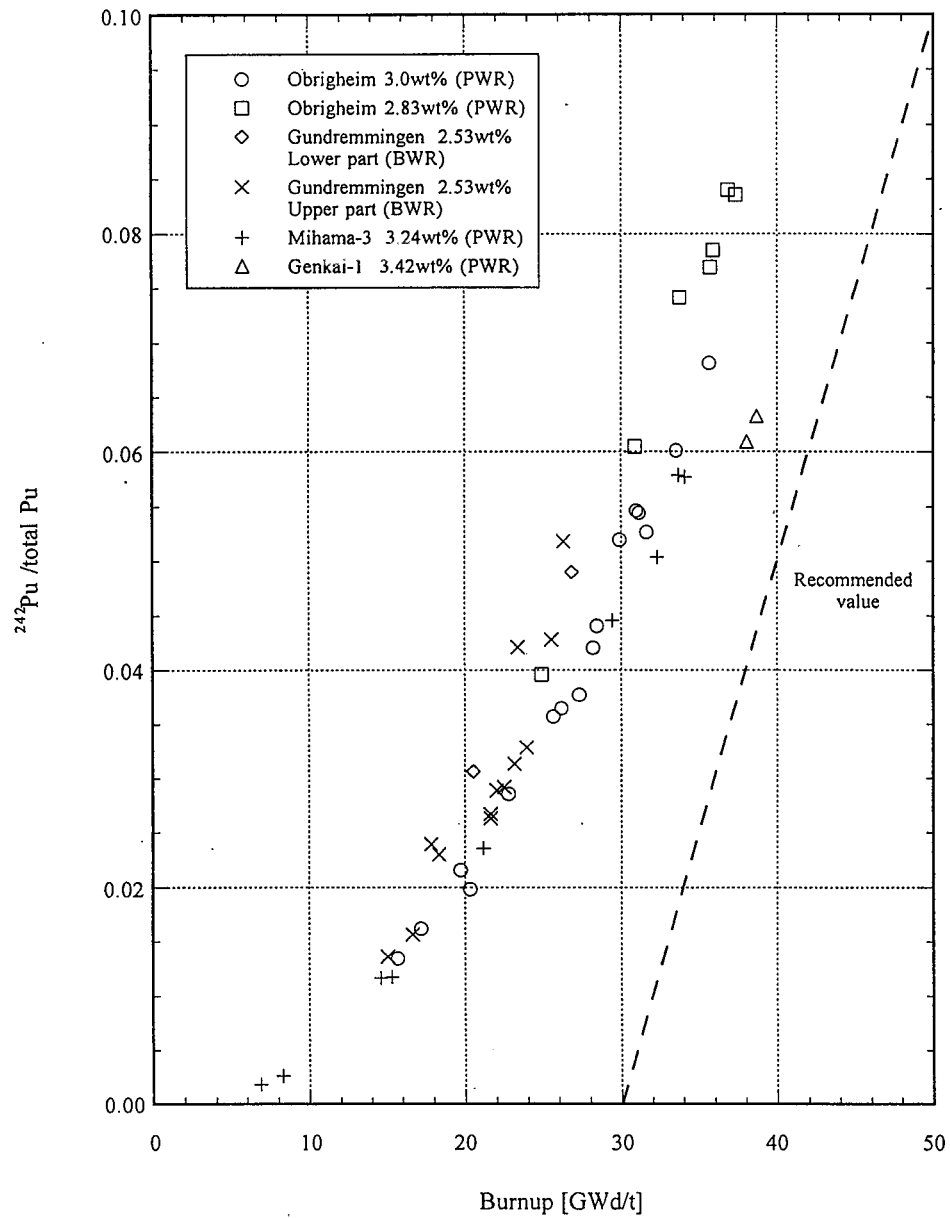


Fig 3.34 Fuel depletion and change in $^{240}\text{Pu}/\text{total Pu}$

Fig 3.35 Fuel depletion and change in $^{241}\text{Pu}/\text{total Pu}$

Fig 3.36 Fuel depletion and change in $^{242}\text{Pu}/\text{total Pu}$

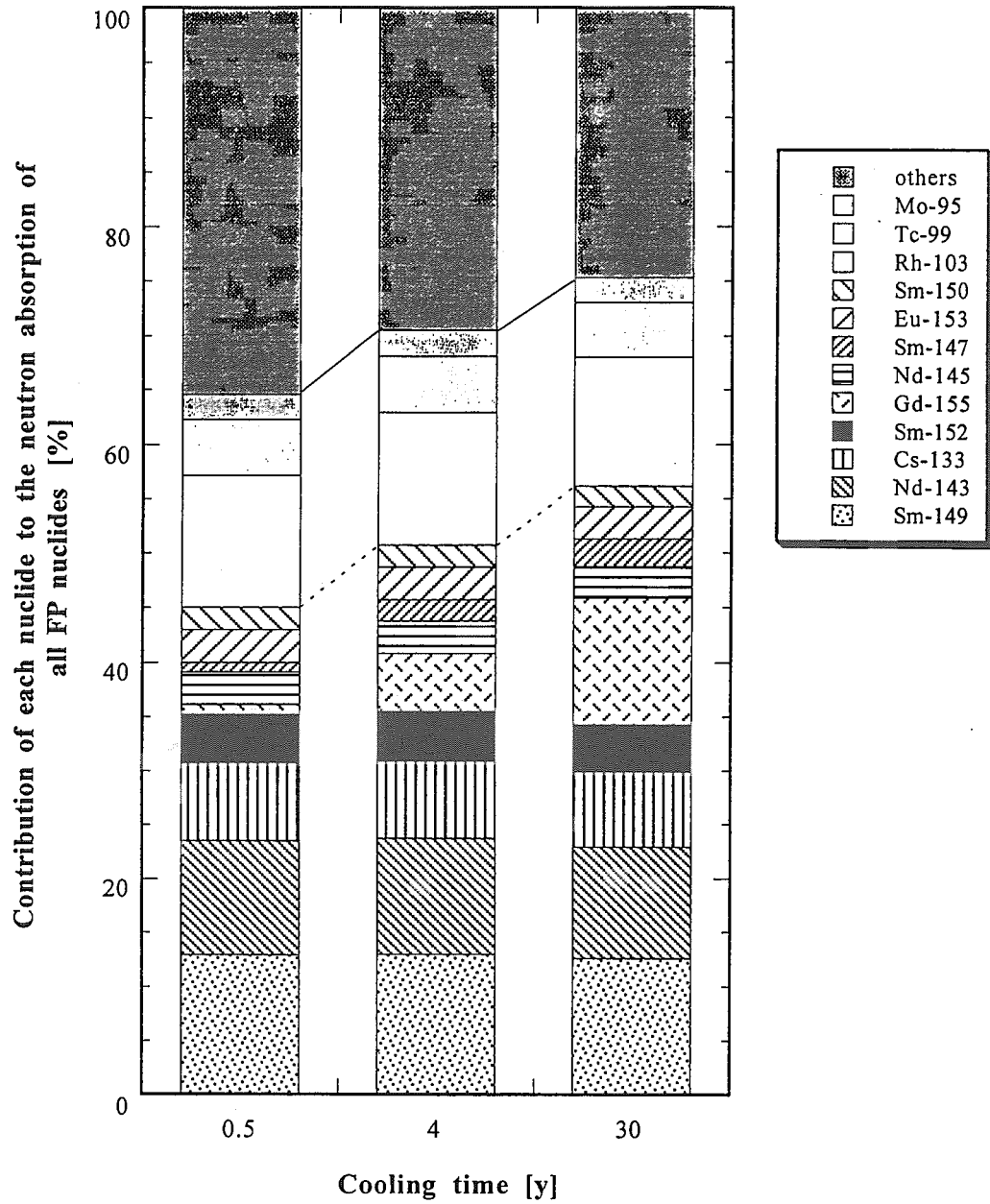


Fig 3.37 Contribution (%) of each nuclide to the neutron absorption of all FP nuclides (Example of PWR $U(4.15)O_2$ and burnup 30 GWd/T)

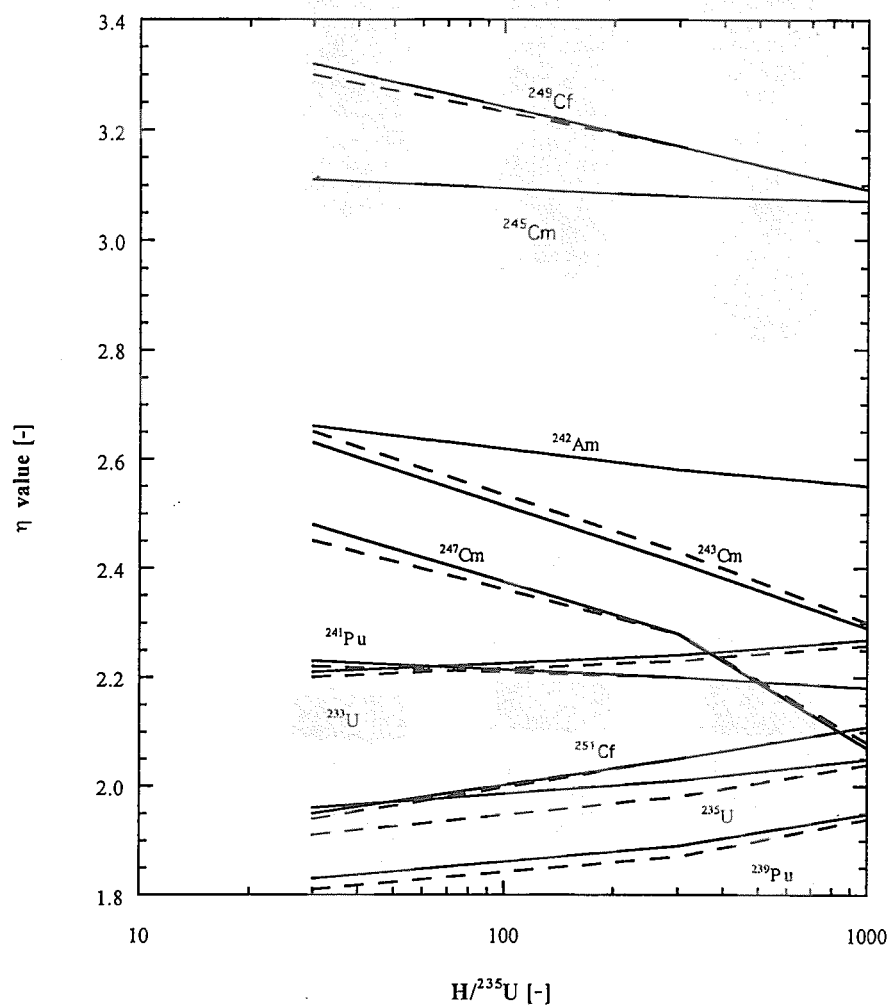


Fig 3.38 η -values for each of the nuclides

(Solid line shows value for $\delta_0 = 10^2 \text{ b}$, and dashed line shows values for $\delta_0 = 10^8 \text{ b}$)

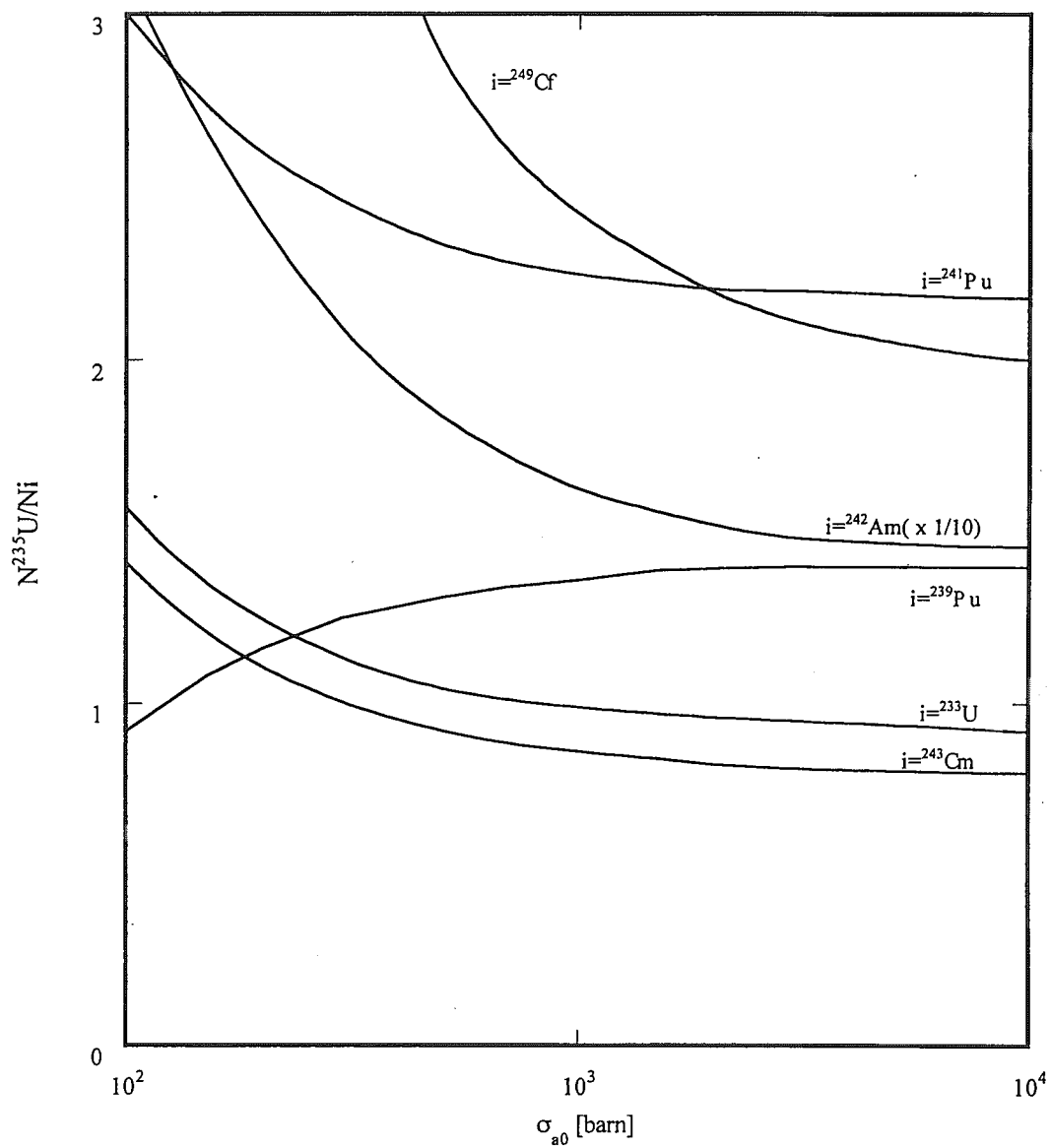


Fig 3.39 Comparisons of nuclide reactivity worth
(number of ^{235}U atoms equivalent in reactivity to one object nuclide atom)

4. METHODOLOGY FOR ANALYTICAL SAFETY ASSESSMENT

In criticality safety assessment for nuclear fuel installations, it is necessary to assess both single units and the mutual interaction of neutrons in multiple units. This assessment must consider the diversity of the physical and chemical forms of the nuclear materials handled. It also must consider that these forms are subject to change and that there a lot of system, facilities and equipment installed containing nuclear materials. The procedures and practices for analytical safety assessments are described in this chapter.

4.1. Procedure for Analytical Safety Assessment

Analytical assessment is usually performed as follows. First, analytical assessments of single units corresponding to individual installations or pieces of equipment are made. Next, multiple units composed of these single units are assessed analytically. The procedure for this analytical assessment is shown in Fig. 4.1. The basic principles for each step of this diagram are summarized below.

(1) Determine the objects to be assessed in the installation

The objects of analytical assessment for critical safety are principally all the installations and equipment handling nuclear fuel materials. However, both depleted uranium and natural uranium may be omitted from the safety assessment. In addition, nuclear fuel materials whose k_{∞} is less than 1.0 may be omitted from the safety assessment. This is because it has been verified that uranium and plutonium fuels of any convex geometry for which the infinite multiplication factor is less than 1.0 do not reach criticality due to the reflection effect.⁽¹⁾ The enrichment and concentration limit values of uranium and plutonium satisfying $k_{\infty} < 1.0$, are dependant on the chemical forms. One example of this is described in Section 4.3.1. An installation or piece of equipment containing no nuclear fuel material in normal operation should be also assessed if there is any possibility that nuclear fuel material would flow into it.

(2) Set up the single units

Normally, a single unit is set up for each piece of equipment. Since the term "single unit" is used for the minimum unit for criticality control, a group of complex pieces of equipment may be set up as a single unit.

(3) Set up assessment conditions for each single unit

First, determine chemical composition, isotopic composition, concentration of the solution, density, etc. If several pieces of equipment are to be grouped to form a single unit in the assessment conditions for nuclear fuel material, examine the neutron moderating conditions, etc., between these pieces of equipment and set up the conditions that are severest from the nuclear standpoint in the

ranges that are technically conceivable, including fluctuations for both the normal and abnormal states. Next, select the method for criticality control of the single unit. In addition, determine the reflection conditions needed for analytical assessment of the single unit. These reflection conditions should be the severest from the nuclear standpoint in the ranges that are technically conceivable. Fluctuations in the peripheral conditions due to submersion in water and the reflection effects of structural materials adjacent to the unit must be considered. When it is necessary to assume submersion of the unit in water, a water reflector having a thickness of more than 20 cm should be used.

(4) Prepare a model for each single unit system

Considering the assessment conditions for a single unit, prepare a geometric model for analysis and calculate the atomic number density for each region of the model.

(5) Perform the criticality analysis for each single unit system

A criticality analysis for each single unit is performed with a point approximation code that is applicable to simple models or with a detailed calculation multigroup or continuous energy code.

When an analytical result is less than the estimated lower limit criticality multiplication factor, the criticality-limits for the single unit should be selected to conform to the modeling conditions. When the analytical result exceeds the estimated lower limit criticality multiplication factor, the assessment conditions for the single unit should be changed to ensure that the analytical value is less than the estimated lower limit criticality multiplication factor. The analysis should then be repeated. If the estimated lower limit criticality value for the single unit corresponding to the assessment conditions in step (3) can be used, the criticality limits for the single unit may be decided according to the estimated lower limit criticality value without going through steps (4) and (5). The estimated lower limit criticality values for the common types of single units are identified in (1) of Section 4.3.1 and are shown in Tables 4.1 through 4.5 and in the Nuclear Criticality Safety Handbook - Data Collection. After deciding the criticality-limits for all single units, the analytical assessment for multiple units should be performed.

(6) Set up the arrangement of the single units and the assessment area for the multiple units

Decide the installation location for single units according to the arrangement plan. If unit movement is a precondition, as for transporting containers, the route and range must be decided in advance. When it is necessary to consider submersion in water, the face-to-face isolation distance between single units must be more than the allowable face-to-face distance. The method to determine this distance is described in Section 4.4.1. When the arrangement of the single units is determined, the mutual interaction evaluation area of the system should be set up considering the existence of the shielding wall, etc.

(7) Set up assessment conditions for multiple units

Examine the neutron moderating conditions between multiple units and the reflection conditions on the periphery of the array of units. Select the severest nuclear conditions in the range technically conceivable including the fluctuations in the normal and abnormal states.

(8) Prepare a model of the multiple unit system

Prepare an array model for analysis according to the assessment conditions for multiple units. Calculate the atomic number density for each region of the model.

(9) Perform criticality safety analysis for the multiple unit system

Criticality safety analyses are performed using simple methods such as the solid-angle method, simple calculation codes, or detailed calculation codes.

If the analytical result satisfies the subcritical conditions for the analytical method used, the criticality safety for the multiple units is confirmed. If the analytical result does not satisfy the subcritical conditions, change the arrangement of the units or, depending on the situation, change the single units by altering their dimensions, etc., so that subcritical conditions can be satisfied.

4.2. Setting up Assessment Conditions

For each of the single units, determine the respective fluctuation ranges for the materials, the physical and chemical forms of the nuclear fuel materials to be handled, and the neutron absorption effect, moderation, and reflection conditions, etc. For multiple units, besides the above determinations, select the respective fluctuation ranges in the normal and abnormal states for the distance between units, the moderator between units, and the neutron reflector and shielding materials. After determining these fluctuation ranges, set up the conditions that impose the severest results for criticality assessment. For this purpose, the following items must be examined in the assessment of single units.

(1) Nuclear fuel material

- Type of nuclear fuel material (chemical composition)
- Isotopic composition of nuclear fuel material
- Quantity of nuclear fuel material
- Concentration or density of nuclear fuel material
- Nonuniformity of nuclear fuel material (distribution of concentration)
- Heterogeneity of nuclear fuel material (dimensions and pitch)
- Kinds of coexisting materials (chemical composition)
- Concentration or density of coexisting materials

- Kind of neutron absorber (chemical composition)
- Concentration or density of neutron absorber
- Temperature

(2) Equipment and container

- Material type (chemical composition)
- Density
- Shapes and dimensions

(3) Others

- Kind of reflector (chemical composition)
- Density or concentration of reflector
- Shape and dimensions of reflector

4.3. Methods for Analytical Assessment of a Single Unit

Analytical assessments of a single unit can be broadly classified into those situations where data of known subcritical conditions are used and those where analyses are performed using codes. Both situations are described below.

4.3.1. Assessment with the Data on Known Subcritical Conditions

The data on subcritical conditions are commonly obtained for simple forms, such as a sphere, an infinite circular cylinder, and an infinite slab. To assess a single unit using these data, the form of the single unit must be simplified. The minimum estimated criticality and the minimum estimated lower limit criticality values of typical nuclear fuel materials are described in the Nuclear Criticality Safety Handbook - Data Collection. Some examples are shown in (1) through (3) below.

(1) Subcriticality conditions for mass, volume, shape, and dimensions

The basic factors for criticality safety include mass, volume, shape, and dimensions (the diameter of an infinite circular cylinder and the thickness of an infinite slab). The minimum estimated criticality values and the estimated lower limit criticality values of these criticality factors are shown in Tables 4.1 through 4.5.⁽²⁾ These values were obtained using the estimated criticality multiplication factor, the estimated lower limit criticality multiplication factor, both determined by analyzing the results of many benchmark calculations, and the JACS Code System⁽³⁾ All object systems assessed have a sufficient water reflector.

(2) Subcritical conditions for an infinite system

One criticality factor concerning the composition, state, or both, of nuclear fuel may satisfy a specified condition that precludes criticality irrespective of other criticality factor, in spite of the dimensions of the system being infinite. The estimated criticality values and the estimated lower limit criticality values of typical criticality factors concerning uranium enrichment, nuclear fuel material concentration, and moderation power are shown in Tables 4.6 through 4.9 ⁽⁴⁾. These values were obtained using the estimated criticality multiplication factor for infinite systems and the estimated lower limit criticality multiplication factor, both determined by analyzing the results of many benchmark calculations, and the JACS Code System. Consequently these data can be applied to any single units of a homogeneous in an arbitrary shape. Fig. 4.2 shows the conditions for a homogeneous U-H₂O system that becomes critical. This figure indicates the following:

- a) The system is subcritical at 11.6 g ²³⁵U/L or less.
- b) The system becomes critical at lower concentrations as the enrichment becomes higher.
- c) The system never becomes critical at any concentration if the ²³⁵U enrichment is less than 0.88 wt%.
- d) When the ²³⁵U enrichment is less than 5 wt%, the system never becomes critical if H/U is less than 0.1.

(3) Subcritical conditions for light water reactor spent fuel

Considering the composition changes of uranium and plutonium associated with fuel burnup, one observes that the subcritical mass (mass equivalent to k_{eff} of 0.98) increases with burnup. Additional consideration of the formation of FP nuclides leads one to observe that the subcritical mass becomes even larger than the subcritical mass calculated ignoring FP nuclides. To evaluate the criticality of spent fuel, three typical spent fuel handling processes -- transportation, dissolution, and Pu purification -- were selected and the subcritical mass (a fuel mass equivalent to $k_{\text{eff}} = 0.98$) of fuel typically handled in these processes, i.e., (a) heterogeneous UO₂ pellets-H₂O, (b) homogeneous UO₂(NO₃)₂-Pu(NO₃)₄ water solution, and (c) homogeneous Pu(NO₃)₄ water solution, were calculated. Two burnup fuel compositions were considered. One composition only considered uranium depletion and plutonium formation, the other also accounted for the formation of FP nuclides. In this analysis, the isotopic composition of uranium and plutonium at each burnup level shown in Table 3.7 were used. The ORIGEN2 code was used to estimate the amount of FP formation. Fuel parameters of each fuel used for the calculations are shown in Table 4.10. As the results in Fig. 4.3 through Fig. 4.5 show, the subcritical mass increased with increased burnup for all the three studied fuels. These figures also show that the subcritical masses including FP formation in the calculations were larger than the masses including only the isotopic composition of uranium and plutonium.

4.3.2. Analysis Using Simple Calculation Codes

The output data with simple calculation codes, such as SIMCRI⁽⁵⁾, that use a space one-point approximation model are normally the multiplication factor and the critical buckling factor in an infinite system. Subsequently, the fuel system may be judged as subcritical if the geometrical buckling of the analytical object calculated from the following equation is larger with sufficient margin than the critical buckling calculated using a simple calculation code.

Infinite slab with thickness T:

$$B^2 = \left\{ \pi / (T + 2d) \right\}^2 \quad (4.1)$$

Infinite circular cylinder with radius R:

$$B^2 = \left\{ 2.405 / (R + d) \right\}^2 \quad (4.2)$$

Sphere with radius r:

$$B^2 = \left\{ \pi / (r + d) \right\}^2 \quad (4.3)$$

When a reflector is provided, the extrapolation distance d varies with the material, shape, and dimensions of the fuel and the reflector material. When a reflector having sufficient thickness around the fuel area is provided, d can be represented by the following formula:

$$d = 2D \cdot (1 + \beta) / (1 - \beta) \quad (4.4)$$

where D represents the diffusion factor of the fuel area and β is the albedo value. β can be determined by the following equations according to the shape of the fuel.

Infinite slab:

$$\beta = \frac{1 - 2\kappa_R D_R}{1 + 2\kappa_R D_R} \quad (4.5)$$

Infinite circular cylinder with radius R:

$$\beta = \frac{1 - 2\kappa_R D_R \frac{K_1(\kappa_R \cdot R)}{K_0(\kappa_R \cdot R)}}{1 + 2\kappa_R D_R \frac{K_1(\kappa_R \cdot R)}{K_0(\kappa_R \cdot R)}} \quad (4.6)$$

Sphere with radius r :

$$\beta = \frac{1 - 2D_R(\kappa_R + 1/r)}{1 + 2D_R(\kappa_R + 1/r)} \quad (4.7)$$

where

D_R : Diffusion constant of the reflector area ,

κ_R : Inverse of diffusion length of the reflector area ,

K_0 : Modified 0th order Bessel function of the second kind , and

K_1 : Modified 1st order Bessel function of the second kind .

For water reflection, the analytical results with $D_R = 0.47$ cm and $\kappa_R = 0.30$ cm⁻¹ show satisfactory agreement with the results of detailed calculations. In Eqs. (4.1) to (4.3), d is the extrapolation distance in the vacuum border condition without a reflector. It can be obtained by substituting $\beta = 0$ into Eq. (4.4).

The geometric buckling, B_g , of a regular polygon is inversely proportional to the square of the radius, R_c , of a circle which circumscribes the effective geometry including the extrapolation distance. The geometric buckling is expressed by the form $B_g^2 = (a_N/R_c)^2$, where a_N is a constant depending on the number of sides of a regular polygon.^{(6),(7)} Values for a_N are shown in Table 4.11.

4.3.3. Analysis Using Detailed Calculation Codes^{(8),(9),(10)}

The effective neutron multiplication factor, k_{eff} , an eigenvalue of a system, is normally obtained by solving a neutron transport equation with detailed calculation codes. If k_{eff} is less than the estimated lower limit criticality effective multiplication factor, k_L , the system can be considered subcritical.

One- and two-dimensional transport codes using the diffusion approximation and the S_N method are widely used for the calculation of k_{eff} of single unit systems having simple shapes. A three-dimensional transport code by the Monte Carlo method is used to analyze complex systems such as storage tanks and columns having complex shapes and complex piping systems in spent fuel reprocessing facilities. When a system is simple, a calculation code using two- or three-dimensional diffusion approximations may be used to minimize computer time. Examples of these detailed

calculation codes for criticality safety assessment are shown in Table 4.12 and 4.13.

To use these codes, libraries of nuclear constants are required. Some nuclear libraries are multigroup libraries that divide the neutron energy spectrum into multiple groups, other libraries are continuous energy libraries that consist of a collection of many energy points. These libraries are prepared by processing nuclear data files such as ENDF/B⁽¹¹⁾ and JENDL⁽¹²⁾. These are also called processed libraries and each processed library is prepared for specific code's format requirements.

Then, multigroup libraries are processed by a cross section processing program to incorporate resonance self-shielding and heterogeneous corrections, and to tailor the format of cross section to the next calculation code. In this way, a cross section data file to be inputted to each calculation code is prepared. A typical method of preparing an effective cross section file that accounts for the resonance self-shielding is by using a f-table. A general method used to correct it for the heterogeneous effect is by applying the Dancoff factor. One advantage of continuous energy libraries is that they do not require additional processing for resonance self-shielding nor heterogeneous effect corrections.

Before calculating the effective multiplication factor using a detailed calculation code, the accuracy of the multigroup constant libraries that will be used must be fully assessed because that accuracy has a direct influence on the accuracy of the effective multiplication factor.

The characteristics and cautions for using calculation codes are described below for each calculation theory.

(1) Diffusion approximation method

The diffusion approximation theory describes the average behavior of neutrons in a system; that is, it describes the macroscopic behavior of neutrons. In this theory, neutron density is treated as a statistically averaged value. When a calculation code utilizing diffusion approximation is to be used, the shape of the system normally must be represented by regular, orthogonal coordinates. Consequently, modeling must be done with great care.

With the diffusion approximation method, it is not possible to handle the neutron movement direction faithfully. The direction variable in the transport equation is eliminated. Instead, current J of the neutrons in the direction of the coordinate axis is introduced according to Fick's Diffusion Law as follows.

$$J_x = -D \cdot \frac{\partial \phi}{\partial x} \quad (4.8)$$

where D is the diffusion constant and ϕ is the neutron flux.

Fick's Diffusion Law is valid when the neutron absorption in the system is weak and most neutrons are scattered due to collisions with atomic nuclei. Namely, $\Sigma_a \ll \Sigma_s$ holds. Furthermore, Fick's Diffusion Law is valid in a region which is more than several times of the neutron mean free

path away from a studied system boundary or neutron source. Therefore, this method is not suitable for a system having small dimensions, a system containing voids, a system containing locally strong neutron sources, or a system unable to scatter neutrons easily. For this reason, diffusion approximation codes are often used for analyzing a very large system such as a total reactor core system. Where the diffusion approximation is not valid (i.e. around fuel elements), a correction given by the transport theory is required. This is called the transport correction.

Usually the diffusion approximation method is simpler than other calculational methods. Even for the systems with two- or three-dimensional complex shapes, the calculation can be made quickly and with high calculational accuracy if a calculational code adopting the finite element method or the finite difference method is used. To obtain the effective neutron multiplication factor with high accuracy, a calculational code based on strict theory must be used. However, the diffusion approximation calculation codes are widely used for quick estimates of neutron flux.

With the diffusion code, the neutron generation distribution corresponding to the maximum eigenvalue (effective multiplication factor of neutron) of the transport equation is obtained by a repetition method until it converges. The determination of convergence should particularly be noted. Convergence is determined by comparing the relative error between the previous and present values for repeated calculations. However, this error is not the difference from the true solutions. Consequently, if the required error range of eigenvalues is 10^{-2} , the standard value to determine convergence in the calculation should be a value set one order of magnitude severer or 10^{-3} .

Also, when using a code in which a parameter is discretized as in the diffusion method, the neutron multiplication factor obtained varies according to the number selected for spatial mesh points and neutron energy groups. These variations are often called the mesh effect and the group effect, respectively, and corrections for these effects may be made.

(2) Discrete ordinate (S_N) method

With the S_N method, it is possible to obtain directly the numerical solution of the transport equation. As with the diffusion approximation method, the S_N method handles neutron density as a statistical average value. In practice, the system must have a regular shape and it must be computed with an orthogonal coordinate system. In calculating systems having complex shapes in two or three dimensions, the computer storage and calculation time requirements are colossal. As a result, only one-dimensional slabs, circular cylinders, and spheres are usually calculated.

The S_N method can faithfully handle the movement direction of neutrons. With this method, the space domain is partitioned into meshes and the domain of direction variables (angle) is divided into several directions. The N in S_N represents the number of scattered azimuths. If N is set at a large value, directional division is very fine. As a result, calculation accuracy improves but the calculation time also becomes colossal and convergence is also aggravated. The same applies with the number of mesh divisions of the space domain. The merit of the S_N method is that it applies even to the systems that the diffusion approximation method cannot handle, including systems having small dimensions

and volume, systems having voids, and systems having locally strong neutron absorption or generating sources. A one-dimensional S_N code is frequently used for surveying the trend of criticality or surveying parameters because of its short calculation time.

With the S_N code as well as the diffusion code, the distribution of neutron generation corresponding to the maximum eigenvalue (effective multiplication factor of neutron) of the transport equation is obtained by a repetition method until it converges. The determination of convergence should particularly be noted. Convergence is determined by comparing the relative error between the value of the previous calculation and the value of the most recent calculation for repeated calculations. However, this error is not the difference from the true solutions. Consequently, if the required error range of eigenvalues is 10^{-2} , the standard value to determine convergence in the calculation should be a value set one order of magnitude severer or 10^{-3} .

The standard value to determine convergence of neutron flux distribution should be a value less than the standard value to determine convergence of the eigenvalue.

When using an S_N code, attention must be paid to the ray effect. The ray effect is a phenomenon resulting in physically impossible oscillations of the neutron flux in a two or three-dimensional S_N code, caused by neutrons moving along the lines defined by discretized angles only. Attention is required, since this ray effect is more significant with a small scattering cross section and a large absorption cross section. Additionally, attention must be paid to the mesh and group effect, as explained in the diffusion approximation method.

(3) Collision probability method

In the diffusion approximation method and the S_N method above, differential equations with discretized variables are solved to obtain a neutron flux distribution. These methods can give accurate solutions with a small number of mesh points when the spatial change of the neutron flux is gradual. When the mean free path of neutrons is shorter than the size of the object system, the spatial changes of the neutron flux within the system is more complicated and a greater number of mesh points are required to obtain accurate solutions to the differential equations. In contrast, the integral transport equation is known to give an accurate solution using a small number of mesh points, since it does not rely on solving discretized differential equations. The kernel of the integral neutron transport equation is related to the probability that a neutron will suffer its next collision with an atomic nuclei after travelling a known distance. The integral diffusion equation, therefore, is solved by so-called the collision probability method. Due to the above-mentioned feature, this method is often used to obtain an average effective cross section of a small region of a large heterogeneous system such as fuel rods in a light water fuel assembly. A widely adopted practice is that based on the average effective cross section of the small region obtained in this manner, the neutron flux distribution or eigenvalue of a larger system is obtained by using a diffusion approximation, S_N , or Monte Carlo code, which is described below.

(4) Monte Carlo method

In the safety assessment of criticality of nuclear fuel installations, calculations by the Monte Carlo method have two advantages over other methods. One is this method's superior ability to handle fuel shapes and the other is its ability to handle nuclear cross-section data. The calculation of fuel solution reaction tanks in spent fuel reprocessing facilities that have complex shapes is difficult with one- or two-dimensional S_N codes. On the other hand, a Monte Carlo code has package input options for general forms and combinations of these options. In addition, more complex shapes can be applied to using a general geometry option; therefore, this method has high adaptability to inputting of fuel shapes. With a Monte Carlo code, the behavior of neutrons in the system is traced one by one. Thus, the value of nuclear cross-section data used is the probability of a reaction when the neutrons collide with atomic nuclei. As a result, it is applied correctly according to the theory of probability. Consequently, it is possible to use the most basic values of nuclear data corresponding to neutron energies, reaction types, and the kinds of nuclei. A typical multi-group Monte Carlo code, such as KENO-Va, divides the neutron energy spectrum into a finite number of ranges and assumes that the cross section remains constant within each energy range, considering limited computer memory capacity. However, computers with a larger memory capacity and a faster speed available today allow some Monte Carlo codes use a continuous neutron energy spectrum instead of a multi-group one. These codes are called continuous energy Monte Carlo codes, and MVP is a typical example.

Monte Carlo code calculations are free of errors resulting from the use of differential calculus, because they dispense with the mesh divisions of space and direction, which are used by the S_N code. However, this is a statistical calculation process that employs a random number sampling method. Therefore, with a Monte Carlo code, the calculation result lies within the range of a statistical error. To minimize this statistical error, it is necessary to increase the number of neutron histories and thus the calculation time.

With a Monte Carlo code, the reaction process of each neutron is traced according to given probability data. The neutron number F that represents the number of neutrons that result from fission, the neutron number A that represents the number of neutrons that are absorbed and vanish, and the neutron number L that represents the number of neutrons lost as they leak from the system are used to calculate the effective multiplication factor, k_{eff} , using the following equation.

$$k_{\text{eff}} = F / (A + L) \quad (4.9)$$

In calculations using a Monte Carlo code, the spatial distribution of neutron generation requires special attention. Since the fundamental mode of neutron generation is not known at the start of the calculation, a flat distribution or cosine distribution is arbitrarily assumed and several hundreds to several thousands of neutrons per generation is normally generated according to the distribution. From the second generation onward, the place where fission occurred in the previous generation is

used. This is assumed as the distribution of neutron generation for this time calculation. Normally this iteration is repeated for 50 to 300 generations. As the generations advance, the distribution of neutron generation approaches the fundamental mode, a stable mode where the distribution changes little from one generation to the next. The distributions for the first several generations are very different from those in the fundamental mode. Consequently, when the statistically averaged k_{eff} of the system is to be calculated, the k_{eff} values for the first several generations are skipped. The procedure for selecting the neutron number, the number of calculated generations, the number of skipped generations, and the initial distribution of neutron generation must be examined for each calculation. The required accuracy and computing time must also be considered. However, it is necessary that the final average k_{eff} agrees with the distribution of the neutron generation in the fundamental mode and that the standard deviation, σ , is limited to an appropriate value.

Most errors that result from the use of a Monte Carlo code are attributable to calculation of the final k_{eff} of the system by summing the calculation results obtained before the distribution of neutron generation reaches the fundamental mode. This will cause errors in the value of the generated average of k_{eff} that is calculated for each generation. It will also cause errors in the estimate of the standard deviation of the calculated value of k_{eff} . There is no definite standard established for determining the number of generations skipped initially. As the distribution of neutron generation approaches the fundamental mode, the variation of distribution for each generation becomes smaller. Thus, the variation of the calculated value of k_{eff} for each generation also should become smaller. Consequently, if the fluctuation of the calculated value of k_{eff} for each generation lies within the error range, the distribution of neutron generation may be understood to have approached the fundamental mode. The calculational result for that generation onward may be used for the calculation of the final statistic.

The above situation applies where the number of neutrons produced per generation is sufficiently large. However, a problem may occur because the calculation involves too few generations (several hundred to several thousands). For example, the distribution of neutrons produced for each generation is unstable; it may approach or may move away from the fundamental mode. In such an instance, a new calculation should be started after increasing the number of neutrons produced per generation. Furthermore, when the number of neutrons per generation is small, a large statistical error in the effective multiplication factor for each generation will result. When coupled with the necessity to increase the number of skipped generations, this will increase the standard deviation of the effective multiplication factor obtained by averaging the remaining generations. When using a Monte Carlo code, therefore, if the number of histories (number of neutrons per generation multiplied by the number of generations) or the calculation time is limited, the user should reduce the number of generations and select a larger number of neutrons per generation.

To make the number of generations as small as possible, the distribution of the initial neutron generation should be made as close to the fundamental mode as possible. Theoretically, when the

calculation involves a simple shape, the fundamental mode distribution can be anticipated. Even if a flat distribution is inputted, it will converge into the fundamental mode after a few generation times. Thus, few problems will occur. In the systems having complex shapes, however, many generations are needed to arrive at the fundamental mode. In this situation, a method may be used where the distribution is estimated in advance using a diffusion approximation or S_N method code with a simplified model. This estimated distribution is inputted into the Monte Carlo code calculation.

4.4. Method for Analytical Assessment of Multiple Units

Analytical assessment of multiple units can be broadly classified into two cases; the case where the mutual interaction of neutrons between single units is assessed using known isolation data, and the case where the analysis is performed with simple calculation methods, such as the solid-angle method, a simple calculation code, or a detailed calculation code. Each method is described in the following sections.

4.4.1. Assessment by Isolation Data

Even where subcriticality is confirmed in a single unit, the unit may become critical due to mutual interaction of neutrons when two or more single units are grouped. Two methods are available to prevent criticality attributable to mutual interaction of neutrons. One method is to keep the distance between single units larger than a certain value (that is, keep the solid angle smaller than a certain value). The other method is to keep the mutual interaction effect at a low level by providing a neutron isolator between the single units. The distance between single units where mutual interaction can be neglected is called adequate isolation distance and the thickness of a neutron isolator where mutual interaction can be neglected is called adequate isolation thickness. Criticality safety can be assessed from these isolation data.

(1) Thickness of neutron isolator

Isolation thickness varies with the material of the isolator, the composition, shape, and dimensions of the fissile material of the single unit, and the distance from the single unit to the isolator. Determination of values that apply uniformly to all situations is not practical because these values would usually be unnecessarily conservative. Therefore, the values of isolation thickness should be decided for each object after considering the applicable conditions.

The Reflector Factor (RF), defined by Eq. (4.10), is calculated, case-by-case, for each object. The isolation thickness is obtained by comparing the calculated RF with a suitable reactivity cutoff value.

$$RF = \left| (k(R) - k_s) / k_s \right| \quad (4.10)$$

where

$k(R)$: Effective neutron multiplication factor of the system where isolator thickness is R and the nuclear fuel systems on the opposite sides of the isolator are assumed to be the same, and

k_s : $=k(R \rightarrow \infty)$, which is the effective neutron multiplication factor where the object nuclear fuel system is surrounded by an infinitely thick isolator.

The nuclear fuel system may be either a single unit or a multiple unit. Two calculational examples using this method are described below.

- (a) Fig. 4.6 shows that various water isolation thicknesses can be obtained due to differences in the configuration of fuel assemblies when the thickness of the water isolator between fuel assemblies varies. These data are for two configurations of BWR type fuel assemblies, each with an enrichment of 2.6 wt%. The data were obtained using the method described above. The RF of a 2×1 configuration system is identified with circle symbols. This was obtained experimentally by the critical water level method. When RF approaches 10^{-3} , it decreases linearly on the logarithmic scale as the water isolator thickness increases.⁽²⁸⁾ The RF of a 3×3 configuration system is identified with triangle symbols. This was obtained using the KENO-IV Code. The two systems have a common trend; the RF decreases with an increase in the thickness of the water isolator.⁽²⁹⁾ However, when the thickness of the water isolator is zero, the larger system, the 3×3 configuration, has a larger value than the smaller system, the 2×1 configuration system.
- (b) Examples of changes in RF versus the thickness of a concrete isolator and a water isolator are shown in Fig. 4.7.⁽²⁹⁾ This figure shows the results of detailed calculations for the changes in RF where the following three types of infinite slab fuel are isolated with concrete or water.
- ^{235}U metal (thickness 2 cm)
 - Homogeneous $^{235}\text{U}\text{-H}_2\text{O}$ (uranium concentration: 0.1 gU/cm^3 , thickness: 7.5 cm)
 - Homogeneous $\text{U-H}_2\text{O}$ (^{235}U enrichment: 5 wt%, uranium concentration: 2 gU/cm^3 , thickness: 12 cm)

The changes in RF obtained from calculations with simple formulas are also shown. These changes will be discussed later. As is evident from Fig. 4.7, despite the combination of fuel and isolator, RF decreases exponentially with an increase in isolator thickness. Except very thin isolator, the gradient of this decrease is approximately the inverse of the neutron migration length ($1/5.93\text{cm}^{-1}$). The gradient of RF in the very thin isolator cases decreases because of the increased direct leakage of the thermal neutrons from the fuel region. The RF decreases exponentially when the isolator thickness becomes larger than the neutron migration length.

Considering the above discussion, isolation thickness can be calculated using this simple formula.

$$RF = \{k(0) - k_s\} / k_s \cdot \exp\{-(R - M)/M\} \quad (4.11)$$

where

$k(0)$: $k(R \rightarrow 0)$, which is the effective multiplication factor of the system where the isolator thickness becomes zero and the nuclear fuel systems are in contact with each other,

R : Isolator thickness, and

M : Migration length of neutron in isolator.

This equation was derived on the assumption that RF decreases exponentially when the isolator thickness becomes larger than the migration length of neutrons in the isolator. As is evident from Fig. 4.7, the RF value obtained from this equation is larger than the RF value obtained from the detailed calculation.

The isolator thickness R , where RF given in Eq. (4.11), becomes equal to the reactivity cutoff value, is called the isolation thickness. If this thickness is expressed as l , the value of l can be given by the following equation.

$$l = M \cdot \left[1 + \ln \left\{ (k(0) - k_s) / (k_s \cdot \epsilon) \right\} \right], \quad (4.12)$$

where

ϵ : reactivity (discussed in a following paragraph).

When Eq. (4.12) is applied to a system consisting of many identical units, the neutron multiplication factor of an infinite array system, k_∞ , can be used for $k(0)$. In a system consisting of a few units, even if the isolation thickness becomes zero and the units contact each other, the system size is finite and has a constant buckling. Thus, the value of $k(0)$ of a finite system is smaller than that of k_∞ . This results in the difference of RF(0) values among the 2×1 system and the 3×3 system shown in Fig. 4.6.

If the k_{eff} of each unit is designed with some safety factor against the limit value k_{limit} , it is possible to maintain subcriticality even when the isolator thickness is reduced. Suppose that reactivity increased by ϵ due to the mutual interaction effect. The effective multiplication factor of the system will then become $(1+\epsilon) \cdot k_s$. Consequently, if the following equation is held, subcriticality can be maintained.

$$k_{\text{limit}} \geq (1 + \epsilon) \cdot k_s \quad (4.13)$$

From this equation, it is possible to calculate ϵ . Substituting ϵ into Eq. (4.12), the isolation thickness, l , is obtained.

The isolation thickness for water isolation for infinite slab fuel can be obtained using a simple formula. When the fuel is enclosed with an adequate water isolator, let the effective multiplication factor, k_s , be 0.8, the infinite multiplication factor, $k(0)$, be 1.2 and limit value $k_{\text{limit}}=0.94$. From Eq. (4.13), $\epsilon = 0.175$ is led as the limiting value; i.e., even if the 17.5% $\Delta k/k$ reactivity increases, subcriticality can be maintained. Then, substituting $\epsilon = 0.175$, $M = 5.93$, $k(0) = 1.2$, and $k_s = 0.8$ in Eq. (4.12), an isolation thickness of 12 cm is obtained.

The RF in a fuel system without a moderator has a trend different from Figs. 4.6 and 4.7 because the neutron energy spectra in the fuel region are significantly changed by the presence of a water isolator. Changes in RF versus the concrete isolator thickness in a BWR fuel assembly storage facility are shown in Fig. 4.8. As is evident from the figure, where the \circ symbols correspond to detailed calculations, as the concrete thickness increases from 0 to 10 cm, RF increases due to the thermalization of neutrons. In a system such as this, F/A or the ratio of neutron generation to neutron absorption in a fuel region of the system enclosed with an isolator having sufficient thickness, identified with the \bullet symbol, should be used for the value of $k(0)$.

(2) Isolation solid angle (isolation distance)

When multiple units are configured keeping the solid angle between single units less than a certain value (keeping the isolation distance larger than a certain value), mutual interaction decreases and the presence of one single unit has little influence on the effective multiplication factor of another single unit. The solid angle (distance) formed by the units in the state described above is called isolation solid angle (isolation distance).

The isolation solid angle is obtained by solving for RF using the following formula and comparing the RF value with the reactivity cutoff value.

$$RF \equiv \frac{\Delta k}{k}(\Omega) = \left| \frac{k_A(\Omega) - k_s}{k_s} \right| \quad (4.14)$$

where

$k_A(\Omega)$: Effective multiplication factor of multiple units where the fractional solid angle (the ratio to 4π) between single units is Ω , and

k_s : Effective multiplication factor of a single unit in the independent state.

When a second single unit having the same fuel composition and shape is placed near the first single unit, the change in reactivity of the system $\Delta k/k(\Omega)$ can be given by the following equation.⁽³¹⁾

$$\frac{\Delta k}{k}(\Omega) = \frac{\Omega}{1/M^2 B^2 + 1 - \Omega} \quad (4.15)$$

where

- M^2 : Migration area of a neutron in a single unit,
 B^2 : Geometrical buckling of a single unit, and
 Ω : Fractional solid angle between units.

Consequently the increment of reactivity of a system can be determined by the function of a solid angle with $M^2 B^2$ as a parameter.

The distance corresponding to the isolation solid angle of a simple form (isolation distance) can be calculated using the solid-angle formula of TID-7016 Rev. 2⁽³²⁾ as follows. The parameter η , which represents the distance, is made dimensionless and is defined for each form. Then, the solid angle is expressed by η as follows. For all cases, H represents the distance from the center of one single unit to the surface of the other single unit. (See Fig. 4.9.)

a) Sphere $\eta \equiv H/R$ (4.16)

R : radius

$$\Omega = (1/2) \cdot \left\{ 1 - \left(1 + 1/\eta^2 \right)^{-1/2} \right\} \quad (4.17)$$

b) Circular cylinder $\eta \equiv H/D$ (4.18)

D : Diameter and L : Height

$$\Omega = \left\{ 1/(4\pi\eta) \right\} \cdot \left\{ 1/4 + (\eta D/L)^2 \right\}^{-1/2} \quad (4.19)$$

c) Cuboid

$$\eta \equiv H/(a \cdot b)^{1/2} \quad (4.20)$$

a, b : Lengths of the two sides of the surfaces facing each other

$$\Omega = (1/\pi) \cdot \sin^{-1} \left[\left\{ 1 + 4\eta^2(b/a) \right\}^{-1/2} \cdot \left\{ 1 + 4\eta^2(a/b) \right\}^{-1/2} \right] \quad (4.21)$$

If the above equations are used, the three elements (reactivity increase, solid angle, and distance) can be interrelated. The results for a sphere, an infinite cylinder ($L \rightarrow \infty$), and a quadrangular prism are shown in Fig. 4.10. As is evident from the figure, for the unit where $M^2 B^2 = 1$,

the fractional solid-angle for $3 \times 10^{-3} \Delta k/k$ is 0.6%. This corresponds to 0.075 sr (steradian). The distances corresponding to this solid angle are 6.4, 26, and 3.6 as the η value for a sphere, an infinite cylinder, and a quadrangular prism, respectively.

4.4.2. Analysis by a Simple Method such as the Solid-Angle Method

The simple methods for assessment of mutual interaction can be broadly classified into the following two methods, each with its own characteristics.⁽³¹⁾

- (i) The solid-angle method and albedo method are used for calculating the mutual interaction between units in an array system. These methods are used effectively when a configuration has a few units and the methods do not depend upon the types of units (triangular lattice and square lattice).
- (ii) The density analog method, the NB_N^2 method, the surface density method, and the hyperbola method are semiexperimental methods. These methods are effectively used when a configuration has many units.

The density analog method and the surface density method are simple to use, but the supporting calculations are complex. The solid-angle method is the most practical method and has a wide range of applications. This method is designed to limit the solid angle or distance between single units by comparing the total solid angle formed by a single unit with the limiting solid angle. Solid-angle methods are described in TID-7016 Rev. 1⁽³³⁾ and CEA-R3114.⁽³⁴⁾ JAERI has also developed a solid-angle method.⁽³⁵⁾ These methods all can be used but attention must be directed to the respective application ranges.

The solid-angle method that JAERI has developed is described below. According to the theory of the solid-angle method, the following equations can be described as a total solid-angle limiting inequality concerning a single unit i of a system having no reflector.

$$\Omega_i < \frac{1}{f_i} \left(1.0 - \frac{k_i}{k_{\text{limit}}} \right) \quad (4.22)$$

$$f_i = \frac{1}{\sum_{j \neq i} \Omega_{ij}} \cdot \sum_{j \neq i} \left(1.0 - \frac{k_j}{k_{\infty j}} \right) \Omega_{ij} \quad (4.23)$$

$$\Omega_i = \sum_{j \neq i} \Omega_{ji} \quad (4.24)$$

where

- k_i : Effective multiplication factor of single unit i under the bare condition,
- k_{limit} : Limit effective multiplication factor,
- k_j : Effective multiplication factor of single unit j present around single unit i under the

bare condition,

- $k_{\infty j}$: Infinite multiplication factor of single unit j ,
 Ω_{ji} : Fractional solid angle viewing single unit i from single unit j , and
 Ω_i : Sum of fractional solid angles when viewing single unit i from all other surrounding single units j (total fractional solid angle).

In these equations, f_i represents the solid-angle weighted average neutron leakage from single units j located around single unit i toward single unit i . The value of f_i becomes $1.0 - k_i / k_{\infty j}$ for a multiple unit system consisting of the same single units. The problems that may be encountered in applying the solid-angle method are described below.

- (1) The total solid angle, Ω_i , is calculated as a sum of the solid angles viewing the object single unit i from the other single units located around single unit i . For this purpose, the calculation may use the "point-plane model," which gives a result close to the precise formula. Parameters for a solid-angle calculation using a point-plane model are illustrated in Fig. 4.9.
- (2) The solid angle may be obtained by considering the shadow generated by the third single unit when the object single unit is viewed from single unit j . This is illustrated in Fig. 4.11.
- (3) The total solid-angle limit inequality may be derived by substituting $(1.0+k_i)/2$ for k_i in Eq. (4.22). This introduces the concept of semireflection when reflectors are provided around the system. To be more precise, the contribution of the reflection from the surface of the reflecting wall when considering the albedo may be added during the calculation of the total solid angle. This is shown in Fig. 4.12. For the albedo, the values of 0.56, 0.73, and 0.79 should be used for water, concrete, and SUS-304, respectively.
- (4) When a single unit is surrounded by a moderator material or by units having a large moderating effect, k_i and k_i should be obtained by calculating the neutron energy spectra in the fuel region by assuming sufficient reflector thickness. This will include the influence of the reflector on the neutron energy spectrum.
- (5) When an inundated state is considered possible, the face-to-face distance between single units should be set to more than the isolation distance so that criticality safety can be maintained even during submersion in water.
- (6) The method described in TID-7016 Rev. 2, the method proposed by CEA-R3114, or the method proposed by JAERI seem to be very similar. All are based on Eq. (4.22) for the determination of the solid angle. However, with TID-7016 Rev. 2, $f_i = 4\pi/19$ and $k_{limit} = 19/20$ (for many application ranges) and with CEA-R3114, $f_i = 1.0$ and $k_{limit} = 1.0$.

When using these solid-angle methods, the calculation of Ω_i requires great care. Using the method described in TID-7016 Rev. 2, Ω_i is calculated as the sum of the solid angles viewing the surrounding other single units j from the object single unit i . When using the method proposed by

JAERI, Ω_i is calculated as the sum of the solid angles viewing the object single unit i from the surrounding other single units j .

The following drawbacks occur due to the differences in the methods of calculating Ω_i . When single unit j has a large volume and if k_{∞} is small, although originally the influence of j on single unit i should be small, Ω_i becomes a large value when using TID-7016 Rev. 2. Thus, the spacing between i and j must be made larger. Therefore, when the TID-7016 Rev. 2 method is used, an unnecessarily large safety margin is provided. Conversely, when single unit j having a small volume is near single unit i , large values of Ω_i will result with the CEA-R3114 and JAERI methods although originally the influence of j on single unit i should be small. The results provide unnecessarily large safety margins.

Any of the three solid-angle methods may be used; criticality safety will be assured if the method satisfies the limit condition of Eq. (4.22).

4.4.3. Analysis by Simple Calculation Codes

Simple calculation codes are normally used for the calculation of the effective multiplication factor of neutrons in a multiple unit system using the neutron balance equation. This equation is derived on the assumption that the neutron migration probability between single units is proportional to the solid angle obtained by using the effective multiplication factor k_{eff} and the infinite multiplication factor k_{∞} of each single unit. In using simple calculation codes, great care must be directed to the application ranges. Consequently, the application range of the simple calculation code concerned should be clarified in advance through a comparison with a calculation using a detailed calculational code.

With the simple calculation code MUTUAL,^{(35),(38)} developed by JAERI for the mutual interaction effect of neutrons, k_{eff} of a multiple unit is calculated using the effective multiplication factor k_{effB} under bare conditions of a single unit and the infinite multiplication factor k_{∞} (or M^2B^2). To clarify the application range of the MUTUAL code, the calculated results of this code and the detailed calculation code KENO-IV, a Monte Carlo code, are compared in Fig. 4.13. As shown in this example, when the calculated results obtained from MUTUAL are compared with KENO-IV, the application range of MUTUAL will be decided as the range where k_{eff} is calculated as a large value. Thus, a calculation with MUTUAL always provides a larger safety margin than a calculation with KENO-IV. In determining the subcriticality of multiple units using k_{eff} calculated with MUTUAL, therefore, the same limit value (k_{limit}) used for KENO-IV should be utilized.

MUTUAL is applicable to an array system of low-enrichment uranium fuel units that satisfy the following conditions

- (1) MUTUAL is applicable to a system without a reflector, where $I' \leq 0.25$ and $\omega \leq 0.06$ or $d \geq 30$ cm (I' : increase rate of neutron multiplication factor due to neutron interaction effect, ω : unit solid angle fraction, d : unit isolation distance).
- (2) MUTUAL is applicable to a system having four surfaces are surrounded by concrete

reflectors by setting the albedo value at 1, but the current version of MUTUAL cannot be applied to an array system having all six surfaces are surrounded by concrete reflectors.

To verify the applicability of MUTUAL to a system without reflectors, calculations were conducted by changing the fuel moderation and the array configuration. Calculations were performed for two types of single unit $\text{UO}_2\text{-H}_2\text{O}$ fuels (moderated system and non-moderated system) having an enrichment of 5 wt%. Fuel composition, dimension, and other parameters for each single unit fuel are shown in Table 4.14. Two types of arrays, $2 \times 1 \times 1$ and $3 \times 3 \times 1$, were used and calculations were conducted by changing the unit isolation distance. (For $\text{UO}_2\text{-H}_2\text{O}$ moderated system, $4 \times 4 \times 1$ and $5 \times 5 \times 1$ array configurations were also considered.) An example result for a moderated system with a $4 \times 4 \times 1$ array is given in Fig. 4.14. For this $4 \times 4 \times 1$ array, when the unit isolation distance became less than or equal to 20 cm (solid angle fraction became greater than or equal to 0.065), the MUTUAL calculated effective multiplication factor was smaller than that the KENO-IV value. Similar calculations were conducted for other $\text{UO}_2\text{-H}_2\text{O}$ moderated array configurations selected. Based on the results, the limits of application for the MUTUAL code for $\text{UO}_2\text{-H}_2\text{O}$ moderated units were found to be 20 cm in regard to the unit isolation distance, 0.065 for the solid angle fraction, and 0.25 for the increase rate of effective multiplication factor.

For $\text{UO}_2\text{-H}_2\text{O}$ non-moderated units with $2 \times 1 \times 1$ and $3 \times 3 \times 1$ array configurations, the effective multiplication factor calculated by MUTUAL never became smaller than the KENO-IV value, even under such conditions as an unit isolation distance of 10 cm, a solid angle fraction of 0.096, and an increase rate of the effective multiplication factor of 0.15.

In addition, calculations were conducted for $\text{UO}_2\text{-H}_2\text{O}$ moderated fuel units having an enrichment of 5 wt% by changing unit dimensions and array configurations. For all cases studied, the maximum value of unit isolation distance was 20 cm, and the minimum value of solid angle fraction was 0.061 where the effective multiplication factor calculated by MUTUAL never became smaller than the value calculated by KENO-IV.

The applicability of MUTUAL to a system having concrete reflectors was evaluated. Cases examined here include two types of fuel units, $\text{UO}_2\text{-H}_2\text{O}$ moderated system and $\text{UO}_2\text{-H}_2\text{O}$ non-moderated system; two array configurations, single unit and a $3 \times 3 \times 1$ array (unit isolation distance 30 cm); two reflector conditions, all six surfaces surrounded by reflectors and four surfaces surrounded by reflectors (top, bottom, and two adjacent side surfaces); and two distances to the concrete walls, 50 cm and 100 cm. For the MUTUAL code calculations, the albedo value was set to 0.73 for all concrete reflectors.

Table 4.15 shows the result of cases in which all six surfaces were surrounded by reflectors. For both the $\text{UO}_2\text{-H}_2\text{O}$ moderated system and $\text{UO}_2\text{-H}_2\text{O}$ non-moderated system, the effective multiplication factor calculated by MUTUAL was smaller than the KENO-IV value. In contrast, for cases where there are only four surface reflectors (top, bottom, and two adjacent side surfaces) the effective multiplication factor calculated by MUTUAL was larger than the KENO-IV value

(excluding an case of a single unit of $\text{UO}_2\text{-H}_2\text{O}$ non-moderated system with a distance to reflector of 50 cm). These results are tabulated in Table 4.16.

The effects of changes in albedo value input into MUTUAL were also examined with respect to reflected systems. Bounding cases were evaluated for a concrete reflector albedo value equal to 1.0, rather than the generally assumed value of 0.73. These results are summarized in Tables 4.15 and 4.16. In cases where four surfaces are surrounded by concrete reflectors (albedo = 1.0) the effective multiplication factor calculated by MUTUAL was larger than the KENO-IV value by no more than $0.037\Delta k$. In contrast, in cases where all six surfaces are surrounded by concrete reflectors, the effective multiplication factor calculated at an albedo value of 1.0 was larger than that of 0.73 by as much as $0.02\Delta k$, but still smaller than the KENO-IV value in all six surface cases.

The above mentioned calculations for concrete reflected systems assumed that fuel array units are positioned inside a structure or a cell. In cases where the structure or cell is small and the distance from a fuel unit to the walls are small for all surfaces, the application of the current version of MUTUAL is considered difficult, whereas in cases where a structure or cell is large, the distance from a fuel unit to two walls is sufficiently large, and the distance to the remaining two walls is at least 50 cm, MUTUAL can be applied by adopting an albedo value of 1.0.

4.4.4. Auxiliary Calculations for Simple Calculation Methods such as the Solid-Angle Method and for Simple Calculation Codes

As described above, when making an assessment of the criticality safety of multiple units using a simple calculation method, such as the solid-angle method or a simple calculation code based on the solid-angle method, preparatory calculations are necessary. These include calculation of the effective multiplication factor k_{eff} , infinite multiplication factor k_{∞} , migration area M^2 , and geometrical buckling B^2 under the bare conditions of each of the single units. To obtain these quantities, the methods described below are recommended.⁽³⁹⁾

(1) Method by means of criticality condition data

The effective multiplication factor of a standard form single unit with a sufficiently thick reflector, k_{effR} , is generally given. The effective multiplication factor in bare conditions, k_{effB} , is obtained using the following relations.

For an infinite slab with thickness T:

$$k_{\text{effB}} = k_{\text{effR}} \cdot \frac{1 + M^2 \left(\frac{\pi}{T + 14.2D} \right)^2}{1 + M^2 \left(\frac{\pi}{T + 4D} \right)^2} \quad (4.25)$$

$$M^2 \cdot B^2 = M^2 \cdot \left\{ \pi / (T + 14.2 \cdot D) \right\}^2 \quad (4.26)$$

$$k_{\infty} = (1 + M^2 \cdot B^2) \cdot k_{\text{effR}} \quad (4.27)$$

For an infinite circular cylinder with radius R:

$$k_{\text{effB}} = k_{\text{effR}} \cdot \frac{1 + M^2 \left(\frac{2.405}{R + 7.1D} \right)^2}{1 + M^2 \left(\frac{2.405}{R + 2D} \right)^2} \quad (4.28)$$

$$M^2 \cdot B^2 = M^2 \cdot \left\{ 2.405 / (R + 7.1 \cdot D) \right\}^2 \quad (4.29)$$

$$k_{\infty} = (1 + M^2 \cdot B^2) \cdot k_{\text{effR}} \quad (4.30)$$

For a sphere with radius r:

$$k_{\text{effB}} = k_{\text{effR}} \cdot \frac{1 + M^2 \left(\frac{\pi}{r + 7.1D} \right)^2}{1 + M^2 \left(\frac{\pi}{r + 2D} \right)^2} \quad (4.31)$$

$$M^2 \cdot B^2 = M^2 \cdot \left\{ \pi / (r + 7.1 \cdot D) \right\}^2 \quad (4.32)$$

$$k_{\infty} = (1 + M^2 \cdot B^2) \cdot k_{\text{effR}} \quad (4.33)$$

where M^2 and D are obtained from a chart or simple calculation code given in the chapter 3 of "Nuclear Criticality Safety Handbook - Data Collection." The ratios of $k_{\text{effB}}/k_{\text{effR}}$ for an infinite slab and an infinite circular cylinder are shown respectively in Figs. 4.15 and 4.16.

(2) Method by means of charts or simple calculation codes

The values of k_{∞} , M^2 , B^2 , and D are given by charts in the Nuclear Criticality Safety

$$k_{\text{effR}} = k_{\infty} / (1 + M^2 \cdot B^2) \quad (4.34)$$

$$k_{\text{effB}} = k_{\text{effR}} \cdot \frac{1 + M^2 B_c^2}{1 + M^2 \left(\frac{\pi}{T + 4D} \right)^2} \quad (4.35)$$

For infinite slab with thickness T:

$$T = \pi / B - 14.2 \cdot D \quad (4.36)$$

For an infinite circular cylinder with radius R:

$$R = 2.405/B - 7.1 \cdot D \quad (4.37)$$

For a sphere with radius r:

$$r = \pi/B - 7.1 \cdot D \quad (4.38)$$

(3) Method by neutron transport calculation code

Since k_{∞} , k_{effR} , and k_{effB} are given with neutron transport calculation codes for the shape of a single unit, other quantities are obtained from the following equation.

$$M^2 B^2 = k_{\infty} / k_{\text{effR}} - 1.0 \quad (4.39)$$

When a Monte Carlo code (for example, KENO-IV) is used, the number of fission neutrons generated divided by the number of absorbed neutrons in the fuel region may be used as k_{∞} .

4.4.5. Analysis by Detailed Calculational Codes

To obtain the multiplication factor of neutrons in a multiple unit system, simple calculation methods including the solid-angle method or simple calculation codes based on that concept are used. However, owing to the advancement of computer technology, the use of detailed calculation codes is now realistic. Analysis of a system having multiple installations or pieces of equipment requires the use of a Monte Carlo code method, such as KENO-IV.

With the Monte Carlo code KENO-IV, the basic shapes including a sphere, circular cylinder, and rectangular parallelepiped are available in calculational packages. This allows easy definition of box configurations consisting of combinations of these shapes. Furthermore, with a Monte Carlo code for multibox types or combination types, configurations of boxes having varied spacing or configurations of misaligned boxes can be analyzed conveniently.

If a Monte Carlo code is to be used for a criticality analysis of a multiple unit system, attention must be directed to the following points.

- (i) A large enough number of neutron histories must be traced if the interactions of neutrons between single units are calculated.
- (ii) The distribution of neutron formation must conform to the fundamental mode.

Specifically, for the system where the neutrons interactions are weak due to the large distances between single units, strict attention must be paid to the items mentioned above. For a system having

a weak mutual interaction, a Monte Carlo code calculation may have difficulty progressing to the fundamental mode. In such a case, it is necessary to select the initial distribution of neutron generation after estimating the fundamental mode. For this method, since k_{effB} , the effective multiplication factor of neutrons for each of the single units is obtained in advance, the number of neutrons generated should be allocated according to the ratio of this value.

4.5. Methodology for Criticality Safety Evaluation of a System Divided into Compartments

The criticality safety evaluation of a system consisting of multiple chambers can be conducted as follows.

Set the perfect reflective condition in the center thickness of the walls surrounding a chamber, and calculate the neutron multiplication factor inside this chamber. The system is judged as subcritical when the neutron multiplication factor of every chamber does not exceed the maximum permissible multiplication factor. However, the wall thickness must be more than the migration length of neutrons. The migration length of neutrons for concrete walls is about 11 cm.

“The Basic Regulatory Guides for Licensing Safety Review of Nuclear Fuel Facilities” state that the criticality safety shall be secured by “criticality safety control of single unit” and “criticality safety control of multiple units”. This document also states that in the “criticality safety control of multiple units,” the neutron interaction among single units shall be considered. The interaction effect with the fuel in the adjacent chambers, however, is not generally considered. This is because each chamber was usually surrounded by sufficiently thick walls or was designed in such a way that the neutron interaction with other chambers does not take place. Therefore, unnecessarily thick walls were often used for the criticality safety. A thinner wall thickness may be used when the neutron multiplication factor of each chamber is sufficiently smaller than the critical state and when it is verified that the system will not reach criticality due to the neutron interaction effect among the different chambers. With the recent advance of computer technologies, we can evaluate the criticality safety of each chamber with the 3-D Monte Carlo method and can account for the neutron interaction among chambers. Considering what was explained above, we describe below a criticality safety evaluation method for a system having multiple chambers.

Consider a system having multiple chambers for storing nuclear fuel. Each chamber contains single units and each chamber is isolated from other chambers by concrete walls. It must be verified that the system will not achieve criticality due to the neutron interaction with surrounding chambers, taking into account a case where the concrete wall thickness is less than the “isolation thickness”.

The higher the neutron multiplication factor of the adjacent chamber, the severer the criticality conditions become. Therefore, in the criticality safety evaluation of all chambers, it must be assumed that the same nuclear fuel is stored in adjacent chambers and it must be verified that none of the chambers will reach criticality. To model this criticality condition, perfect reflection conditions are

set at the center of the wall thickness. When there is no adjacent chamber containing nuclear fuel, only the neutron reflective effect of structural materials, such as the wall, is considered. Using this method allows us to calculate criticality conditions according to the actual fuel storage conditions, eliminating the need to design the system in such a manner that any nuclear fuel can be stored in the adjacent chambers.

However, when the neutron energy spectrum of a chamber has a possibility of significantly changing due to the existence of an adjacent chamber, such a chamber cannot be treated as an independent chamber. One example of this case is where a chamber of powdered fuel with a low hydrogen content is situated next to a chamber of a low fuel concentration solution. Neutrons emitted from the powder fuel may be thermalized in the solution fuel and come back to the powder fuel. In this case, neutrons have a better chance of escaping the resonance absorption. As the result, the neutron multiplication factor of the system having both the powder and solution fuel chambers may be higher than that of the powder fuel chamber with a perfect reflector surrounding the chamber. When the concrete wall thickness between the both fuel chambers is larger than the neutron migration length of concrete (11 cm), the impact of the shift in the neutron energy spectrum can be ignored, since most of the returning thermalized neutrons are absorbed by the concrete wall.

References for Chapter 4

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Table 4.1 Minimum estimated criticality value and minimum estimated lower limit criticality value of a homogeneous $\text{UO}_2\text{-H}_2\text{O}$ with a water reflector

| Enrichment (wt%) | Mass (kgU) | | Sphere volume (L) | | Infinite circular cylinder diameter (cm) | | Infinite slab thickness (cm) | |
|---------------------|----------------------|-------------------------------------|----------------------|-------------------------------------|---|-------------------------------------|---------------------------------|-------------------------------------|
| | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value |
| 3 | 92.2 | 84.2 | 50.0 | 45.8 | 31.8 | 30.5 | 16.1 | 15.0 |
| 4 | 56.8 | 50.1 | 34.4 | 30.3 | 28.0 | 27.0 | 13.5 | 12.7 |
| 5 | 38.2 | 34.1 | 28.1 | 24.8 | 25.5 | 24.6 | 12.0 | 11.3 |
| 10 | 14.0 | 12.7 | 15.8 | 14.6 | 20.7 | 19.8 | 8.8 | 8.3 |
| 20 | 7.43 | 7.00 | 11.5 | 10.5 | 17.9 | 17.4 | 7.1 | 6.7 |

Table 4.2 Minimum estimated criticality value and minimum estimated lower limit criticality value of a Homogeneous $\text{ADU(II)}^{*1}\text{-H}_2\text{O}$ with a water reflector

| Enrichment (wt%) | Mass (kgU) | | Sphere volume (L) | | Infinite circular cylinder diameter (cm) | | Infinite slab thickness (cm) | |
|---------------------|----------------------|-------------------------------------|----------------------|-------------------------------------|---|-------------------------------------|---------------------------------|-------------------------------------|
| | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value |
| 3 | 112. | 102. | 62.2 | 55.1 | 35.1 | 32.8 | 17.5 | 16.6 |
| 4 | 63.0 | 54.1 | 38.7 | 36.0 | 29.4 | 28.1 | 14.1 | 13.8 |
| 5 | 41.4 | 37.7 | 30.1 | 27.9 | 26.7 | 25.8 | 12.7 | 12.0 |

*1 $\text{ADU(II)} : 3\text{UO}_2 \cdot \text{NH}_3 \cdot 5\text{H}_2\text{O}$ **Table 4.3 Minimum estimated criticality value and minimum estimated lower limit criticality value of a homogeneous UO_2F_2 aqueous solution with a water reflector**

| Enrichment (wt%) | Mass (kgU) | | Sphere volume (L) | | Infinite circular cylinder diameter (cm) | | Infinite slab thickness (cm) | |
|---------------------|----------------------|-------------------------------------|----------------------|-------------------------------------|---|-------------------------------------|---------------------------------|-------------------------------------|
| | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value |
| 3 | 102. | 91.5 | 68.0 | 58.0 | 35.0 | 33.8 | 18.1 | 17.0 |
| 4 | 58.1 | 50.7 | 44.0 | 40.5 | 30.4 | 28.9 | 15.0 | 14.2 |
| 5 | 39.4 | 34.6 | 33.8 | 30.7 | 27.3 | 26.3 | 13.2 | 12.6 |
| 10 | 13.6 | 11.9 | 18.1 | 16.8 | 22.0 | 21.3 | 9.8 | 9.3 |
| 20 | 5.60 | 4.97 | 12.0 | 11.0 | 18.7 | 18.1 | 7.6 | 7.3 |

Table 4.4 Minimum estimated criticality value and minimum estimated lower limit criticality value of a homogeneous $\text{UO}_2(\text{NO}_3)_2$ aqueous solution with a water reflector

| Enrichment (wt%) | Mass (kgU) | | Sphere volume (L) | | Infinite circular cylinder diameter (cm) | | Infinite slab thickness (cm) | |
|------------------|-------------------|-------------------------------|-------------------|-------------------------------|--|-------------------------------|------------------------------|-------------------------------|
| | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value | Criticality value | Lower limit criticality value |
| 3 | 470 | 369 | 393 | 329 | 67.4 | 61.5 | 37.6 | 34.1 |
| 4 | 164 | 124 | 160 | 129 | 47.9 | 44.8 | 25.9 | 24.1 |
| 5 | 82.2 | 70.0 | 88.7 | 74.3 | 38.0 | 36.3 | 20.2 | 19.1 |

Table 4.5 Minimum estimated criticality value and minimum estimated lower limit criticality value of a homogeneous $\text{Pu}(\text{NO}_3)_4$ aqueous solution with a water reflector

| | Minimum estimated criticality value | | | | | Minimum estimated lower limit criticality value | | | | |
|--|-------------------------------------|------|------|------|------|---|------|------|------|------|
| $^{240}\text{Pu}/^{239}\text{Pu}$ [wt%] | 0 | 5 | 10 | 20 | 30 | 0 | 5 | 10 | 20 | 30 |
| Infinite circular cylinder diameter (cm) | 16.2 | 18.7 | 20.1 | 22.2 | 24.3 | 15.5 | 17.6 | 18.8 | 20.9 | 22.8 |
| Infinite slab thickness (cm) | 5.97 | 7.29 | 8.30 | 9.93 | 11.3 | 5.49 | 6.67 | 7.59 | 9.03 | 10.3 |
| Sphere volume (L) | 8.24 | 11.3 | 14.1 | 18.7 | 24.1 | 7.31 | 10.1 | 12.3 | 16.2 | 20.6 |

Table 4.6 Estimated critical enrichment and estimated lower limit critical enrichment (^{235}U wt%) in an infinite system of homogeneous $\text{U-H}_2\text{O}$, $\text{UO}_2\text{-H}_2\text{O}$, and UO_2F_2 aqueous solutions

| | Estimated critical enrichment ^{*1} | Estimated lower limit critical enrichment ^{*2} |
|--|---|---|
| $\text{U-H}_2\text{O}$ | 0.90 | 0.88 |
| $\text{UO}_2\text{-H}_2\text{O}$ | 0.94 | 0.91 |
| UO_2F_2 aqueous solution | 0.98 | 0.95 |

*1 $k_\infty = 0.991$ *2 $k_\infty = 0.980$

Table 4.7 Estimated critical enrichment and estimated lower limit critical enrichment (^{235}U wt%) in an infinite system of homogeneous $\text{UO}_2(\text{NO}_3)_2$ aqueous solution

| | Estimated critical enrichment ^{*1} | Estimated lower limit critical enrichment ^{*2} |
|---|---|---|
| $\text{UO}_2(\text{NO}_3)_2$ aqueous solution | 1.95 | 1.90 |

*1 $k_{\infty} = 0.991$ *2 $k_{\infty} = 0.980$ **Table 4.8 Estimated critical Pu enrichment and estimated lower limit critical Pu enrichment (oxide weight ratio: $\text{PuO}_2/(\text{PuO}_2+\text{UO}_2)$ wt%)**

| | Estimated critical enrichment ^{*1} | Estimated lower limit critical enrichment ^{*2} |
|--|---|---|
| $^{239}\text{PuO}_2 \cdot \text{U}(\text{nat})\text{O}_2 \cdot \text{H}_2\text{O}$ | 0.14 | 0.11 |

*1 $k_{\infty} = 1.013$ *2 $k_{\infty} = 0.980$ **Table 4.9 Estimated critical concentration and estimated lower limit critical concentration in an infinite system for selected nuclear fuel materials**

| Chemical form | Estimated critical concentration | Estimated lower limit critical concentration |
|---|---------------------------------------|--|
| $^{235}\text{U}-\text{H}_2\text{O}$ | $11.8\text{g}^{235}\text{U}/\text{L}$ | $11.6\text{g}^{235}\text{U}/\text{L}$ |
| $^{235}\text{UO}_2-\text{H}_2\text{O}$ | $11.7\text{g}^{235}\text{U}/\text{L}$ | $11.5\text{g}^{235}\text{U}/\text{L}$ |
| $^{235}\text{UO}_2\text{F}_2$ aqueous solution | $12.2\text{g}^{235}\text{U}/\text{L}$ | $12.0\text{g}^{235}\text{U}/\text{L}$ |
| $^{235}\text{UO}_2(\text{NO}_3)_2$ aqueous solution | $12.5\text{g}^{235}\text{U}/\text{L}$ | $11.8\text{g}^{235}\text{U}/\text{L}$ |
| $^{239}\text{Pu}-\text{H}_2\text{O}$ | $7.4\text{g}^{239}\text{Pu}/\text{L}$ | $7.0\text{g}^{239}\text{Pu}/\text{L}$ |
| $^{239}\text{PuO}_2-\text{H}_2\text{O}$ | $7.5\text{g}^{239}\text{Pu}/\text{L}$ | $7.0\text{g}^{239}\text{Pu}/\text{L}$ |
| $^{239}\text{Pu}(\text{NO}_3)_4$ aqueous solution | $7.5\text{g}^{239}\text{Pu}/\text{L}$ | $7.0\text{g}^{239}\text{Pu}/\text{L}$ |

Table 4.10 Parameters used for calculating subcritical masses of burnup fuels

| UO ₂ pellet-H ₂ O (for transportation) | |
|--|-------------------------------|
| Pellet diameter | 0.7 cm |
| Pellet density | 95 wt% of theoretical density |
| Uranium enrichment | 5 wt% |
| Burnup | 0, 15, 30 GWd/t |
| UO ₂ (NO ₃) ₂ - Pu(NO ₃) ₄ solution (for dissolution) | |
| Uranium enrichment | 5 wt% |
| Free nitric acid concentration | 0 |
| (U+Pu) concentration | 500 - 1500 g(U+Pu)/ L |
| Burnup | 0, 15, 30 GWd/ t |
| Pu(NO ₃) ₄ solution (for Pu processing) | |
| Free nitric acid concentration | 0 |
| Pu concentration | 50 - 800 gPu/ L |
| Burnup | 0, 5, 15, 30, 50 GWd/ t |

Table 4.11 Geometric buckling of regular polygons

$$B_g^2 = (a_N/R_c)^2$$

| Geometry | Number of sides N | Geometric factor (constant) a_N |
|------------------|----------------------|--------------------------------------|
| Regular Triangle | 3 | 4.190 |
| Square | 4 | 3.142 (π) |
| Regular Pentagon | 5 | 2.821 |
| Regular Hexagon | 6 | 2.675 |
| Regular Octagon | 8 | 2.547 |
| Circle | ∞ | (2.405) |

Table 4.12 Examples of code systems and their classifications

| Code name | Nuclear data files from which a nuclear constant library to be used for a code is created (Name of pre-process libraries) | Name of post-process library | Code or method for resonance self-shielding and heterogeneous corrections | Code or method for obtaining cell averaged cross section | Code or method for calculating multiplication factor of a system | Remarks |
|-----------|---|---|---|--|--|--|
| SCALE4.3 | Hansen-Roach ENDF/B-IV ENDF/B-V | Nine Libraries: 218GROUPNDF4, 27GROUPNDF4, 44GROUPNDF5, etc, | BONAMI* ² NITAWL-II* ³ | XSDRNP | XSDRNP KENO V.a KENO-VI | CSAS series driver (program) runs an analysis by combining multiple codes. |
| JACS | ENDF/B-IV JENDL-3.1 JENDL-3.2 | MGCL-B4 MGCL-J3.1 MGCL-J3.2 | MAIL | ANISN | ANISN KENO IV MULTI-KENO | The concept of a driver is non-existent. A user performs calculations by connecting codes. |
| SRAC95 | ENDF/B-IV ENDF/B-V ENDF/B-VI JEF-2.2 JENDL-2 JENDL-3.1 JENDL-3.2 | No particular names. | PEACO (NR approximation)* ¹ (IR approximation)* ¹ | (Collision probability method)* ¹ ANISN TWOTRAN | (Collision probability method)* ¹ ANISN TWOTRAN CITATION | The concept of a driver is non-existent. One input file can control codes. |

*1 Names of calculation methods are enclosed in parentheses. (A calculation method when there is not an independent code, is denoted in this manner.)

*2 BONAMI is a code for resonance self-shielding and heterogeneous corrections by NR approximation.

*3 NITAWL-II is a code for spectrum calculations by an integral type treatment of Nordheim

Table 4.13 Examples of calculation codes and their classifications

| Method | Code name | Dimension | Energy group |
|-------------------------|------------|-----------|-------------------------------|
| Diffusion approximation | CITATION | 3-D | Multi-group |
| SN | ANISN | 1-D | Multi-group |
| | XSDRNPM | 1-D | Multi-group |
| | DOT | 2-D | Multi-group |
| | TWOTRAN | 2-D | Multi-group |
| Collision probability | RABTH | 1-D | Ultra-fine group (64194group) |
| Monte Carlo | KENO-IV | 3-D | Multi-group |
| | KENO V.a | 3-D | Multi-group |
| | KENO-VI | 3-D | Multi-group |
| | MULTI-KENO | 3-D | Multi-group |
| | VIM | 3-D | Continuous energy |
| | MCNP4B | 3-D | Continuous energy |
| | MVP | 3-D | Continuous energy |

Table 4.14 Specifications of single units used to verify the applicability of MUTUAL

| Unit type | A | B |
|---|-----------------------------------|-----------------------------------|
| Chemical formula | UO ₂ -H ₂ O | UO ₂ -H ₂ O |
| Enrichment [wt%] | 5.0 | 5.0 |
| Uranium concentration [gU/cm ³] | 2.0 | --- |
| UO ₂ bulk density [gUO ₂ /cm ³] | --- | 4.0 |
| Degree of moderation H/U | 10 | 0.5 |
| Height of circular cylinder [cm] | 100 | 100 |
| Diameter of circular cylinder [cm] | 23 | 100 |
| Calculated k _∞ by ANISN-JR | 1.472 | 1.058 |
| Calculated k _∞ by KENO-IV | 0.6808±0.0020 | 0.5972±0.0013 |
| A (neutrons absorption) | 0.4371 | 0.5428 |
| F (neutrons production) | 0.6808 | 0.5972 |
| F/A | 1.439 | 1.100 |

Table 4.15 Comparisons of KENO-IV and MUTUAL calculations
(cases where all six surfaces surrounded by reflectors)

| Unit type | Number of units | Unit isolation distance [cm] | Distance to reflector [cm] | KENO-IV calculations | | MUTUAL calculations Albedo 0.73 | | MUTUAL calculations Albedo 1.0 | | |
|-----------|-----------------|------------------------------|----------------------------|------------------------|----------|---------------------------------|---|--------------------------------|---|---|
| | | | | $k_{\text{eff}}^{(1)}$ | σ | $k_{\text{eff}}^{(2)}$ | $k_{\text{eff}}^{(2)} - k_{\text{eff}}^{(1)}$ | $k_{\text{eff}}^{(3)}$ | $k_{\text{eff}}^{(3)} - k_{\text{eff}}^{(1)}$ | $k_{\text{eff}}^{(3)} - k_{\text{eff}}^{(2)}$ |
| A | 1 | - | 100 | 0.6886 | 0.0018 | 0.6851 | -0.0035 | 0.6866 | -0.0020 | 0.0015 |
| A | 1 | - | 50 | 0.7016 | 0.0016 | 0.6943 | -0.0073 | 0.6994 | -0.0022 | 0.0051 |
| A | 9 | 30 | 100 | 0.8110 | 0.0018 | 0.7886 | -0.0224 | 0.7928 | -0.0182 | 0.0042 |
| A | 9 | 30 | 50 | 0.8374 | 0.0018 | 0.8071 | -0.0303 | 0.8189 | -0.0185 | 0.0118 |
| B | 1 | - | 100 | 0.6233 | 0.0013 | 0.6074 | -0.0159 | 0.6113 | -0.0120 | 0.0039 |
| B | 1 | - | 50 | 0.6452 | 0.0012 | 0.6194 | -0.0258 | 0.6281 | -0.0171 | 0.0087 |
| B | 9 | 10 | 100 | 0.7617 | 0.0014 | 0.7445 | -0.0172 | 0.7589 | -0.0028 | 0.0144 |
| B | 9 | 10 | 50 | 0.7850 | 0.0015 | 0.7554 | -0.0296 | 0.7749 | -0.0101 | 0.0195 |

Table 4.16 Comparisons of KENO-IV and MUTUAL calculations
(cases where four surfaces (top, bottom, and two adjacent side surfaces) surrounded by reflectors)

| Unit type | Number of units | Unit isolation distance [cm] | Distance to reflector [cm] | KENO-IV calculations | | MUTUAL calculations Albedo 0.73 | | MUTUAL calculations Albedo 1.0 | | |
|-----------|-----------------|------------------------------|----------------------------|------------------------|----------|---------------------------------|---|--------------------------------|---|---|
| | | | | $k_{\text{eff}}^{(1)}$ | σ | $k_{\text{eff}}^{(2)}$ | $k_{\text{eff}}^{(2)} - k_{\text{eff}}^{(1)}$ | $k_{\text{eff}}^{(3)}$ | $k_{\text{eff}}^{(3)} - k_{\text{eff}}^{(1)}$ | $k_{\text{eff}}^{(3)} - k_{\text{eff}}^{(2)}$ |
| A | 1 | - | 100 | 0.6802 | 0.0017 | 0.6830 | 0.0028 | 0.6838 | 0.0036 | 0.0008 |
| A | 1 | - | 50 | 0.6843 | 0.0016 | 0.6876 | 0.0033 | 0.6901 | 0.0058 | 0.0025 |
| A | 9 | 30 | 100 | 0.7798 | 0.0021 | 0.7846 | 0.0048 | 0.7874 | 0.0076 | 0.0028 |
| A | 9 | 30 | 50 | 0.7878 | 0.0020 | 0.7984 | 0.0106 | 0.8074 | 0.0196 | 0.0090 |
| B | 1 | - | 100 | 0.6014 | 0.0011 | 0.6034 | 0.0020 | 0.6057 | 0.0043 | 0.0023 |
| B | 1 | - | 50 | 0.6099 | 0.0012 | 0.6092 | -0.0007 | 0.6138 | 0.0039 | 0.0046 |
| B | 9 | 10 | 100 | 0.7170 | 0.0013 | 0.7401 | 0.0231 | 0.7525 | 0.0355 | 0.0124 |
| B | 9 | 10 | 50 | 0.7245 | 0.0014 | 0.7459 | 0.0214 | 0.7611 | 0.0366 | 0.0152 |

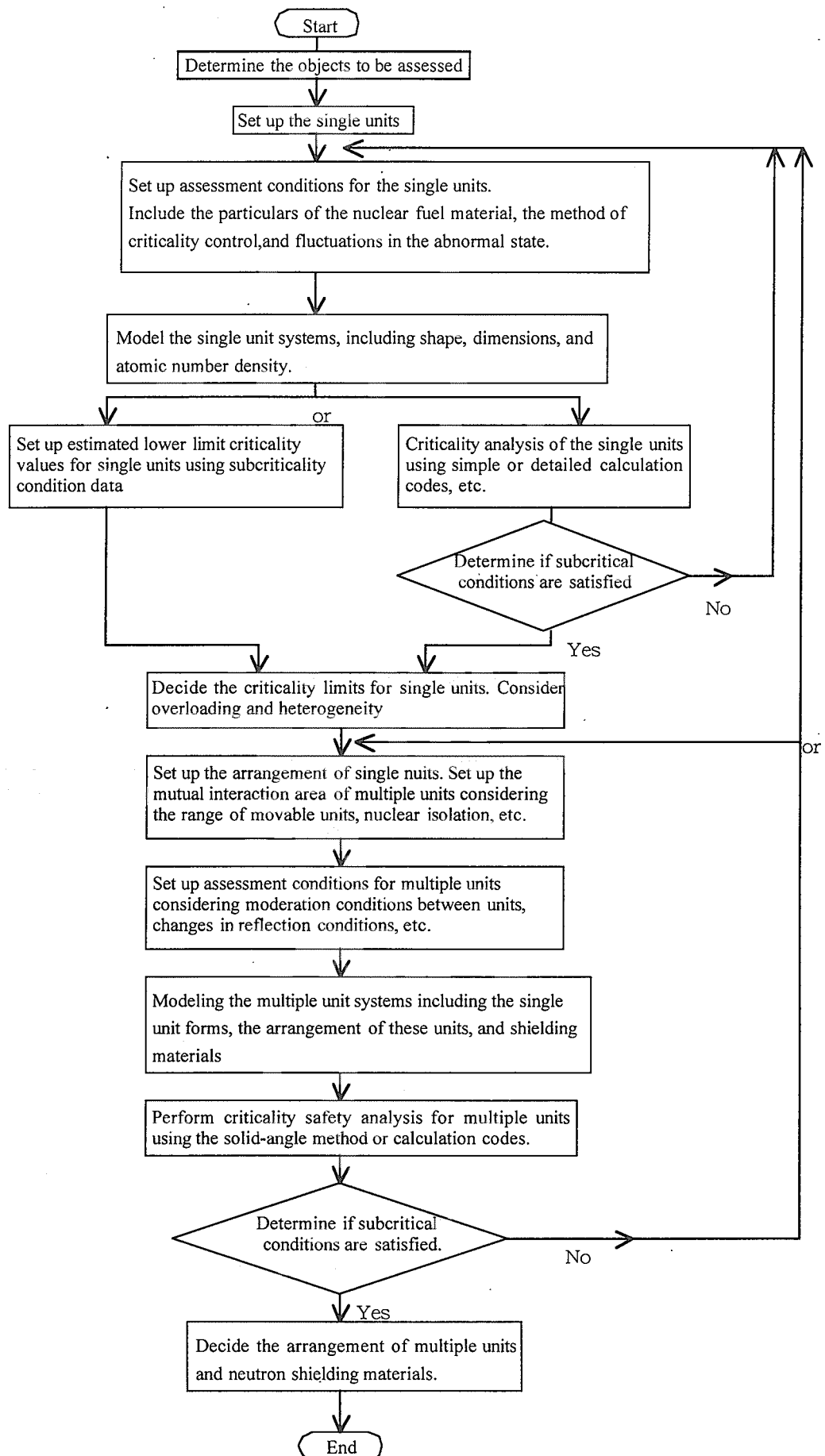


Fig 4.1 Procedure for criticality safety assessment

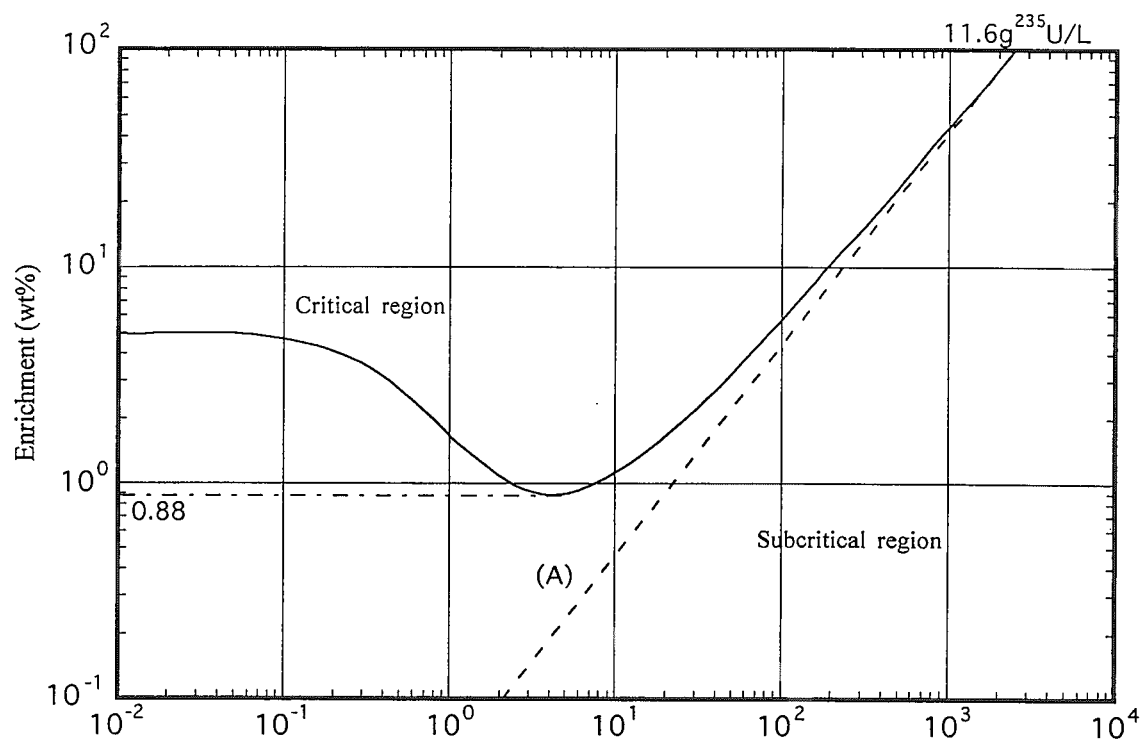


Fig 4.2 Criticality judgment diagram in terms of H/U atomic ratio and enrichment for an infinite system of homogeneous U-H₂O

The dotted line (A) shows a relation between enrichment and H/U atomicratio at a constant ²³⁵U concentration of 11.6g ²³⁵U/L

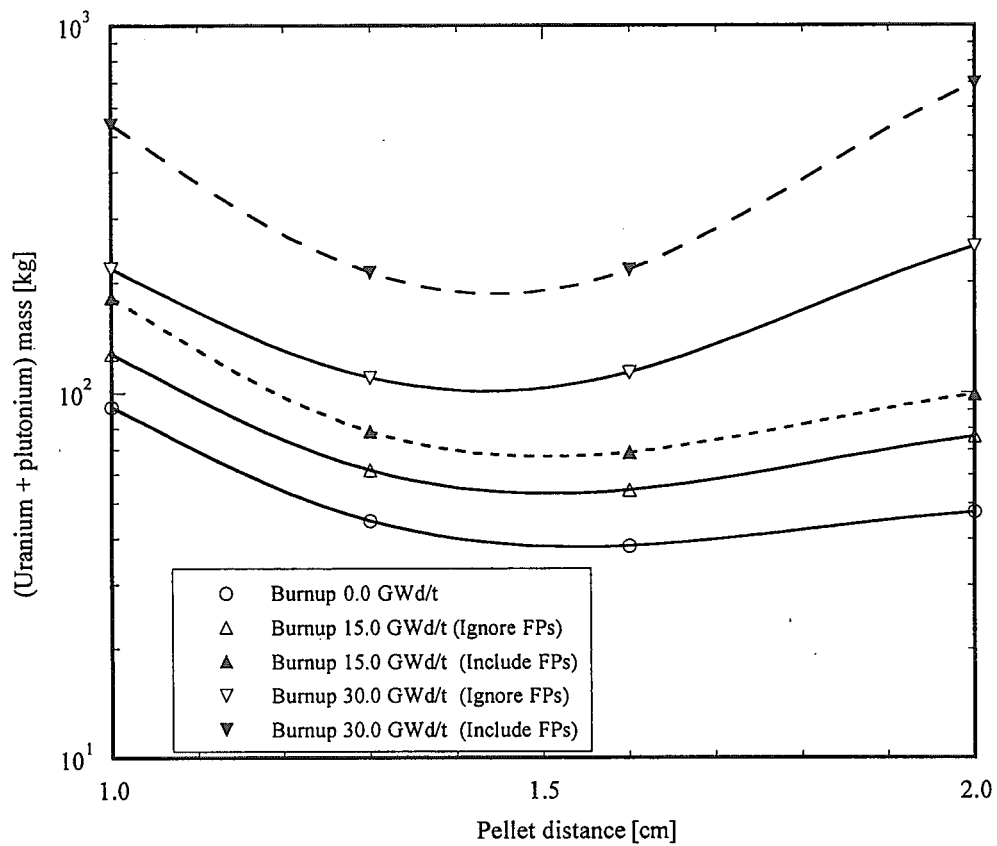


Fig 4.3 Subcritical mass of fuel (1)
 (Heterogeneous UO_2 pellet- H_2O , sphere, asufficiently thick reflector,
 (uranium+plutonium)mass equivalent to $k_{\text{eff}}=0.98$)

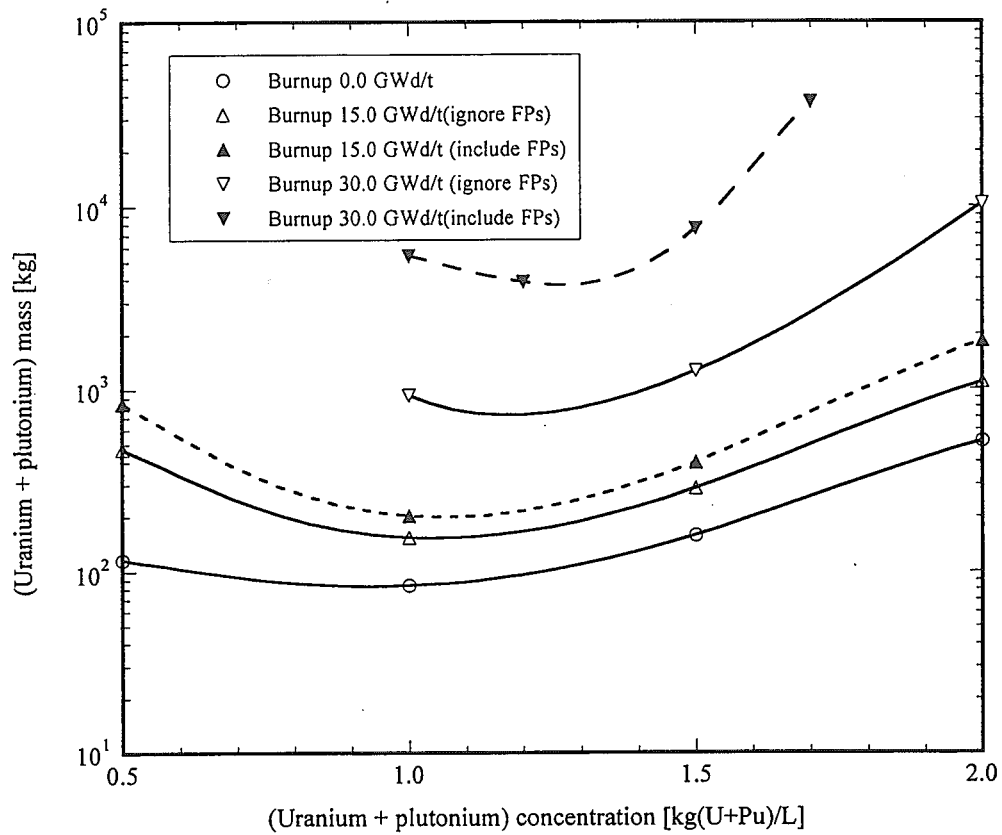


Fig 4.4 Subcritical mass of fuel (2)
 (homogeneous $\text{UO}_2(\text{NO}_3)_4$ - $\text{Pu}(\text{NO}_3)_4$ water solution, sphere, a sufficiently thick reflector, (uranium+plutonium) mass equivalent $k_{\text{eff}}=0.98$)

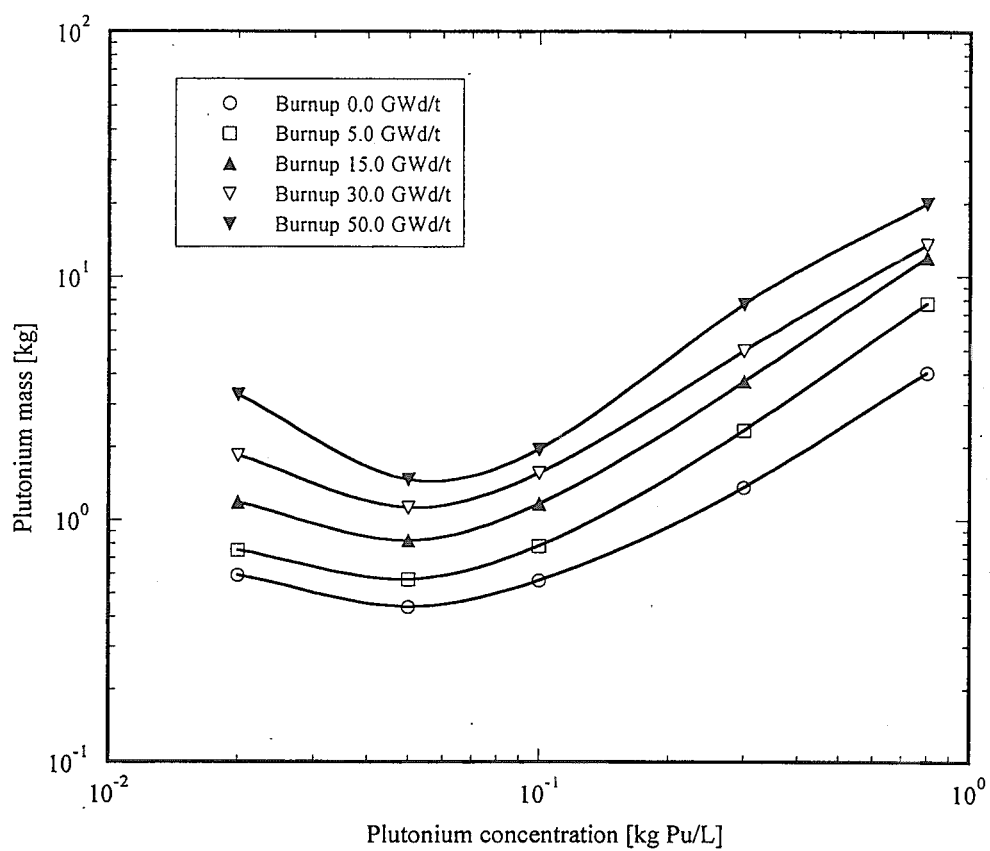


Fig 4.5 Subcritical mass of fuel (3)
 (homogeneous $\text{Pu}(\text{NO}_3)_4$ water solution, sphere, a sufficiently thick reflector, (plutonium) mass equivalent to $k_{\text{eff}}=0.98$)

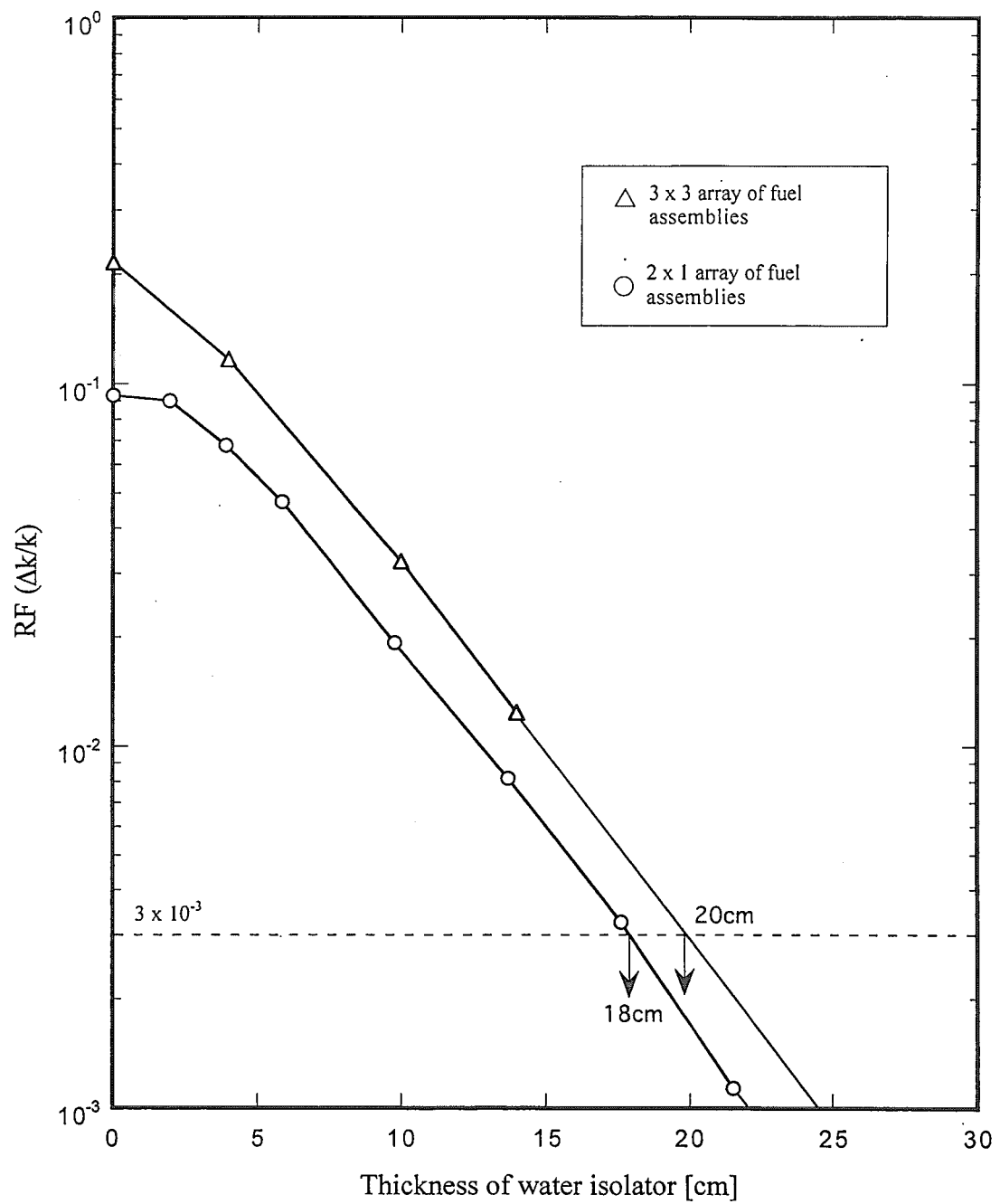


Fig 4.6 Changes in RF versus the thickness of a water isolator
(BWR fuel assembly in water)

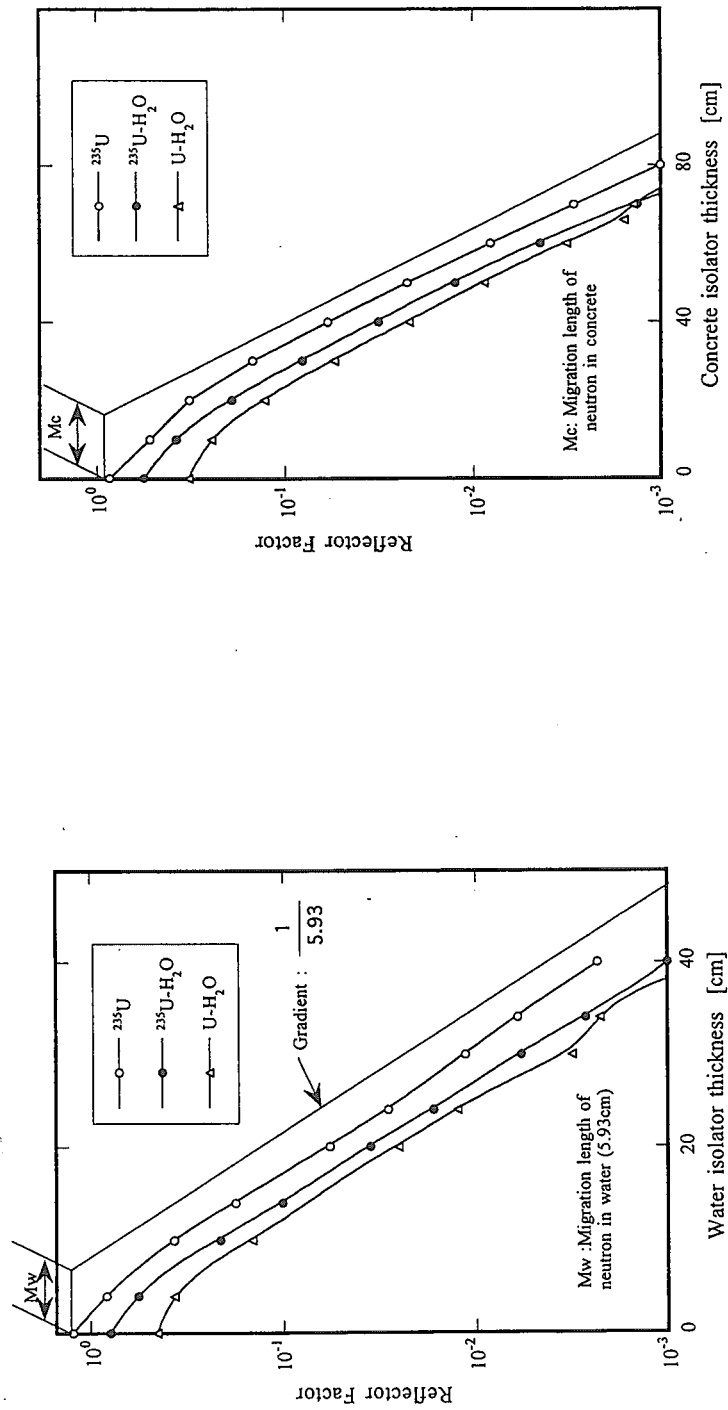


Fig 4.7 Changes in Reflector Factor versus the thickness of water and concrete isolator

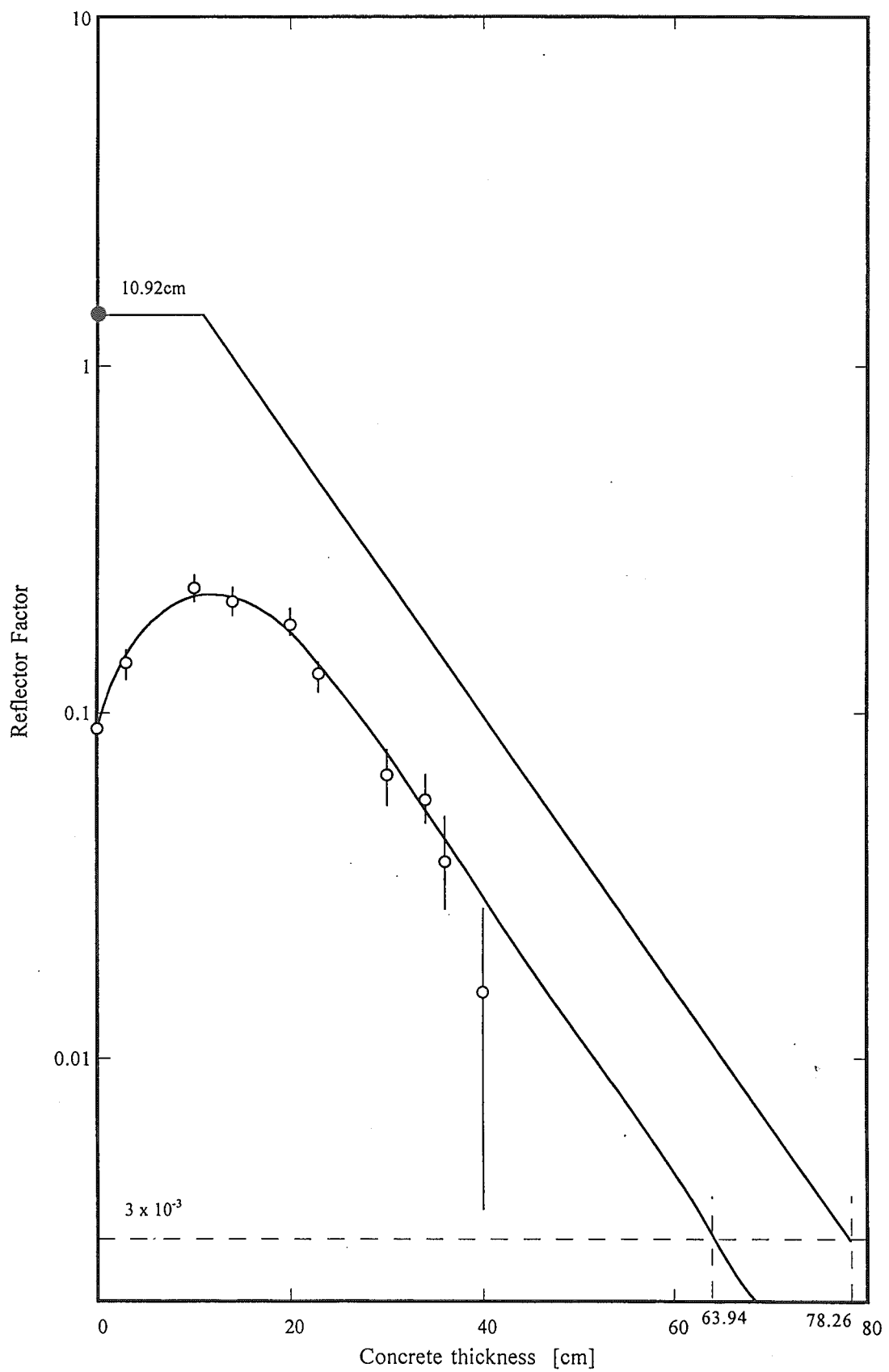
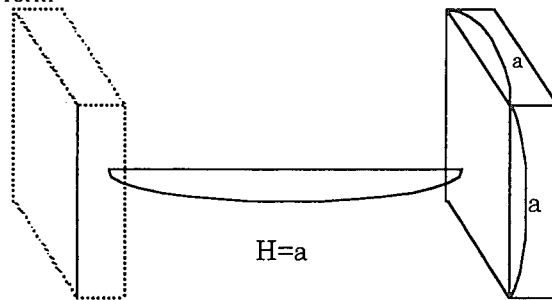


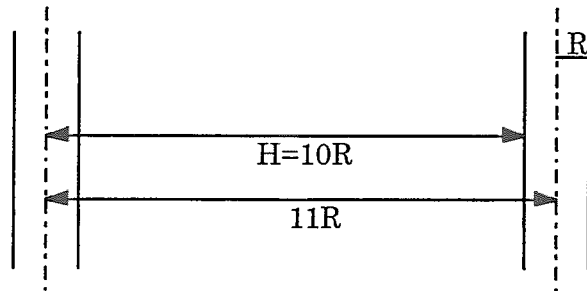
Fig 4.8 Changes in RF with respect to thickness of concrete isolator (BWR fuel assembly in storage facility).

(1) Slab having parallel square form



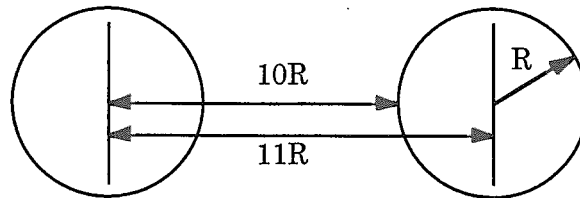
$$\Omega = 4 \sin^{-1} \left[\frac{\frac{a}{2} \cdot \frac{a}{2}}{\sqrt{\frac{a^2}{4} + a^2} \cdot \sqrt{\frac{a^2}{4} + a^2}} \right] = 0.805 \text{ sr (steradian)}$$

(2) Infinite circular cylinder



$$\Omega = \lim_{L \rightarrow \infty} \frac{L \cdot 2R}{\sqrt{(L/2)^2 + (10R)^2}} \cdot \frac{1}{10R} = 0.4 \text{ sr (steradian)}$$

(3) Sphere



$$\Omega = 2\pi \left[1 - \frac{1}{\sqrt{1 + (R/10R)^2}} \right] = 0.03 \text{ sr (steradian)}$$

Fig 4.9 Examples of solid-angle calculations by the point-plane model of TID-7016 Rev. 2

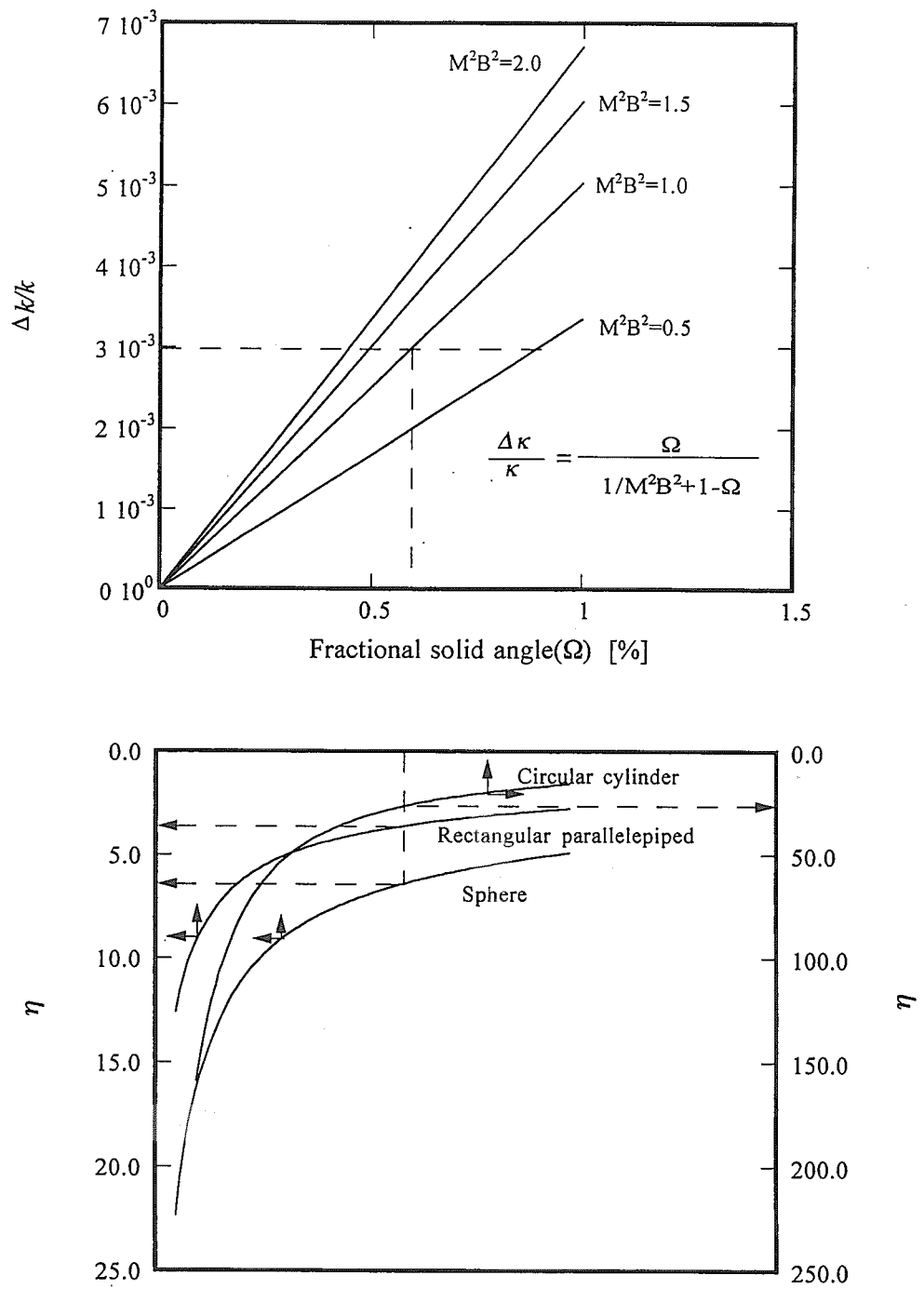


Fig 4.10 Relationship of fractional solid angles and distance between single units

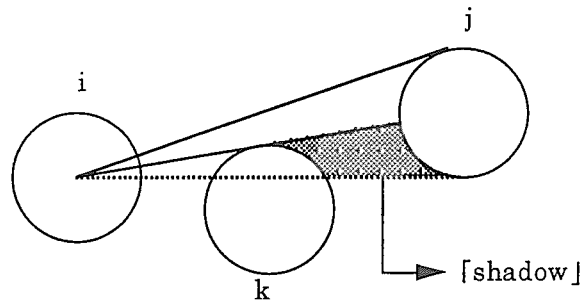


Fig.4.11 Effect of "Shadow"

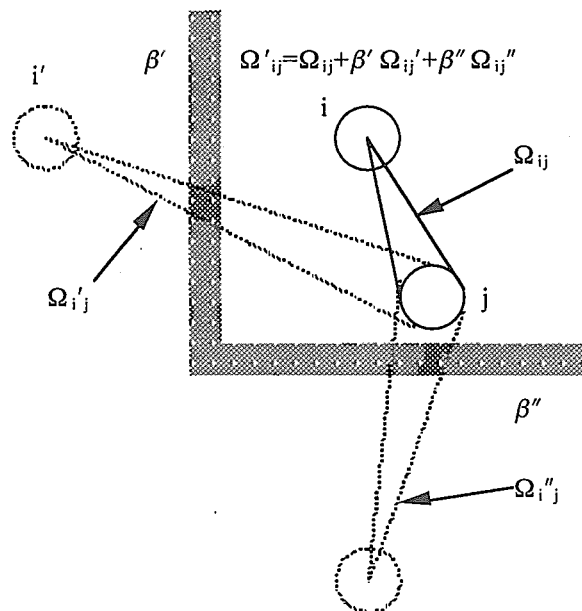
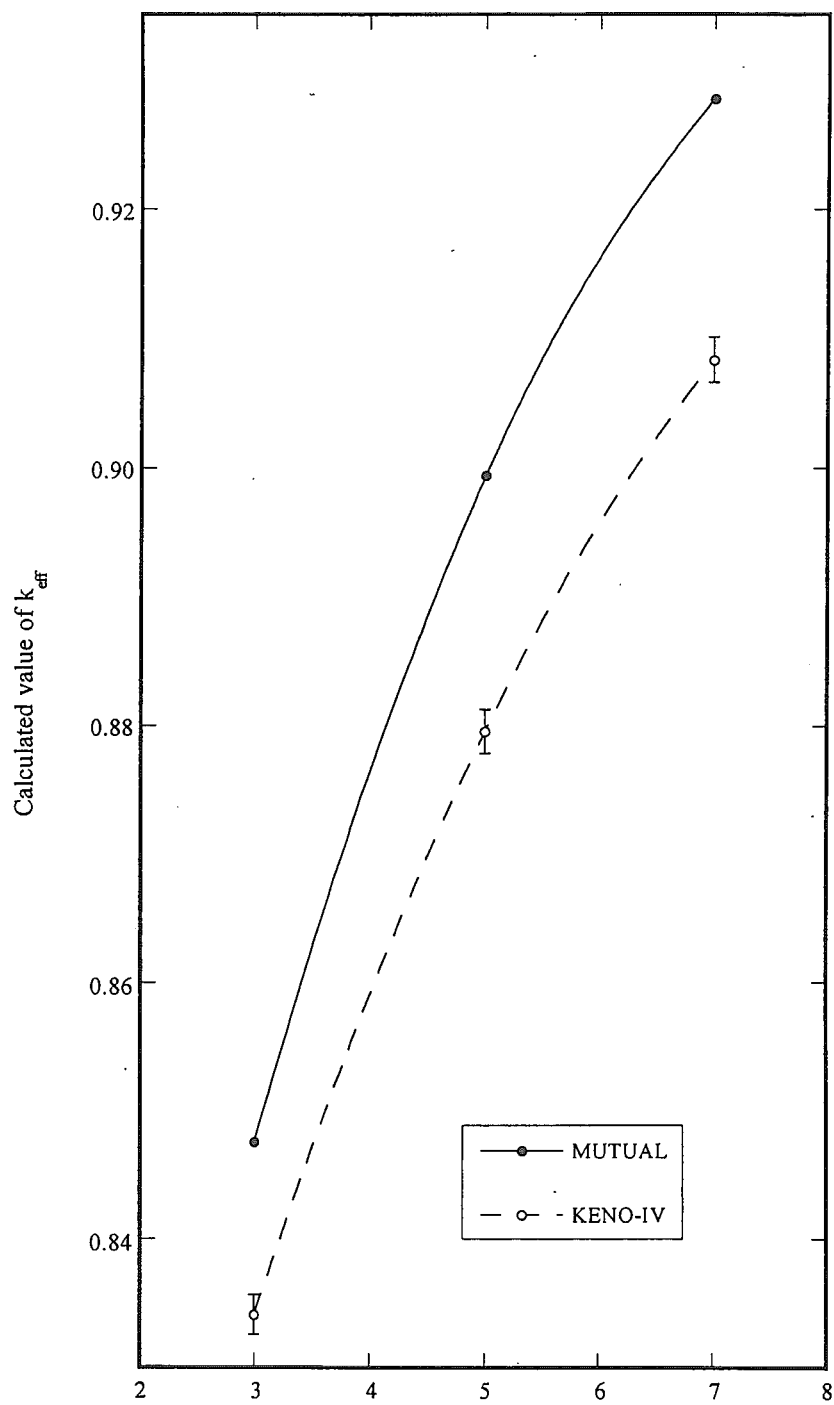


Fig.4.12 Effect of Reflector



Number of units in the configuration : N
 Unit shape : Circular cylinder, diameter 25.54cm, height 100cm
 Unit composition : Homogeneous UO_2-H_2O , ^{235}U enrichment 5wt%,
 uranium concentration 1000gU/L, H/U=24.0
 distance between units : 30cm
 Configuration type : N x N x 1
 KENO-IV History : 1000neutrons x 200generations

Fig 4.13 Changes in k_{eff} against the number of units in the configuration

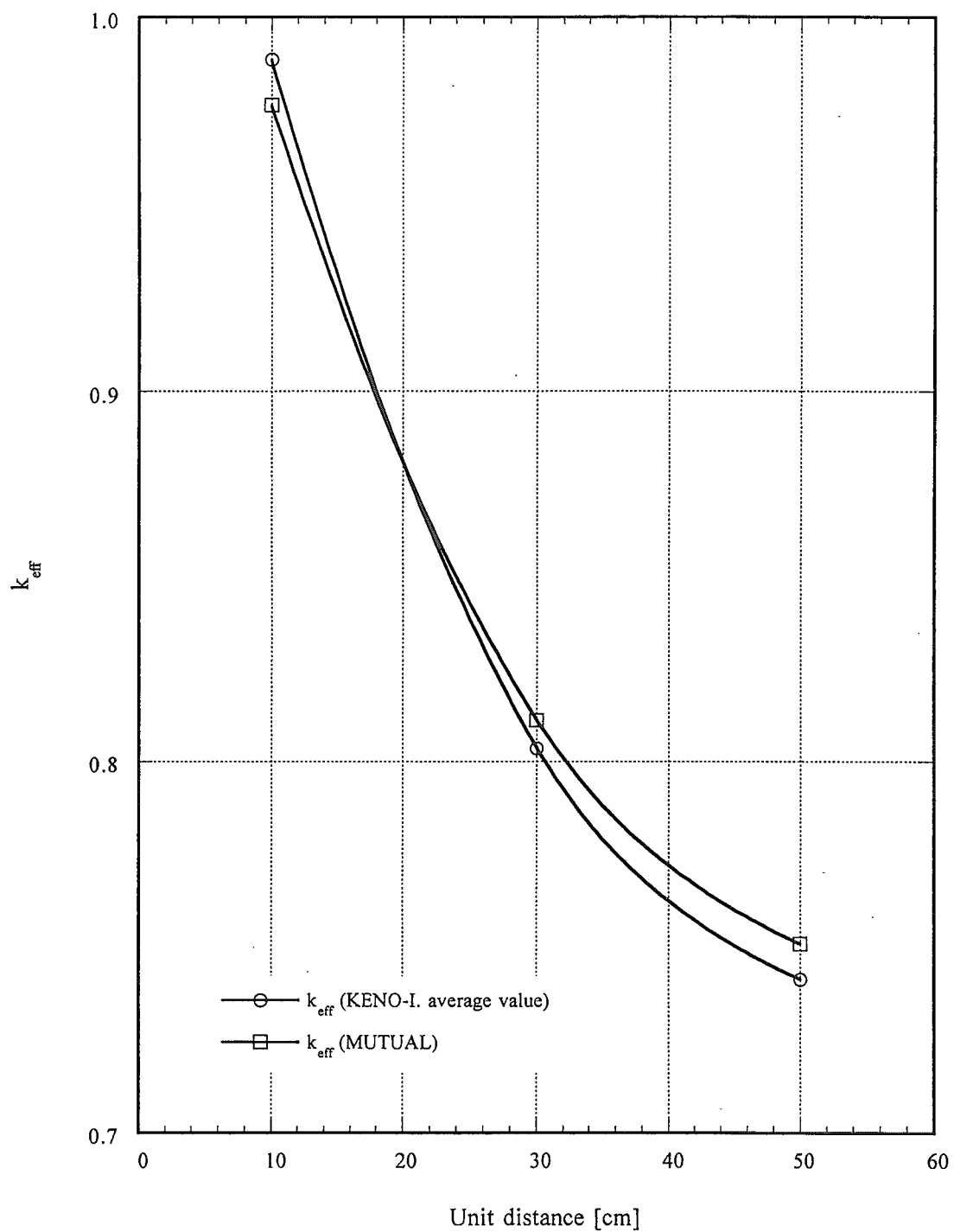


Fig 4.14 Dependence of effective multiplication factor on distance from one unit to another (UO₂-H₂O moderated system, 4x4x1 array)

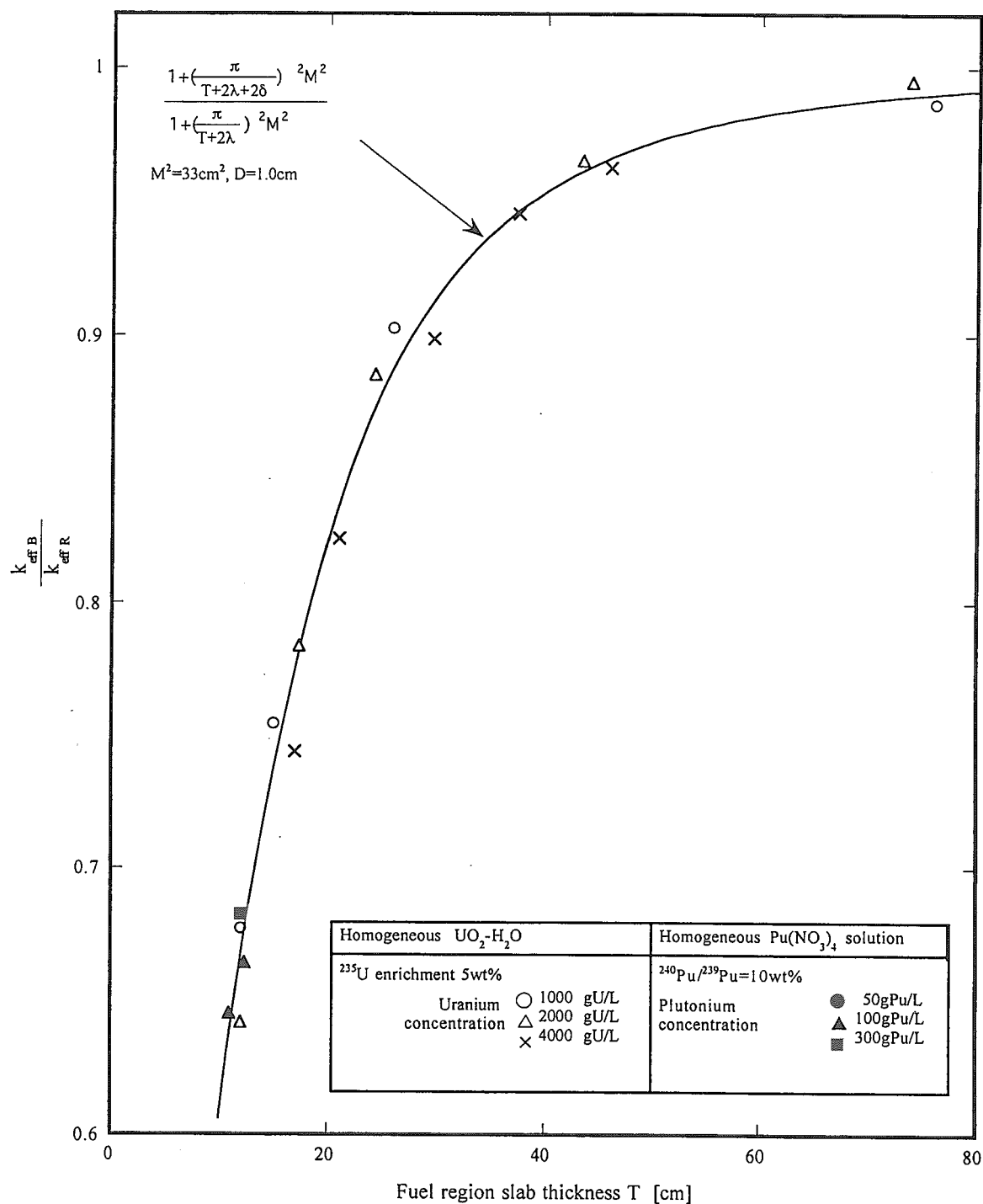


Fig 4.15 Changes in the $k_{\text{effB}}/k_{\text{effR}}$ ratio versus the thickness of a homogeneous $\text{UO}_2\text{-H}_2\text{O}$ infinite slab and a homogeneous $\text{Pu}(\text{NO}_3)_4$ aqueous solution infinite slab

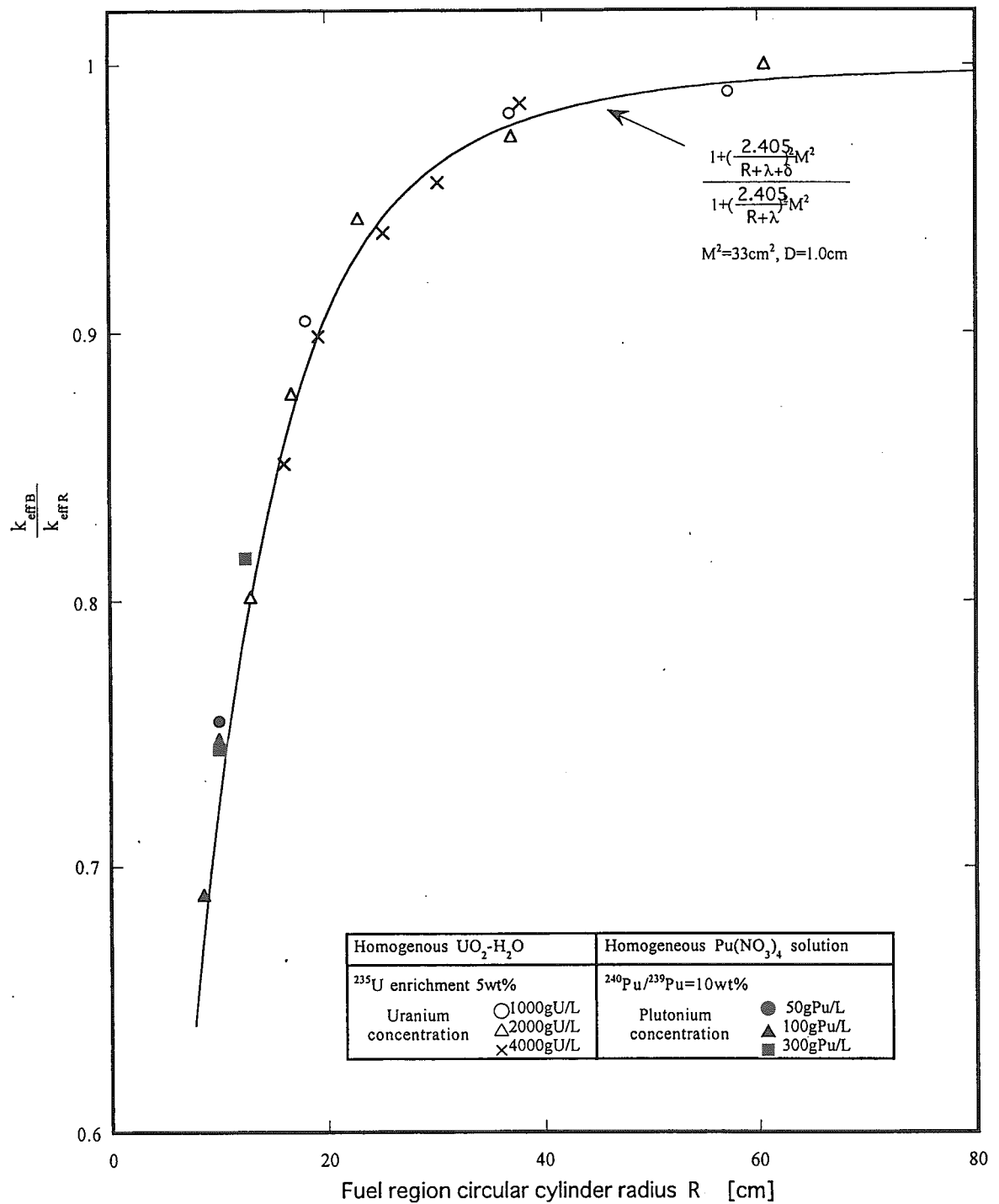


Fig 4.16 Changes in the k_{effB}/k_{effR} ratio versus the thickness of a homogeneous $\text{UO}_2\text{-H}_2\text{O}$ infinite circular cylinder and a homogeneous $\text{Pu}(\text{NO}_3)_4$ aqueous solution infinite circular cylinder

5. CRITICALITY SAFETY OF CHEMICAL PROCESSES

5.1. Introduction

Of processes treating nuclear fuels, those processes which change the physical and chemical forms of nuclear fuels or separate their elements by using chemical reagents, are generally called "chemical processes". In this chapter, as typical examples of chemical processes, we introduce a dissolution and an extraction process in "reprocessing," and address the issue of criticality safety of the chemical processes.

When spent fuels are reprocessed by wet methods (e.g., chop & leach, Purex), nitric acid or organic solvents are used as chemical reagents. Modeling and analytical methods used to evaluate criticality safety of the chemical processes do not differ from those used in other processes. The criticality safety evaluation of the chemical processes, however, because of the use of the chemical reagents, requires special considerations as outlined below:

- (1) Chemical reagents such as nitric acid aqueous solution or organic solvent solutions, which are essential process mediums coexisting with nuclear fuels, act as neutron moderators. In addition, the abundance ratio between nuclear fuels and the mediums changes with varying concentrations of the mediums. Therefore, in the criticality safety evaluation of the chemical processes, the changes in moderating conditions associated with the changes of this abundance ratio between the nuclear fuels and the mediums are considered.
- (2) Due to the chemical reagents as essential process mediums coexisting with nuclear fuels or a small amount of other chemical reagents added in the processes, changes sometimes occur in the physical and chemical forms or in the compositions of nuclear fuels. As a result, the number of critical factors affecting the results of the criticality safety evaluation increases and their effects become more complicated. In the criticality safety evaluation, therefore, attention is given to the sensitivity of each criticality factor to the neutron multiplication factor, and parameters for the criticality safety calculations are carefully set so that the calculations provide conservative results.

In addition, the criticality safety design of the chemical processes can be conducted in a more reasonable manner by taking account of the following points:

- (a) At those processes where the plutonium content in nuclear fuels is low, or at those processes where the plutonium concentration in process mediums is low or the probability of the plutonium concentration increasing is quite small, it is sometimes more reasonable to set control limits for nuclear fuel or plutonium concentration and control actual concentrations within the control limits than adopting geometrical control over the full plutonium concentration range.

- (b) At those processes where the plutonium content in nuclear fuels is high, or at those processes where the plutonium concentration in process mediums is high or the probability of the plutonium concentration increasing is quite large, it is possible to relax some of the criticality safety design constraints by placing a restriction on the plutonium composition in criticality safety control.

Therefore, at some processes of a big reprocessing plant, one criticality control method to be selected is to set nuclear control limits for such criticality factors as concentration of nuclear fuels or plutonium, burnup or composition of spent fuel, and to use various instruments (including sampling and analysis) for monitoring and controlling these "criticality control factors". It is important to note that where this control method is adopted, special considerations are given to maintaining the reliability of the instruments used.

5.2. Cautions for Criticality Safety Design of Chemical Processes

The concept of defense in depth is one of the basic principles of nuclear facility design. Defense in depth is comprised of three general levels. The first level consists of those measures taken to "prevent the occurrence of an abnormal event" from developing into an accident. The second level consists of those measures taken to detect the abnormality and "prevent the abnormality from propagating to an accident," should an abnormal event occur. The third level consists of those measures taken to "reduce or mitigate the consequences of an accident," should an accident occur. Since criticality accidents have the potential to be quite severe, the criticality safety design is required to "prevent criticality in any technically postulated contingency," and the criticality safety design corresponding to the first level of defense in depth is of primary importance.

In the conventional criticality safety design of the chemical process, as an action to "prevent the occurrence of an abnormal event," geometry control in the full fuel concentration range (including the combined use of neutron absorbers) was adopted for every component in order to eliminate the possibility of criticality. Namely, this method is equivalent to guaranteeing that criticality factors are kept within subcritical limits by adopting geometrical design.

As the size of the chemical processes enlarged, however, adopting geometry control at all processes became more difficult, and criticality factors to be controlled during plant operations were introduced. Specifically, a highly reliable criticality safety design utilizing the proper instrumentation and control devices became necessary. In this method, instrumentation and control devices are categorized into two ways in the criticality safety design according to their expected functions: the first method is using them as measures to "prevent the occurrence of an abnormal event" and the second method is using them as measures to "prevent an abnormality from propagating to a criticality accident". Specifically, the first method is to monitor process parameters such as flow and temperature through the use of instrumentation and control devices and to maintain the actual values

within control limits, and the second method is to monitor criticality factors such as concentration and mass of fissile material directly and control the actual values within control limits.

The first method is based on the philosophy that controlling process parameters such as flow rate and temperature within a proper control range is the most essential in the criticality safety of the chemical processes. In this method, the control range of each process parameter is set such that a significant abnormal event does not occur in criticality factors such as fissile material concentration and mass, thus, this can be classified as the first level of the defense in depth. When relying on this method, however, we must identify every single process parameter affecting criticality and control each parameter within a specified limit without fail.

In contrast, in the second method, we set subcritical limits for concentration and mass of fissile nuclides, monitor that variable criticality factors are maintained within the subcritical limits during operations through the use of instrumentation and sample analyses, and if an abnormality is detected, take immediate actions such as emergency process shutdown to “prevent an abnormality from propagating to a criticality accident.” Though this method relies on the criticality safety based on the second level of the defense in depth, it goes without saying that control functions for maintaining normal operating conditions, i.e., measures to “prevent an abnormality from propagating to a criticality accident,” remain the most important.

Precautions in case of guaranteeing criticality safety by controlling those criticality control factors during operations through the use of instrumentation devices are discussed below:

- (1) When criticality safety in the chemical processes is secured through the use of instrumentation and control devices, the instrumentation and control devices must be highly reliable so that operation continues after a single failure, or loss of external electrical power. Operating limits or control limits must be set well below the maximum permissible limit after allowing for the overshoot of interlock systems and instrumentation measurement errors.
- (2) When criticality safety in the chemical process is secured by controlling process parameters, the relationship between each process parameter and criticality factors such as concentration and mass of fissile materials must be well understood in order to verify the validity of the control limit range of each parameter.
- (3) When criticality safety is secured by controlling mass or concentration of fissile nuclides through the use of instrumentation and control devices, control limits of these criticality control factors are to be set by taking full account of interference of other nuclides and possible pertinent nuclide compositional changes.
- (4) When criticality safety is secured by off-line analyses or measurements, operating procedures must be set such that the next operational step cannot be initiated without confirmation of analytical or measurement results. It must be made sure that actual values of criticality safety factors being measured or analyzed do not change until the next operational step. In addition, potential sample representation errors and errors

associated with measuring and analyzing must be fully considered

- (5) When a control limit is set on burnup as a criticality control factor, reliable measurement or calculated data must be used. Measurement or calculation errors must be fully considered.

5.3. An Example for Criticality Safety Analysis of a Dissolution Process ⁽¹⁾

5.3.1. Design of a Dissolution Process

The process design of the dissolution process includes performing a structural design of the dissolver to achieve a target throughput, setting dissolving parameters to secure a dissolving solution with desirable specifications, and performing a system design including instrumentation in order to realize the desired dissolving parameters. The criticality safety design is closely related with both the structural design and the system design, and it competes with the structure design for achieving a target throughput in particular.

The evaluation model for the dissolution process assumes a cylindrical batch dissolver vessel. In the batch type dissolver, since series of individual operations, such as filling the vessel with nitric acid, loading the fuel, dissolution, extraction of the dissolved solution, and cleaning and expelling of the hull, have a significant influence on throughput, the shape and dimensions, etc. are designed by taking account of each individual operation's efficiency. An example flow chart of the dissolution process design is shown in Fig. 5.1.

5.3.2. Setting of Assessment Condition for a Dissolver

In general, criticality factors in the process design can be classified as below:

- (a) Criticality factors of which subcriticality is guaranteed by process design

These criticality factors are divided into two groups: one group of criticality factors of which values set by the geometry design do not vary during operations and the other group of criticality factors of which actual values vary during operations, but by imposing most conservative parameters in design calculations, it is not necessary to consider the variability of those criticality factors during operations in terms of criticality safety.

- (b) Criticality factors of which subcriticality is guaranteed during operations

These criticality factors take various values or states during operations. In the process design stage, specific control limits are formulated, and subcriticality is guaranteed by maintaining actual values and states during operations within control limits through the use of instrumentation and manual operator actions.

In the criticality safety design of the chemical process, since the criticality factors described above have some uncertainty associated with operational fluctuations, selection of parameters and their values to be used for each criticality factor in the design becomes very crucial, and thus it is vital that we fully understand the margins associated with the criticality factors. The criticality factors can roughly be classified into geometrical parameters, fuel parameters and the states of fissile materials. With these in mind, we selected the following parameters for this evaluation model.

(1) Geometrical parameters

- Diameter of dissolver (fabrication dimension): 29 cm
- Diameter of bucket (fabrication dimension): 24 cm

Other geometrical parameters include the bucket height and plate thickness. The height of the heterogeneous region of the bucket affects moderation conditions and thus affects the loading amount of fuel pieces, and when the system size is small, the height gives a significant effect on the reactivity. Additionally, the criticality evaluation must consider the effects of the structural materials. Although the material composition and plate thickness have minimal impact on the reactivity, the structural materials can still act as absorber or reflector materials and impact system criticality. In the evaluation of this document, however, they were omitted for the sake of convenience.

The amount of deformation or size changes due to corrosion, thermal expansion, and fabrication permissible should be determined in a conservative manner accounting for operating and fabrication conditions and they were not addressed in this evaluation. The contributions of these three factors to reactivity are relatively small. "Other deformation margins" are considered in some evaluations, but not in others. When they are considered, specific causes for deformation, e.g. earthquake, are identified in some cases, but not in others. Whether earthquakes should be superimposed on the criticality safety design, however, depends on which has larger safety margins, the criticality safety design or the seismic design of the pertinent component?

- Reflector: vacuum, 2.5 cm of water reflector, 30 cm of water reflector

Three reflector conditions above were used: 30 cm of water reflector assuming a water submersion event, 2.5 cm of water reflector considering the reflector effects of surrounding structural materials and personnel, and vacuum condition to create a baseline of comparison for the two water models mentioned above.

(2) Fuel parameters

- Fuel type: PWR fuel
- Initial enrichment: 4.1 wt%
- Specific power: 38 MW/t

- Cooling time: 4 years

Although the above mentioned fuel parameters are not directly input into any criticality calculation code, they are treated as criticality factors because of their impact on the fissile nuclide composition, which ultimately influences the criticality results. Though these parameters vary from one fuel type to another, actual fuel specifications are usually available at the time of the process design. It is usually reasonable to select the most conservative values of these parameters with respect to reactivity. As a model case, the above values were selected.

- Burnup: 0 ~ 48 GWd/t
- Nuclides: U+Pu, U+Pu+Ac, U+Pu+FP, U+Pu+Ac+FP
 (Ac: ^{237}Np , ^{241}Am , ^{243}Am)
 (FP: ^{149}Sm , ^{143}Nd , ^{133}Cs , ^{152}Sm , ^{155}Gd , ^{145}Nd , ^{147}Sm , ^{153}Eu , ^{150}Sm)

Among the fuel parameters, it is important to understand the changes in nuclide compositions associated with the fuel burnup, since the burnup level has a significant impact on the margin of safety.

When the target throughput is small, assuming the maximum initial enrichment of the pertinent fuel type (4.1 wt% in this evaluation) and no burnup gives the most conservative margin of safety.

When the target throughput is large and highly reliable burnup data are available, we can secure some safety margins by assuming nuclide compositions based on the formation of actinides and FP associated with the burnup. The actinides and FP considered in the evaluation must be stable nuclides that definitely exist in the pertinent process. The percentage of each nuclide in the fuel can be directly obtained from a burnup calculation code or more conservative values may be obtained by multiplying the calculated values by some safety factor.

In this evaluation, nuclides identified above that are stable ones in the dissolution process and a burnup of up to the present maximum 48GWd/t were used.

(3) States of fissile materials

- Bucket region: optimal or near optimum moderator lattice configuration

One feature of the criticality safety design of the dissolver is that the heterogeneous system of sheared fuel pieces gives a dominant effect on reactivity. Therefore, the lattice configuration and fuel pieces and their associated moderating conditions give a significant impact on the effective multiplication factor. When the lattice configuration and distribution of fuel pieces cannot be specified, assuming the optimum moderating condition gives the most conservative results. In order to understand the moderating conditions and their associated safety margins, however, the sensitivity of the lattice configuration to the effective multiplication in the vicinity of the optimum moderating

condition were evaluated.

- Solution region: water, fuel solution (400g(U+Pu)/L, free nitric acid 0, 3N)

Modeling the dissolver using water reflector in the solution region generally gives the largest reactivity. If actual fissile material and nitric acid concentration values can be determined through the use of highly reliable instruments or analyses, those values should be incorporated into the criticality safety design. In this evaluation, the fuel solution concentration above was selected as a model case.

- Solution temperature: 15 °C, 100 °C

Since the solution temperature is used to calculate the solution density, temperatures in the vicinity of 15 °C are considered conservative since they result in the highest solution density. In addition, this temperature well suits the use of nuclear data, which were evaluated at a room temperature. The operating temperature of the dissolver may also be used in the calculation, assuming that the temperature can be validated by highly reliable methods. We also evaluated 100 °C to simulate process boiling conditions.

Fig. 5.2 shows a schematic model for this evaluation.

5.3.3. Results of Criticality Analysis for a Dissolver

This section provides example criticality safety evaluations for each of the criticality factors discussed in the previous section. Three cases of fuel burnup, 0, 30, and 48 GWd/t, were selected from the full range of 0 - 48GWd/t. Two solution temperatures, 15 °C and 100 °C, were used to evaluate the change in solution density with respect to temperature. As for the solution region, when the dissolving solution is used, free nitric acid concentrations of 3 N(normal) and 0 N were selected.

(1) Lattice configuration of fuels in a bucket region under optimum moderating condition

The maximum fuel pin radius was selected based on a 0.929 cm diameter PWR fuel pin, and the optimum lattice configuration for this pin radius or lattice spacing was evaluated. One example of the evaluations is shown in Fig. 5.3. The two graphs show the results for uranium and plutonium fuels with a burnup of 30 GWd/t; one graph is for water in the solution region and the other is for 3N nitric acid.

In this example, the neutron multiplication factor of the fuel pin with the maximum diameter (0.929 cm) shows the maximum. Other calculations varying burnup and fuel composition (considering FP and actinides) showed similar behavior. The optimum moderating condition for each fuel composition was searched in this manner, and the criticality calculations of the dissolver in the next section were conducted based on the lattice configuration under the optimum moderating

condition or in its vicinity for each selected fuel composition.

(2) Dissolver Criticality Calculation Results

The criticality calculation results under the optimum moderating condition of the dissolver are given in Tables 5.1 to 5.5. In addition, Fig. 5.4 shows the plots of the above calculations for 30 cm water reflector in the solution region, and it quantifies the changes in the effective multiplication factor due to burnup, FP, and actinides. As the figure shows, k_{eff} for the 30 GWd/t burnup fuel was more than $0.1\Delta k$ lower than the k_{eff} of the new fuel. This can be primarily explained by the changes in uranium and plutonium composition associated with an increased burnup. Adding FP further reduced k_{eff} by about $0.05\Delta k$, while the addition of non-uranium/plutonium actinides resulted in a non-significant change of about $0.01\Delta k$.

The effective neutron multiplication factor with fuel solution (400g(U+Pu)/L) plus nitric acid solution in the solution region was lower than that of water in the solution region by about $0.07\Delta k$ for 3N nitric acid solution and by about $0.04\Delta k$ for 0 N nitric acid solution (all cases with 30 cm water reflector).

The effective multiplication factor for a 100°C water in the solution region was $0.01\Delta k$ lower than a 15°C water in the solution region, and the effective multiplication factor for a 100°C fuel solution in the solution region was about $0.03\Delta k$ lower than a 15°C fuel solution in the solution region (all cases with 30 cm water reflector).

In addition, in order to evaluate the safety margins of this optimum moderating condition, criticality calculations were performed in the range of 1.14 to 1.97 cm (volume ratio of water to fuel from 0.5 to 3.5) of lattice pitch with a fuel pin diameter of 0.929 cm. The results are shown in Fig. 5.5 (burnup: 30 GWd/t, fuel composition: U+Pu, solution region: water). Assuming that the actual dissolver water to fuel volume ratio widely varies between 1.0 and 1.5, the effective multiplication factor under the optimum moderating condition contains about $0.06\Delta k$ to $0.01\Delta k$ of safety margins in case of 30 cm-thick water reflector.

5.3.4. Safety Margins Found in an Example of a Dissolver

As shown in the previous section, consideration of burnup and the existence of FP and non-uranium/plutonium actinides decrease the effective multiplication factor by more than $0.1\Delta k$ for 30 GWd/t burnup fuel. The effective multiplication factor would further decrease if other factors, such as fuel dissolution in nitric acid, solution temperature increase, and lattice configurations that do not result in the creation of optimal moderator conditions, are considered. Among these factors, fuel dissolution and solution temperature increases cannot be expected to occur always in a series of dissolving steps. By simply adopting the fuel composition for new fuel and the sheared fuel configuration under optimal moderator conditions, k_{eff} becomes significantly higher than the value associated with the actual process conditions. Thus, this difference can be considered as a potential

margin of safety.

5.4. An Example for Criticality Safety Analysis of an Extraction Process

The co-decontamination process in the Purex method separates fission products from uranium and plutonium by processing the nitric acid solution of the spent fuels after shearing and dissolving processes with the use of 30% TBP + dodecane solvent. Typical extractors include the mixer-settler, pulse column, and centrifugal extractor. In this document, we used a mixer-settler in the co-decontamination process as an example, and described an example of criticality safety design and potential safety margins for this chemical process.

5.4.1. Design of an Extraction Process

The objective of the extraction process is to separate fission products from the product (uranium and plutonium). The process design for a given fuel type and processing capacity is carried out in such a manner that the fission product concentration in the product is kept below an upper limit and the loss of uranium and plutonium to raffinate is minimized.

As for feed parameters for dissolved fuel solution, a uranium and nitric acid concentration (3N) that gives the optimum extraction are determined, and the feed flow rate is determined from the processing capacity. Plutonium and fission product feed concentrations are determined from design based uranium concentrations and processing fuel.

The size of the mixer-settler, number of mixer-settler stages, feed stage location, and scrubbing solution and solvent feed parameters, etc. are determined based on k sensitivity evaluations and experimental data of such key process parameters as the aqueous to organic phase volume ratio (A/O ratio), the upper limit for fission product concentration of the product, the upper limit for uranium and plutonium concentration in the raffinate, the scrubbing solution flow rate, and the solvent flow rate. The structure and the process control of the mixer-settler are determined based on considerations of the start-up of the extraction operation, flush out, decontamination, etc., and the structural materials are determined from the operating environment.

Process control methods and conditions are established for key process parameters (e.g., flow rate of scrubbing solution) such that the operational variability of the parameters does not result in either (1) exceeding the specified upper limits for a) fission product concentration of the product, and for b) uranium and plutonium concentration in the raffinate, nor (2) mixer-settler plutonium accumulation.

5.4.2. Setting of Assessment Condition for Mixer-Settler

(1) Geometrical parameters

- The number of stages and dimensions of the mixer-settler

Fig. 5.6 shows a process model schematic.

Stages one through nine, which comprise the first bank of the mixer-settler, were considered in the criticality evaluation. This is due to two reasons: (1) the existence of fission products and plutonium accumulation are only observed in bank one, and (2) the neutron interactions between the first and second bank can be ignored. Fig. 5.7 shows a schematic model of the first bank of the mixer-settler.

- Reflector: vacuum, 30cm water reflector

As mentioned in section 5.3.2, various size deformation factors should be included in a complete criticality evaluation, but for the sake of convenience, these considerations were omitted from the evaluation presented here.

(2) Fuel parameters

- Fuel type : PWR fuel
- Initial enrichment : 4.1 wt%
- Cooling time : 4 years

Since specifications of the fuel to be processed are available at the time of design, the following values were used to construct the model case.

- Burnup : 48 GWd/t
- Nuclides : U+Pu, U+Pu+Ac, U+Pu+FP, U+Pu+Ac+FP
(Ac: ^{237}Np , ^{241}Am , ^{243}Am)
(FP: ^{149}Sm , ^{143}Nd , ^{133}Cs , ^{152}Sm , ^{155}Gd , ^{147}Nd , ^{153}Sm , ^{150}Sm)

These fuel parameters are dictated by the fuel burnup history and do not vary during the operation of the mixer-settler. However, fuels of different burnup values may be mixed and processed in the mixer-settler. As mentioned in section 5.3.2 for the dissolver, when highly reliable burnup data are available and the actual burnup can be determined conservatively, we can consider actinides even other than uranium and plutonium and FP. In this document, we evaluated the effects of FP and actinides other than uranium and plutonium, using the fuel burnup of 48GWd/t and above-mentioned nuclides.

The uranium and plutonium compositions and the atomic number density ratio of uranium to plutonium for the fuel with 48 GWd/t burnup were selected as follows based on the description given in section 3.5.2.

U composition ^{235}U : 0.82 wt%
Pu composition ratio ^{239}Pu : ^{240}Pu : ^{241}Pu : ^{242}Pu =50:25:15:10

$$\text{Pu/U} \times 100 = 1.05$$

In addition, one case where the ^{239}Pu concentration was equal to 100 wt% was evaluated for comparison. Using the Pu/U ratio given above and a feed solution uranium concentration of 180g U/L, the plutonium concentration is calculated as 1.9g Pu/L, but 2g Pu/L was used in the actual evaluation. The concentrations of FP and actinide nuclides other than uranium and plutonium were calculated by ORIGEN-2 using the uranium concentration of 180g U/L.

(3) State of fissile materials

- Concentration of fissile materials: concentration variability due to abnormal process parameters.

The concentration of fissile materials, FP, and actinides may increase in the mixer-settler process due to abnormal process parameters. In this evaluation, we evaluated the concentration increase due to a low flow rate of organic solvent (30% TBP + dodecane) using the EXTRA.M code⁽²⁾. An initial normal operating organic solvent feed flow of 465 L/h was selected and the flow rate was assumed to decrease from 465 L/h to 418 L/h at eight hours from the start up. Since a spent fuel composition with a residual uranium enrichment of only 0.82 wt% was used for this evaluation, only the process plutonium concentration needs to be considered for criticality safety. Fig. 5.8 shows the changes of the maximum plutonium concentration in the process caused by the reduced organic solvent feed flow rate. A maximum plutonium concentration of about 36 gPu/L was reached 50 hours after the reduction of organic solvent feed flow rate to 418 L/h (about 60 hours from the operation start).

In addition, The two graphs in Fig. 5.9 show the plutonium concentration distribution in the aqueous and organic phases across the stages of the mixer-settler; the left graph represents the situation before the organic solvent feed flow rate reduction and the right graph depicts when the maximum plutonium concentration was reached. As a comparison, we evaluated the most conservative scenario associated with the organic solvent feed flow rate reduction by assuming that only ^{239}Pu of the maximum plutonium concentration following the organic solvent feed flow rate reduction is distributed uniformly within the entire mixer-settler.

The use of actual conditions such as plutonium concentration in the process, plutonium isotope composition, and the existence of FP and actinides other than uranium and plutonium would allow the criticality safety evaluation to become more realistic. However, this would require more stringent validation of these conditions based on reliable measurements and calculations.

5.4.3. Results of Criticality Analysis for a Mixer-Settler

This section presents the results of the mixer-settler criticality safety assessment with respect to the criticality factors discussed in section 5.4.2. The neutron multiplication factor was calculated at two stages, once at a normal operating condition, before the organic solvent

feed flow rate decreases, and the other at the maximum plutonium concentration. Various fuel types were considered for these evaluations: one fuel type contained only uranium and plutonium, and other fuel types contained other actinides and/or fission products (FP) in addition to uranium and plutonium. Table 5.6 shows the evaluation results.

The results are shown graphically in Fig. 5.10. The fuel type numbers (No.) in Fig. 5.10 correspond with those in Table 5.6. Note that No. 0 in Table 5.6 is the case where the maximum concentration of ^{239}Pu is uniformly distributed throughout the entire system. The solid and dotted lines in Fig. 5.10 represent this case.

5.4.4. Safety Margins Found in an Example of a Mixer-Settler

The calculations in the previous section show that an unattended abnormality will lead to a gradual increase in the mixer-settler plutonium concentration. It takes approximately 50 hours for the plutonium concentration to reach its maximum value, then the concentration value begins to decrease as some plutonium escapes into the raffinate. At the maximum plutonium concentration, the effective multiplication factor of U-Pu fuel is close to 1; however, when FP and actinides other than uranium and plutonium are considered, which is not the case with the existing design, the effective multiplication factor decreases by about 0.2 Δk . The effective multiplication factor at a normal operating condition is less than 0.2, which is very low.

In the actual plant design, since the raffinate and final product concentration of uranium and plutonium is tightly monitored, it is not likely that an abnormal condition would be left unattended for a long time. For example, in the scrub and organic solvent solution feed lines, of which flow rate changes result in critical process condition changes, flow meters are installed double on each line to automatically detect flow rate changes. If the measured flow rate is found outside of a specified range, action is taken to automatically stop the mixer-settler. In addition, an in-line neutron monitor is installed for direct plutonium build-up detection in the mixer-settler, thus abnormalities in extraction process conditions can be detected quickly and efficiently.

As mentioned above, large fluctuations in the nuclear fuel material concentration can occur during extraction process abnormalities. Thus, the criticality safety design calculations assume the highest possible fuel concentration, taking account of optimal moderating conditions and postulated abnormal events. This assumed maximum fuel concentration, however, it is much higher than the actual plant design control limit. Thus, this difference in concentration offers an extra margin of safety during normal operation of the concentration control system. In addition, ignoring the neutron absorption effects of the FP and actinides in the criticality calculation provides an additional margin of safety since these materials will inevitably exist in the co-decontamination extractors.

5.5. An Example for Criticality Safety Design and Control of Chemical Process

Next we demonstrate an example of criticality safety design and control at the solvent extraction process of a very large, modern reprocessing plant.

The solvent extraction process for this reprocessing plant consists of two main facilities. The separation facility houses the separation and partition equipment. The purification facility contains the equipment to purify both plutonium and uranium isotopes. All these facilities, except the uranium purification facility, which handles uranyl nitrate solutions of subcritically enriched uranium, are subject to criticality safety control. Fig. 5.11 and 5.12 outline the process flow of these facilities.

The logic diagram in Fig. 5.13 summarizes criticality control approaches for devices at the extraction process. The basic strategy for the criticality design opts for geometrical criticality control over the entire plutonium concentration range. This strategy is adopted for all devices situated directly along the plutonium product flow path. The same strategy is adopted for other devices outside of this main flow path when physically possible, but this is not always the case with this large reprocessing facility. In situations where it is not possible to depend on geometrical criticality control, either a batch transfer scheme with Pu-concentration analysis or a continuous transfer scheme with a highly reliable instrumentation must be used to provide an adequate margin of safety against criticality accidents. Specific examples corresponding with [1] – [10] in Fig. 5.13 are presented in Fig. 5.11 and 5.12.

In performing the concentration control at the continuous process with the use of instrumentation, the selection of a concentration limit and a resultant set value by the instrument design is very crucial in view of getting a sufficient safety margin. Below we introduce details of how they relate each other for this reprocessing facility.

In the criticality safety design of this reprocessing plant, the maximum allowable plutonium concentration limit is based on the two criteria which follow below:

- (a) The plutonium concentration must be equivalent to an infinite multiplication factor of 0.95, and
- (b) the plutonium concentration limit should be set based on multiple unit criticality calculations.

Once the maximum allowable limit has been set, a limit for normal operating conditions is determined using the following logic:

- (1) The solution is continuously transported with continuous monitoring of the plutonium concentration by in-line neutron monitors. Case [8]

The concentration limit shall be set such that an appropriate safety margin is obtained between the maximum allowable limit and the concentration limit, taking account of the time required between the detection of an abnormality by the neutron monitor and completing actions such as stopping the process and also taking account of resultant concentration overshooting after the detection of the abnormality.

- (2) The solution is shipped to devices outside of the criticality safety control with continuous monitoring of the plutonium concentration by in-line alpha monitors. Case [4],[10]

Since this is the end point of the criticality safety control area, the concentration limit shall be set significantly lower than the maximum allowable limit despite downstream operations which might increase the concentration.

- (3) The solution is batch transferred after confirming the plutonium concentration by sample analysis. Case [5],[7]

The maximum allowable limit shall be used as the normal operation concentration limit since there is no time constraints associated with this batch transfer, and there are no uncertainty factors other than actual concentration analysis error. Thus, in the actual criticality safety control, we must verify that the measured concentration is below the concentration limit, within the uncertainty limits posed by the analysis error. However, if the solution is shipped to devices outside of the criticality safety control area, as mentioned in (2) above, the concentration limit shall be significantly lower than the maximum allowable limit, and the actual concentration shall be controlled at a sufficiently low level.

- (4) The solution is continuously transported with continuous monitoring of process parameters by in-line flow meter monitors. Case [9]

Plutonium concentration in raffinate where there is no direct control of the plutonium concentration, a range of process parameters shall be set in order to control indirectly the plutonium concentration in raffinate.

Since these safeguards outlined in cases (1) through (4) above depend on preventative measures to keep abnormalities under control, the safety control for each specific case must be designed with redundant features such that any single failure would not compromise the integrity of these safeguards.

References for Chapter 5

- (1) T. Yamamoto and M. Shiota, "A Study of Safety Margins in Criticality Safety assessment of a Dissolving Process," JAERI-M93-218 (1993) (in Japanese)
- (2) S. Tachimori, "EXTRA.M : A Computing Code System for Analysis of the PUREX Process with Mixer-Settlers for Reprocessing," JEARI-1331(1994) (in Japanese)

Table 5.1 Evaluation result of dissolver tank (1)

| PWR fuel : Initial concentration 4.1wt% Solution : Water, 15 degree C | | | | |
|---|-----------------------|-------------------------------|---------------|---------------|
| Fuel composition | Reflector | Neutron multiplication factor | | |
| | | 0 MWd/t | 30GWd/t | 48GWd/t |
| U+Pu | No reflector | 0.8422±0.0048 | 0.7205±0.0039 | 0.6708±0.0036 |
| | 2.5cm Water reflector | 0.9031±0.0039 | 0.7834±0.0042 | 0.7287±0.0037 |
| | 30cm Water reflector | 0.9479±0.0040 | 0.8067±0.0041 | 0.7588±0.0034 |
| U+Pu+FP | No reflector | | 0.6761±0.0036 | 0.6048±0.0032 |
| | 2.5cm Water reflector | | 0.7199±0.0039 | 0.6478±0.0029 |
| | 30cm Water reflector | | 0.7543±0.0033 | 0.6651±0.0031 |
| U+Pu + Actinides | No reflector | | 0.7224±0.0041 | 0.6551±0.0034 |
| | 2.5cm Water reflector | | 0.7682±0.0043 | 0.7120±0.0037 |
| | 30cm Water reflector | | 0.7973±0.0033 | 0.7345±0.0035 |
| U+Pu+FP + Actinides | No reflector | | 0.6655±0.0040 | 0.6001±0.0032 |
| | 2.5cm Water reflector | | 0.7120±0.0034 | 0.6452±0.0030 |
| | 30cm Water reflector | | 0.7433±0.0034 | 0.6674±0.0031 |

Table 5.2 Evaluation result of dissolver tank (2)

| PWR fuel: Initial concentration 4.1wt% Solution : nitric acid 3N, 15degree C, 400g(U+Pu)/L | | | | |
|--|-----------------------|-------------------------------|---------------|---------------|
| Fuel composition | reflector | Neutron multiplication factor | | |
| | | 0 MWd/t | 30GWd/t | 48GWd/t |
| U+Pu | No reflector | 0.7287±0.0047 | 0.6173±0.0038 | 0.5703±0.0030 |
| | 2.5cm Water reflector | 0.8179±0.0041 | 0.6849±0.0036 | 0.6468±0.0030 |
| | 30cm Water reflector | 0.8765±0.0045 | 0.7336±0.0031 | 0.6864±0.0029 |
| U+Pu+FP | No reflector | | 0.5847±0.0036 | 0.5149±0.0028 |
| | 2.5cm Water reflector | | 0.6449±0.0034 | 0.5711±0.0029 |
| | 30cm Water reflector | | 0.6769±0.0036 | 0.6107±0.0031 |
| U+Pu + Actinides | No reflector | | 0.6106±0.0033 | 0.5649±0.0033 |
| | 2.5cm Water reflector | | 0.6913±0.0036 | 0.6288±0.0034 |
| | 30cm Water reflector | | 0.7264±0.0036 | 0.6743±0.0033 |
| U+Pu+FP + Actinides | No reflector | | 0.5735±0.0034 | 0.5106±0.0034 |
| | 2.5cm Water reflector | | 0.6369±0.0032 | 0.5587±0.0031 |
| | 30cm Water reflector | | 0.6774±0.0034 | 0.6004±0.0029 |

Table 5.3 Evaluation result of dissolver tank (3)

| PWR fuel: Initial concentration 4.1wt% Solution : nitric acid 0N, 15degree C, 400g(U+Pu)/L | | | | |
|--|-----------------------|-------------------------------|---------------|---------------|
| Fuel composition | Reflector | Neutron multiplication factor | | |
| | | 0 MWd/t | 30GWd/t | 48GWd/t |
| U+Pu | No reflector | 0.7796±0.0043 | 0.6547±0.0043 | 0.6143±0.0030 |
| | 2.5cm Water reflector | 0.8590±0.0042 | 0.7250±0.0040 | 0.6709±0.0033 |
| | 30cm Water reflector | 0.9167±0.0042 | 0.7705±0.0031 | 0.7158±0.0031 |
| U+Pu+FP | No reflector | | 0.6174±0.0032 | 0.5528±0.0034 |
| | 2.5cm Water reflector | | 0.6829±0.0036 | 0.6078±0.0031 |
| | 30cm Water reflector | | 0.7180±0.0034 | 0.6382±0.0032 |
| U+Pu + Actinides | No reflector | | 0.6524±0.0035 | 0.6022±0.0034 |
| | 2.5cm Water reflector | | 0.7159±0.0038 | 0.6540±0.0033 |
| | 30cm Water reflector | | 0.7642±0.0032 | 0.6990±0.0038 |
| U+Pu+FP + Actinides | No reflector | | 0.6087±0.0034 | 0.5416±0.0027 |
| | 2.5cm Water reflector | | 0.6692±0.0036 | 0.5997±0.0027 |
| | 30cm Water reflector | | 0.7096±0.0031 | 0.6245±0.0027 |

Table 5.4 Evaluation result of dissolver tank (4)

| PWR fuel : Initial concentration 4.1wt% Solution : Water 100 degree C | | | | |
|---|-----------------------|-------------------------------|---------------|---------------|
| Fuel composition | Reflector | Neutron multiplication factor | | |
| | | 0 MWd/t | 30GMWd/t | 48GWd/t |
| U+Pu | No reflector | 0.8169±0.0048 | 0.7035±0.0043 | 0.6516±0.0036 |
| | 2.5cm Water reflector | 0.8883±0.0044 | 0.7612±0.0037 | 0.7134±0.0035 |
| | 30cm Water reflector | 0.9218±0.0044 | 0.7909±0.0033 | 0.7492±0.0038 |
| U+Pu+FP | No reflector | | 0.6534±0.0035 | 0.5825±0.0028 |
| | 2.5cm Water reflector | | 0.7086±0.0033 | 0.6348±0.0029 |
| | 30cm Water reflector | | 0.7411±0.0034 | 0.6633±0.0031 |
| U+Pu + Actinides | No reflector | | 0.6937±0.0040 | 0.6473±0.0038 |
| | 2.5cm Water reflector | | 0.7482±0.0036 | 0.6907±0.0034 |
| | 30cm Water reflector | | 0.7832±0.0033 | 0.7329±0.0031 |
| U+Pu+FP + Actinides | No reflector | | 0.6486±0.0034 | 0.5788±0.0030 |
| | 2.5cm Water reflector | | 0.7020±0.0033 | 0.6258±0.0031 |
| | 30cm Water reflector | | 0.7330±0.0038 | 0.6542±0.0034 |

Table 5.5 Evaluation result of dissolver tank (5)

| PWR fuel: Initial concentration 4.1wt%, Solution : nitric acid3N, 100 degree C, 400g(U+Pu)/L | | | | |
|--|-----------------------|-------------------------------|---------------|---------------|
| Fuel composition | Reflector | Neutron multiplication factor | | |
| | | 0 MWd/t | 30GWd/t | 48GWd/t |
| U+Pu | No reflector | 0.7084±0.0038 | 0.5653±0.0032 | 0.5206±0.0032 |
| | 2.5cm Water reflector | 0.7878±0.0046 | 0.6471±0.0033 | 0.5942±0.0034 |
| | 30cm Water reflector | 0.8485±0.0041 | 0.6948±0.0034 | 0.6406±0.0027 |
| U+Pu+FP | No reflector | / | 0.5207±0.0035 | 0.4740±0.0030 |
| | 2.5cm Water reflector | | 0.5956±0.0031 | 0.5373±0.0026 |
| | 30cm Water reflector | | 0.6451±0.0032 | 0.5762±0.0026 |
| U+Pu + Actinides | No reflector | | 0.5529±0.0034 | 0.5086±0.0029 |
| | 2.5cm Water reflector | | 0.6374±0.0034 | 0.5923±0.0028 |
| | 30cm Water reflector | | 0.6813±0.0031 | 0.6348±0.0032 |
| U+Pu+FP + Actinides | No reflector | | 0.5240±0.0033 | 0.4631±0.0029 |
| | 2.5cm Water reflector | | 0.5920±0.0037 | 0.5225±0.0029 |
| | 30cm Water reflector | | 0.6396±0.0030 | 0.5671±0.0025 |

Table 5.6 Evaluation result of extractor at co-decontamination process

| Fuel | | Before flow change (8 hrs from the experiment start) | | After flow change (max. Pu concentration 50 hrs from the experiment start) | |
|------|------------------------|--|---------------|--|---------------|
| No. | Fuel type | Complete Water reflector | No reflector | Complete Water reflector | No reflector |
| 0 | U- ²³⁹ Pu*) | ----- | ----- | 1.2561±0.0039 | 1.1814±0.0043 |
| 1 | U- ²³⁹ Pu | ----- | ----- | 1.1116±0.0042 | 1.0547±0.0044 |
| 2 | U-Pu | 0.1297±0.0006 | 0.1122±0.0007 | 0.8914±0.0029 | 0.8367±0.0034 |
| 3 | U-Pu-FP | 0.1135±0.0006 | 0.0941±0.0007 | 0.6797±0.0033 | 0.5888±0.0039 |
| 4 | U-Pu-Ac | 0.1201±0.0006 | 0.1009±0.0006 | 0.7149±0.0037 | 0.6289±0.0039 |
| 5 | U-Pu-FP-Ac | 0.1140±0.0006 | 0.0953±0.0007 | 0.6801±0.0031 | 0.5954±0.0040 |

*) ²³⁹Pu is assumed to be uniformly distributed in the entire system.

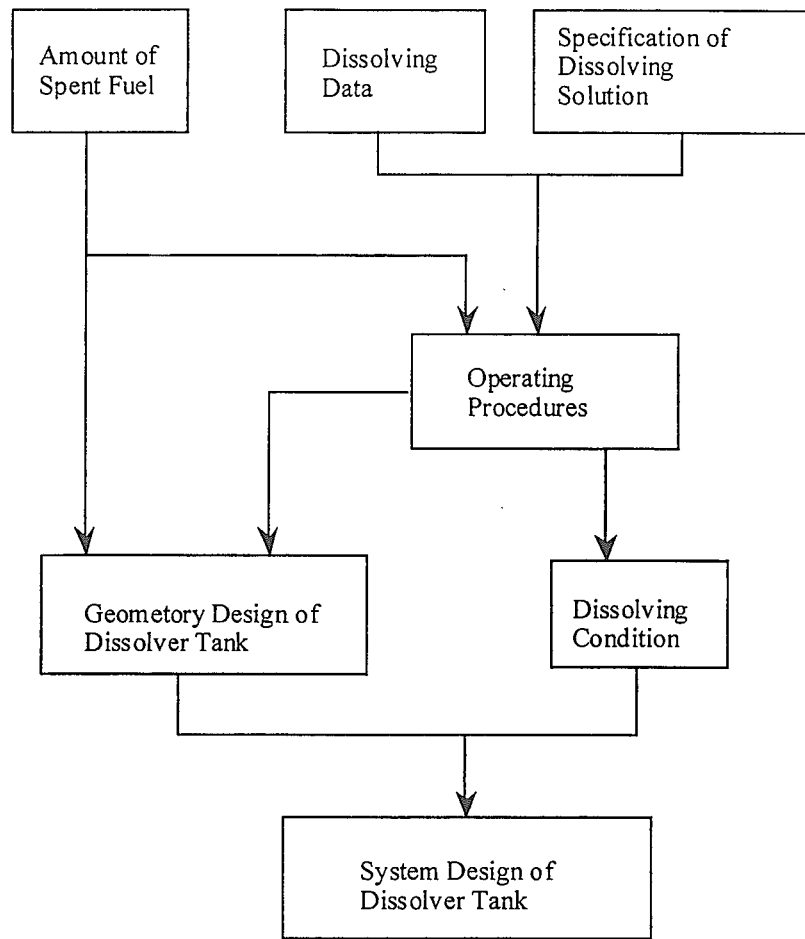


Fig. 5.1 Process design of dissolver tank

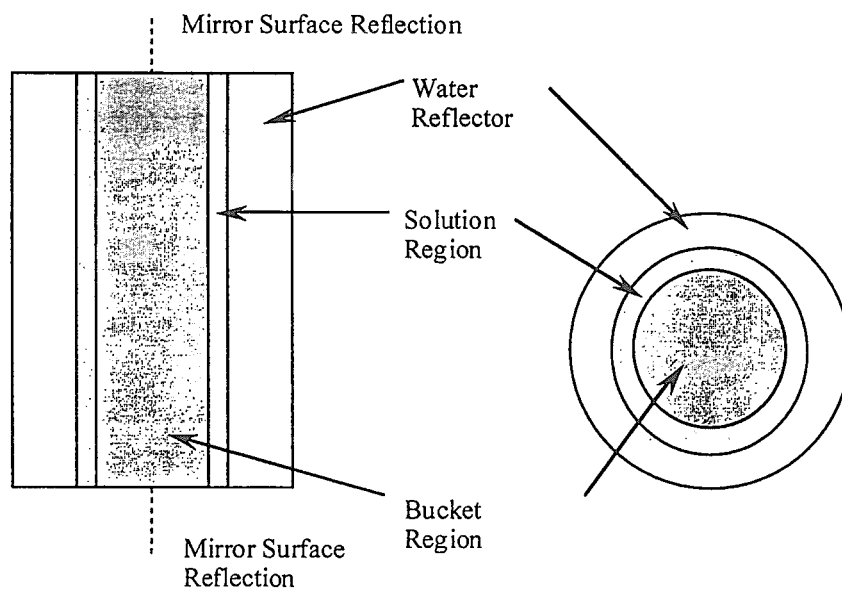


Fig. 5.2 Calculation model of a batch typed dissolver tank

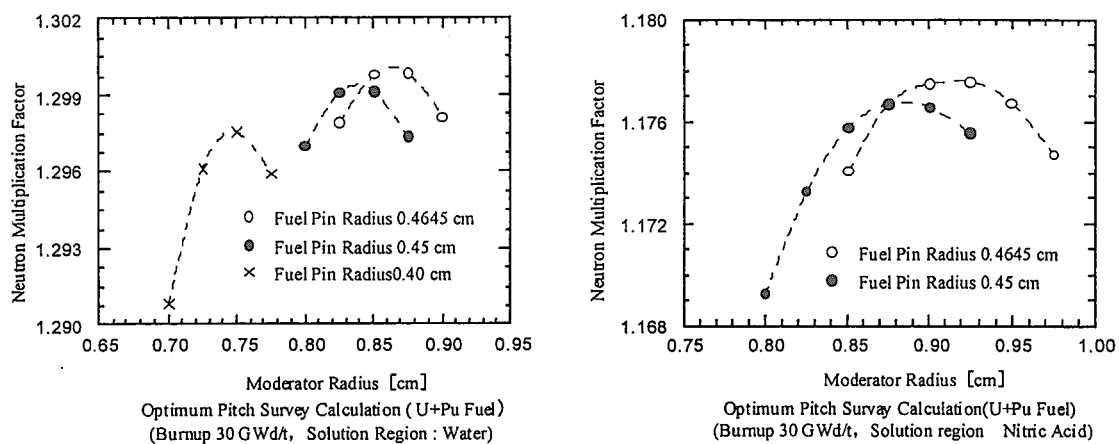


Fig. 5.3 Calculation examples for optimum moderation condition

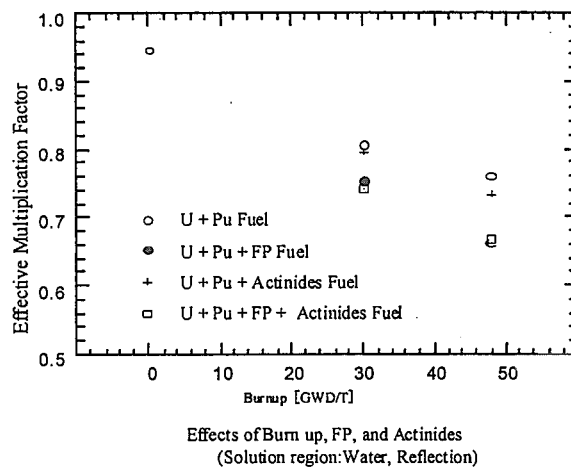


Fig. 5.4 Effect of burnup, FP, and actinides at dissolver tank (example calculation)

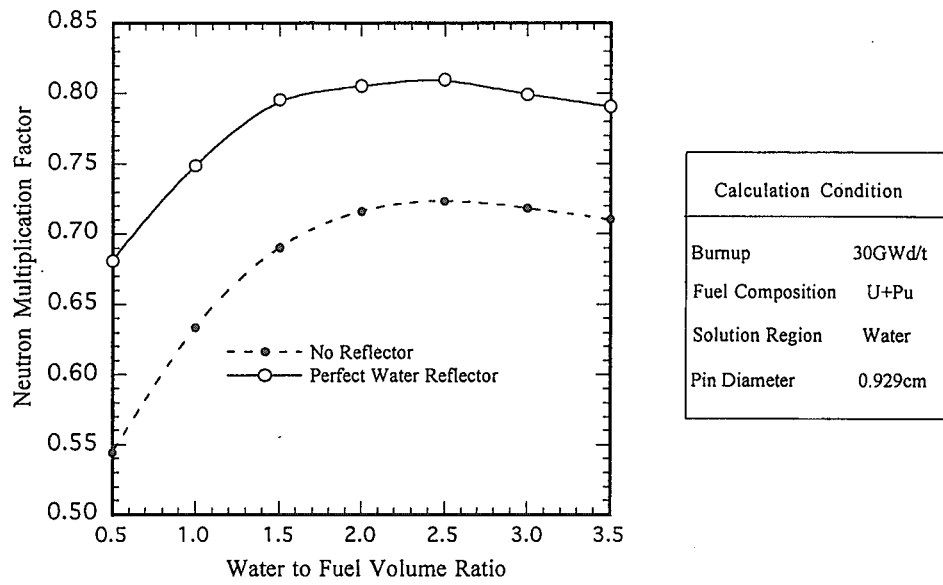


Fig. 5.5 Effective neutron multiplication factor vs. water to fuel volume ratio

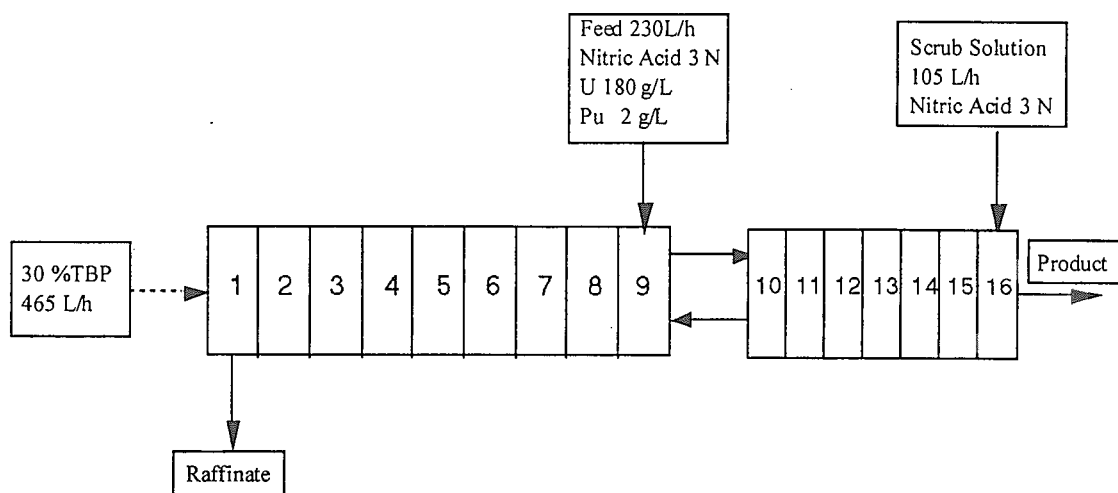


Fig. 5.6 Model of co-decontamination process

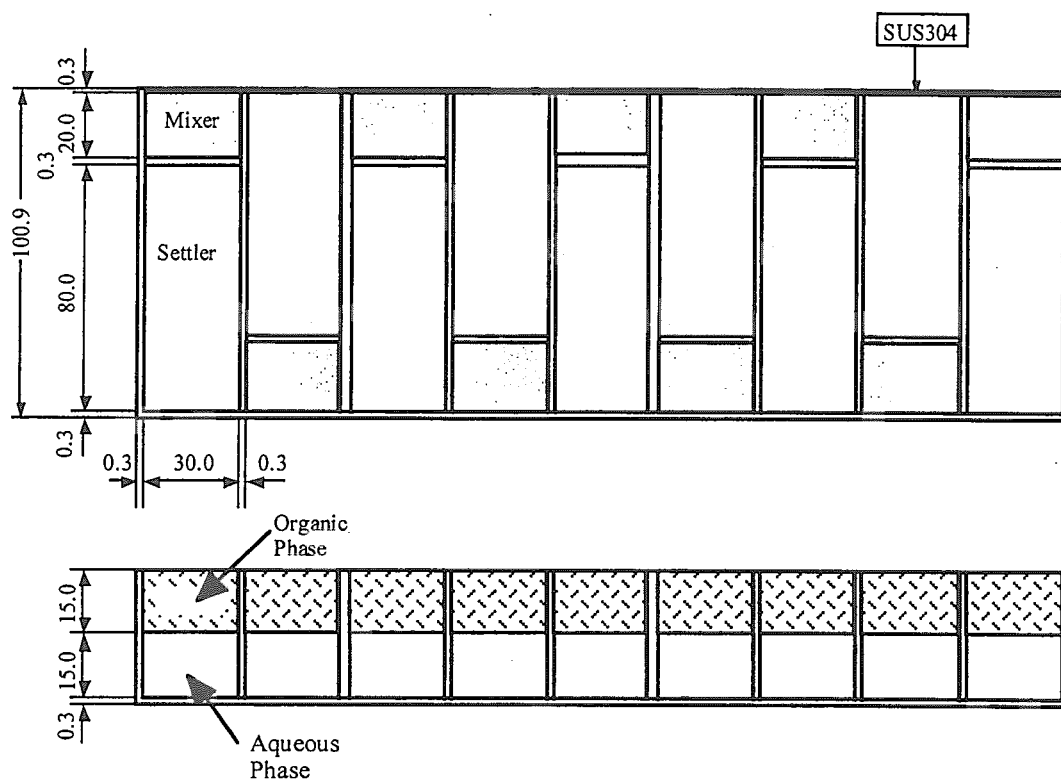


Fig. 5.7 Mixer – settler model

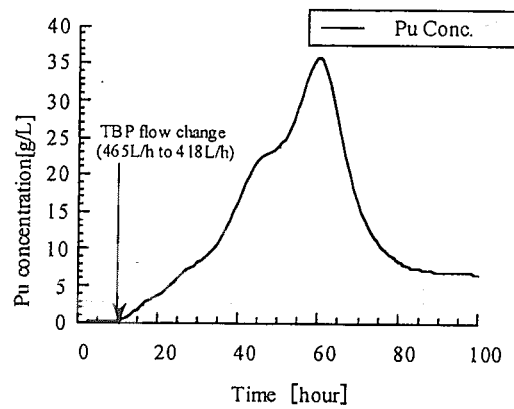


Fig. 5.8 Time variance of the maximum Pu concentration in the process

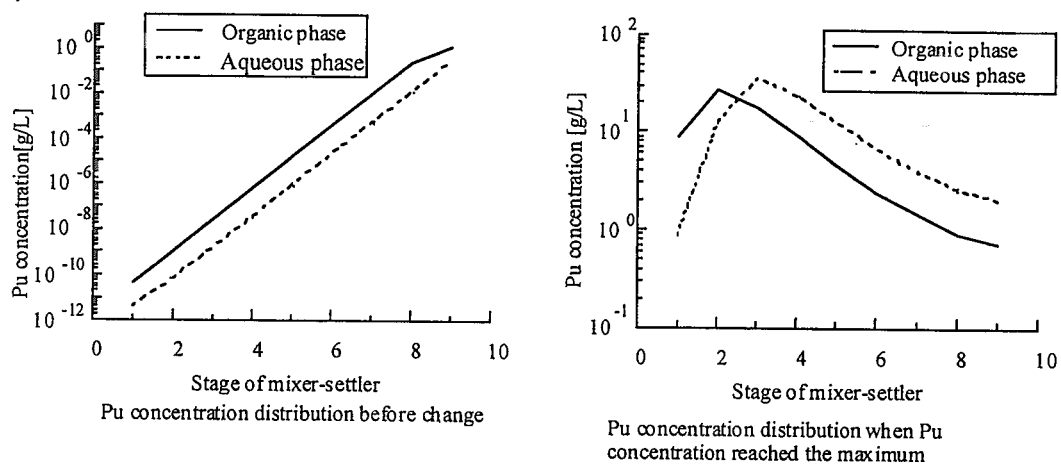


Fig. 5.9 Pu concentration distribution in mixer – settler

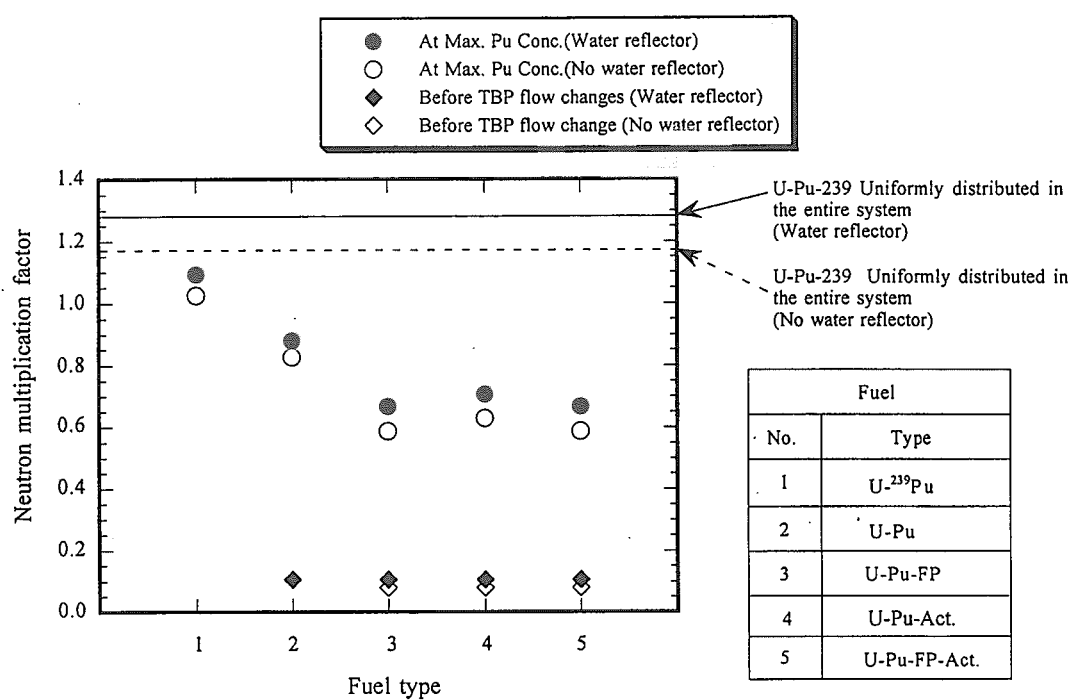


Fig. 5.10 Effects of FP and actinides at co-decontamination extractor (example calculation)

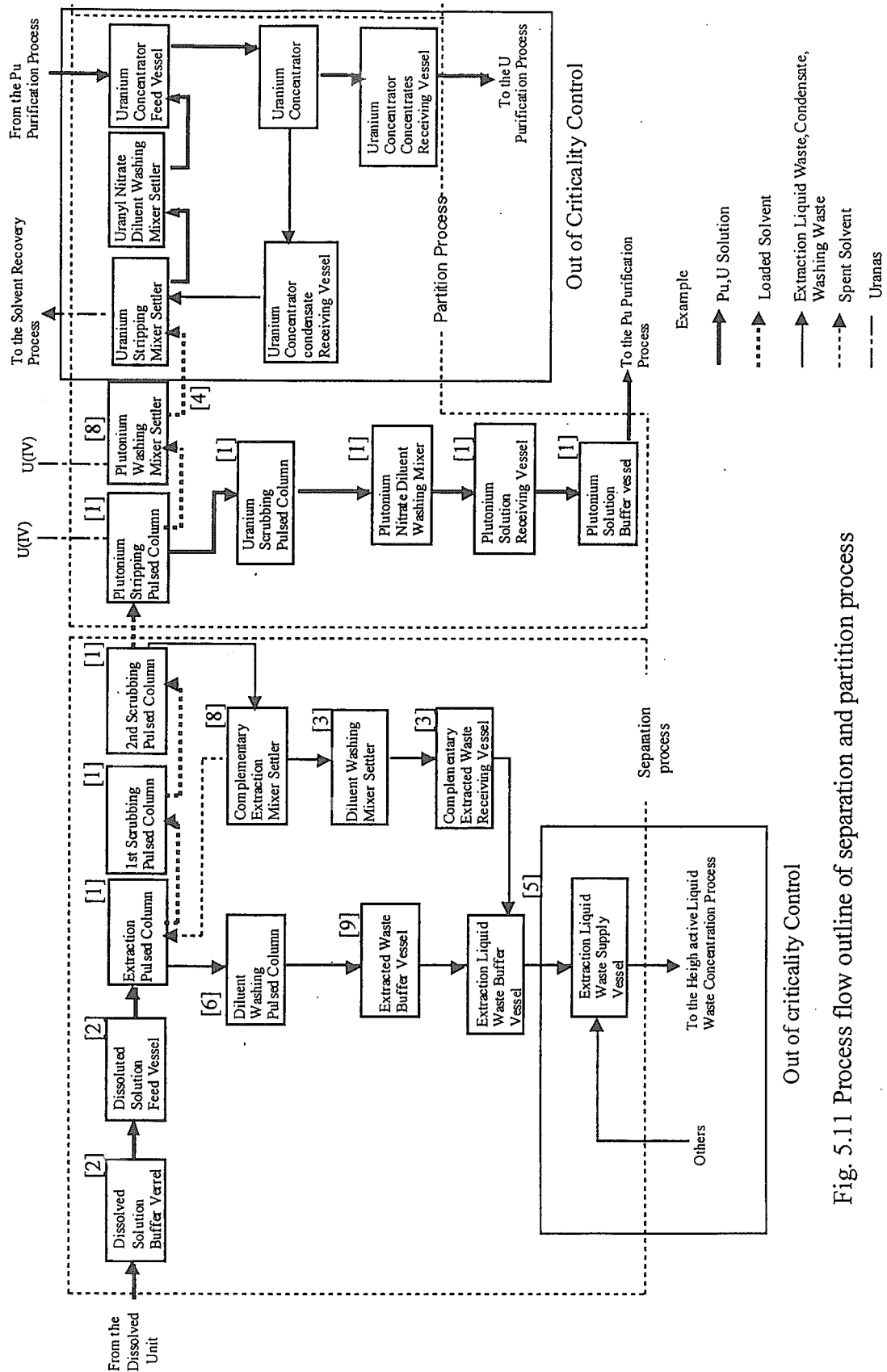
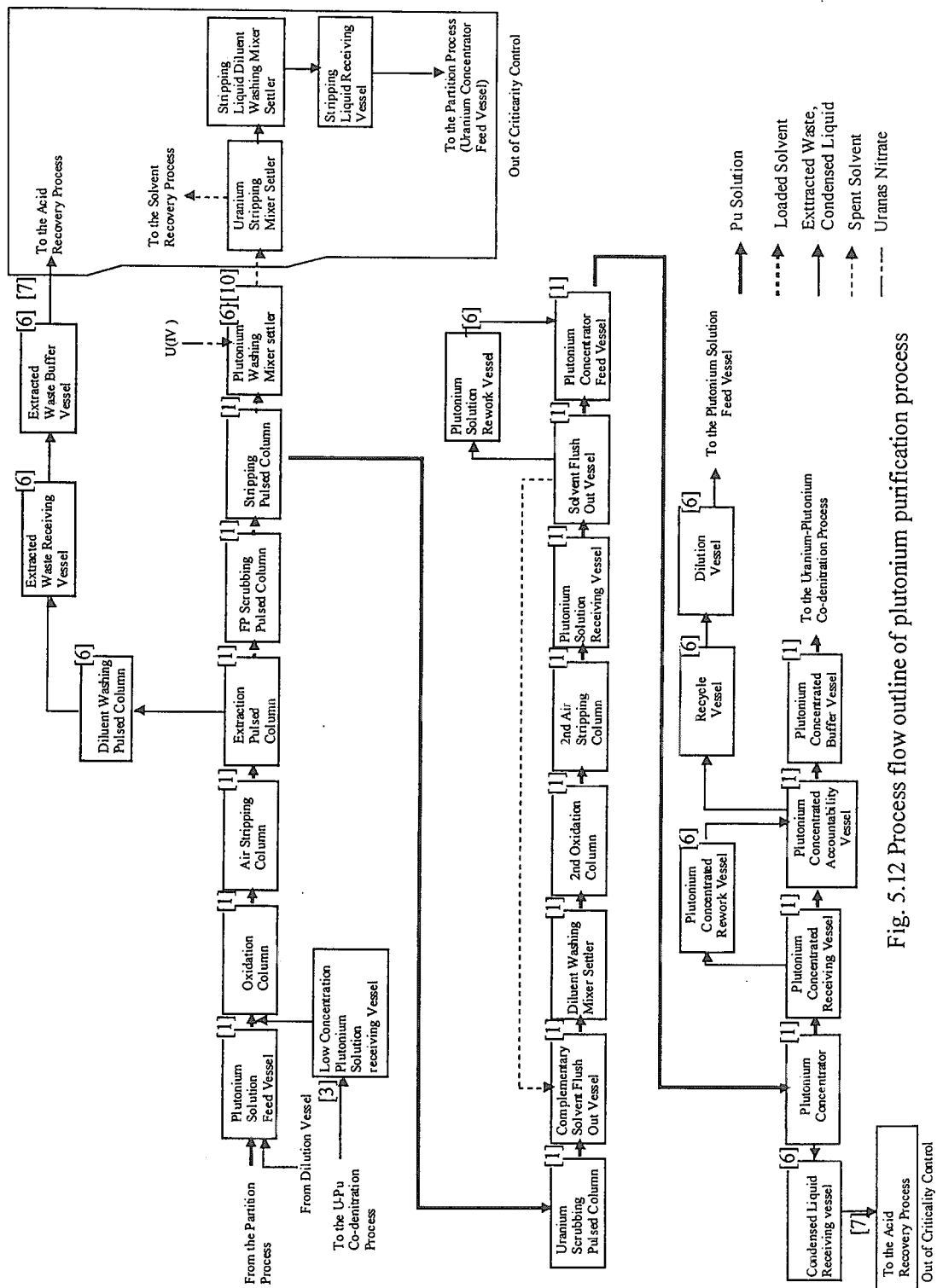


Fig. 5.11 Process flow outline of separation and partition process



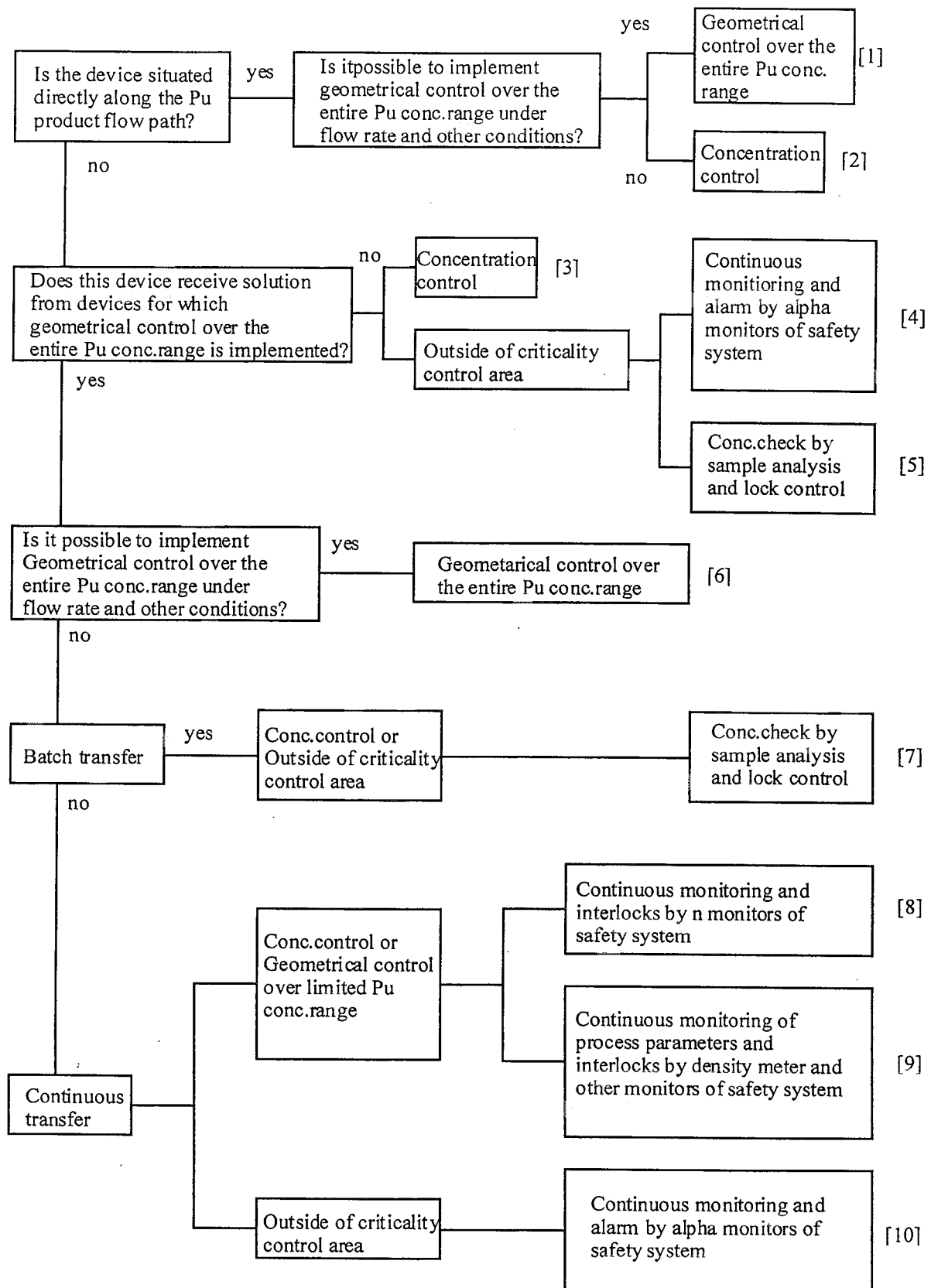


Fig. 5.13 Criticality safety design example at extraction process

6 CRITICALITY ACCIDENTS AND THEIR ASSESSMENT METHODS

6.1 Outline of Criticality Accidents and Simulating Experiments⁽¹⁾

Prior to September 1995, only eight criticality accidents at nuclear fuel facilities (seven accidents in America, one accident in U.K.) had been reported.⁽²⁾⁻⁽⁴⁾ Table 6.1 shows a list of these accidents.⁽⁴⁾ Of the eight accidents, five involved highly enriched uranium fuels, and three remaining accidents involved plutonium fuels. All of these accidents occurred during fuel handling operations. Six of the eight process accidents were concentrated in the seven years between 1958 and 1964, and two accidents occurred in 1970 and 1978. As a result of these accidents, nineteen people received a significant radiation dose; two of the nineteen people died, but none of the accident facilities reported serious damage to the process equipment or any radiological impact on environment.

In September 1995, Russia revealed information about additional criticality accidents which had not been disclosed until the Fifth International Conference on Nuclear Criticality Safety (ICNC95) in Albuquerque, New Mexico, U.S.A.⁽⁵⁾ It outlined twelve criticality accidents, which were summarized in Table 6.2⁽¹⁾. Five accidents involved plutonium fuels, five involved highly-enriched uranium fuels, and the remaining two accidents involved low-enriched (6.5% and 22.6%) uranium fuels. In another aspect, eleven accidents involved water- (or hydrogen-) moderated systems and one accident related with (Pu) metal system. These accidents resulted in a total of five personnel deaths and eight serious injuries or radiation damages due to a significant amount of radiation exposure. No damage was inflicted on the process equipment. Of the twelve accidents, ten accidents are considered as accidents at nuclear fuel facilities, while one accident occurred at a criticality experiment facility and another one while handling plutonium metal. These additional Russian accidents more than doubled the number of reported nuclear fuel facility criticality accidents, raising the total from 8 to 18 nuclear fuel facility criticality accidents worldwide.

All of the above occurred while handling fuel solutions or combined fuels of solution and solid (e.g., precipitates). When a certain amount of reactivity is inadvertently inserted to a solution nuclear fuel system, the system becomes supercritical, causing the nuclear power and the radiation dose rate increases. As a result, such phenomena occur as solution temperature increases, radiolytic gas void formation, and steam void formation after the onset of solution boiling. Possible direct causes for the excursion may be such events as fuel solution transfer and transfer or agitation of the extracted solvent, but each of these events occurs as a result of combinations of various fundamental causes. Eventually, criticality terminates due to inherent sources of negative reactivity. There are various specific reasons for termination: temperature increases, void formation, dilution of the fissile solution, concentration decreases, solution transfer, splash losses, and precipitation product formation.

One of the French criticality accident simulation experiments CRAC, shown in Fig. 6.1, is a good example of power excursion during a criticality accident.⁽⁶⁾ As this figure shows, the fuel

solution criticality event consists of three stages.

[1] First burst stage

When the prompt critical reactivity is exceeded due to an inserted reactivity, a power excursion, or burst, occurs in a short period. The power increases sharply until the negative reactivity due to temperature increase halts the power surge, and radiolytic gas voids thereafter sharply drops the power. When the reactivity insertion is below that is necessary for making prompt critical, the power rises gradually and the possibility of a burst is avoided.

[2] Power oscillation stage

If the system reactivity exceeds prompt critical conditions, an oscillating criticality situation is created after the first burst stage in [1]. As the radiolytic voids escape the solution surface, the negative void reactivity decreases. This causes the power to ramp as the system begins to re-approach prompt critical. This power ramp in turn creates new gas voids, increasing the negative reactivity insertion, and causing the power to drop. As this cycle repeats, the power oscillates. These power oscillations continue till the inserted reactivity effect is offset by solution temperature rise or steam void formation due to boiling.

[3] Slow power decrease stage

As the inserted reactivity is offset by continuous feedback effect, the system becomes slightly subcritical, and the power decreases gradually.

France and the United States have been conducting criticality accident simulation experiments for several years. In 1996, Japan also began conducting criticality accident simulations. The experiments conducted in each of these countries to date is outlined below:

(a) France

France uses the highly enriched uranyl nitrate fueled reactors CRAC⁽⁶⁾ and SILENE⁽⁷⁾ at the Institut de Protection et de Sûreté Nucléaire (IPSN) Valduc Laboratory of Commissariat à l'Énergie Atomique (CEA) to conduct its criticality accident simulation experiments.

The CRAC experiments have been completed and a lot of data have been published. In this facility, experiments on nuclear behavior (kinetics) of solution nuclear fuels at criticality accidents have been conducted, and such data as power, energy, temperature, pressure, and gamma ray dose rate have been measured as a function of time. These experimental data have been used to validate the analysis and evaluation methods for criticality accidents.

A limited amount of data from the SILENE reactor has been presented at international conferences. This reactor has been primarily used as a pulse radiation source for testing criticality alarm devices and biological irradiation effects. Although details are unknown, it has also been

reported that SILENE has been used to measure radiolytic void gas formation data.

(b) The United States of America

During the second half of the 1950's, the United States conducted kinetics experiments for boiling water nuclear reactors using the KEWB reactors at Atomic International.⁽⁸⁾ Although these experiments were not designed for criticality accident research, important criticality data on the mechanisms of radiolytic gas formation in fuel solutions was gathered.

More recently, the SHEBA⁽⁹⁾ reactor at Los Alamos National Laboratory (LANL), has been used to conduct criticality accident simulation experiments with low-enriched uranyl fluoride solutions. SHEBA has been used to study slow accident events under low reactivity insertion conditions (below prompt criticality) as well as to test criticality alarm device performance. Future experiments plan to focus on prompt critical simulations. However, very few experiment data of the SHEBA experiments have been published.

(c) Japan

In 1996, Japan started criticality accident simulation experiments at the Japan Atomic Energy Research Institute (JAERI) using uranyl nitrate solutions in TRACY^{(1),(10),(11)}. Unlike the French experiments, these experiments use low-enrichment uranium and are intended to further the understanding of the accident events of low-enriched uranium system, of which the kinetics constants are different from that of high-enriched uranium systems. In these experiments, the radiolytic gas void production as a function of time, of which not much data have been gathered worldwide, will be measured for better understanding of the behavior of power excursions. In addition, the transfer rates of fission products from the core tank to the vent system will be measured in order to accumulate the data necessary to analyze the radiation environment during criticality accidents.

Note that all the experiments (a)-(c) above deal with fuel solution systems. Experimental data are available for other systems such as heterogeneous systems (e.g., dissolver tank) and powder fuel systems. France has conducted experiments on a powder fuel system at the IPSN, where data were measured as water penetrates the UO₂ powder fuel⁽¹²⁾, however no neutron behavior simulations have been conducted.

6.2 Methodology for Assessing a Criticality Accident

Safety design at nuclear fuel facilities is required to include appropriately the concept of defense in depth. A safety system evaluation shall be conducted based on postulated abnormalities and accidents prior to the installation of any nuclear fuel facility. According to *the Regulatory Guide for Licensing Safety Review of Fuel Fabrication Processing Facilities*, any facility dealing with low-enriched uranium (less than 5 %) is not required to evaluate criticality accidents in the safety

assessment as long as the criticality safety design satisfies all regulatory guide requirements. In contrast, *Regulatory Guides for the Licensing Safety Review of Reprocessing Facilities* specify the criticality of nuclear fuel material as an example to be included in the safety assessment. Therefore, when installing “Facilities for which one needs to consider measures for preventing criticality accidents” under section 2.2, we need to carry out safety evaluation on criticality accidents including whether the postulated regulatory accidents are required or not.

When criticality accident scenarios are included in the safety assessment, the following two methods are used depending on the type of facility under consideration.

The objective of the safety assessment is to evaluate the radiation exposure to the public living near the facility. In case of criticality accidents, noble gases and iodine are major sources of the radiation exposure. The first method of defining a criticality accident scenario in the safety assessment is to select a criticality accident without specifying a particular process, or when specifying a particular process, to select a criticality accident that envelopes the entire facility without taking account of specifications of the particular process. The second method is to hypothesize possible abnormal events at “each process of the reprocessing facility where radioactive materials exist” and select design basis events “in relation to the probability of occurrence,” as stated by *Safety Review Guidelines for Reprocessing Facilities*. In this case, since a specific process is considered in selecting an accident event, the safety design specifications of the process is often reflected upon the selected accident scenario.

As mentioned above, the magnitude of radiation exposure of the public at criticality accidents, main sources being noble gases and iodine, depends on the severity of the postulated criticality accident, that is, the total number of fissions. When selecting an accident scenario for a particular process, the expected process-specific reliability must be carefully considered when estimating the number of fissions associated with the accident scenario. When selecting an accident scenario enveloping the entire facility, the number of fissions must be estimated conservatively by using past criticality accident data, data from criticality accident simulation experiments, and regulatory guides based on these accidents and experiment data, while fully analyzing the applicability of these data to the process under study.

Table 6.3⁽¹⁴⁾⁻⁽¹⁸⁾ shows examples of criticality accident scenarios used in the safety analyses of several reprocessing facilities. Subsequent sections of this report introduce simplified analysis formula and criticality accident analysis codes for estimating the total fissions in a criticality accident.

6.3 Estimation of Scale of Criticality Accident by Simplified Evaluation Models

6.3.1 Derivation of Simplified Models and Parameter Studies⁽¹⁹⁾

When a sufficient amount of reactivity is added to a solution system containing fissile materials such as uranium and plutonium, the system becomes supercritical. When the added reactivity is

relatively small, the majority of produced energy is consumed for heating up the solution, and generally the solution does not boil and the reaction terminates eventually due to the temperature negative reactivity coefficient effect. The total fissions F can be given from reactivity balance model, using both the one-point and adiabatic approximations.

$$F = 3.3 \times 10^{10} \cdot C_p \cdot \rho_{\text{add}} \cdot |1/\alpha_T| \cdot V_s \cdot d_s \quad (6.1)$$

where

| | |
|---------------------|---|
| C_p | :specific heat (J/(°C·g)) |
| ρ_{add} | :added reactivity ($\Delta k/k$) |
| α_T | :reactivity temperature coefficient ($\Delta k/k/^\circ\text{C}$) |
| V_s | :solution volume (cm^3) |
| d_s | :solution density (g/cm^3). |

The solution composition terms in Equation (6.1) can be combined into a single factor denoted by Θ , where

$$\Theta = [3.3 \times 10^{10} \cdot C_p \cdot d_s / |\alpha_T|]^{-1} = 3 \times 10^{-11} \cdot |\alpha_T| / C_p / d_s$$

Equation (6.1) can now be expressed as

$$F = \rho_{\text{add}} \cdot V_s / \Theta \quad (6.2)$$

Fig. 6.2 shows a correlation between ρ_{add} and V_s of various fuel compositions parameterized in Θ , assuming that total number of fissions (e.g., $F = 10^{19}$ fissions) is constant.

Since Equation (6.2) is used when the temperature rise by the fission energy is not enough to boil the solution, this equation can be simplified by adopting the temperature rise ΔT instead of addressing the added reactivity directly. In this way, the temperature application range of this equation can be clearly identified. By inserting $\Delta T = \rho_{\text{add}} / |\alpha_T|$ into Equation (6.1), using an assumed temperature rise range from room temperature (20°C) to boiling temperature (110°C), and selecting values of other parameters conservatively within an actual application range such that calculated total fissions will be on a safe side, we obtain

$$F_1(90^\circ\text{C}) = 2.6 \times 10^{13} \cdot V_s \quad (6.3)$$

The continuous void production which occurs when boiling commences, causes a rapid decline in power. When this process continues, the fission reaction will terminate after a relatively long period due to decrease in water level by evaporation of solution or moderator (water) losses. In reality, however, criticality monitors would alert system operators for an accident situation, prompting

them to terminate the reaction forcefully by taking corrective measures such as solution damping, dilution, and poison addition. Should abrupt boiling occur, the reaction would also quickly self-terminate due to the large splash losses. Accounting for both the self-termination and the operator induced termination processes during an actual accident allows us to assume that even a large excess reactivity insertion fission reaction would terminate quickly following the onset of boiling. Under conditions where all of the above assumptions apply, Equation (6.3) provides a conservative estimate of the total number of fissions independent of individual accident scenarios. For example, the total number of fissions for critical systems and devices which are continuously monitored and controlled and have built-in corrective actions against potential criticality accidents can be calculated from Equation (6.3).

In systems and devices, where the boiling continues for a long time during an excursion and as a result, the solution condenses by evaporation, it is considered sufficient to calculate the total fissions from Equation (6.4).

$$F_2 = 6 \times 10^{13} \cdot V_s \quad (6.4)$$

6.3.2 Comparison of Simplified Models with CRAC Experiments and Data on Past Criticality Accidents Occurred in Solution Systems

In addition to the equation above, several different simple formulas have been proposed for the calculation of the total number of fissions for a criticality accident. Some representative equations are introduced below.

Tuck equation⁽²⁰⁾: $F = V \times 10^{17}$

This equation assumes the event terminates by solution evaporation. It represents the energy necessary to evaporate a volume, V (in liters), with an assumption that about 20% of the heat is lost to the environment. If additional solution is fed to the tank in the course of the event, then this volume should be included in the volume V . Note that the factor in this equation is 1.7 times larger than that of Equation (6.4).

Olsen equation⁽²¹⁾: $F = 2.95 \times 10^{15} \cdot V_B^{0.82} + 3.2 \times 10^{18} \cdot (1 - t^{-0.15})$

The Olsen equation expressed total fissions as the integrated sum of the burst and slow power decrease stage fissions. This equation is applicable to a vessel diameter of 300-800mm and a solution feed rate of 0.027-0.52 L/sec ranged in the CRAC experiment. V_B is the fuel solution volume in liters at the burst stage, and t is duration time (sec.) of the slow power decrease stage.

Barby equation⁽²²⁾: $F = V \cdot t / (3.55 \times 10^{-15} + 6.38 \times 10^{-17} \cdot t)$

Barbry, in this equation, described a simple empirical relation deduced by the most severe CRAC and SILENE experiments. V is volume in liters of the fuel solution, t is duration time (sec), and applicable to a duration time less than 600sec. This equation is applicable to homogeneous high-enriched uranium systems with a neutron source, and also applicable to plutonium systems. The solution concentration ranged between 20 and 360gU/L, and cylindrical vessels with 30cm and 80cm (CRAC), and with 36cm (SILENE) in diameter were used.

The simple analysis equations (6.3) and (6.4) in the previous section for calculating the total fissions, as it is obvious from the derivation, it is considered to be generally applicable to fuel solution systems with negative reactivity coefficients, irrespective of the fuel type of uranium or plutonium, concentration, composition, enrichment, nitric acid concentration and vessel shape and size.

Fig. 6.3 compares the calculated total fission values from the simple analysis equations, the CRAC experiment data⁽²³⁾, and the historical criticality accident data occurred in solution system⁽²¹⁾ that were summarized by Olsen et al.

6.4 Development of Computer Codes for Analyzing a Criticality Accident

6.4.1 Present Status of Development

The United Kingdom, Germany, the United States of America, and Japan are developing criticality accident analysis codes based on the CRAC and SILENE data of highly enriched U. The development status of each country is described below.

(a) United Kingdom

The United Kingdom Atomic Energy Authority (UKAEA, currently AEA Technology) developed a criticality accident analysis code CRITEX⁽²⁴⁾ in collaboration with the French IPSN, based on the CRAC and SILENE experiment data. This code uses the point reactor kinetics approximation with radiolytic gas feedback effects being incorporated. Parameters needed for calculating radiolytic gas production (volume) were derived from the CRAC and SILENE experiment data. The CRITEX code has been benchmarked many times against the CRAC and SILENE data, and all of the calculations showed good agreement with the experimental results. The IPSN conducted experiments on powder UO_2 , where the wetness was measured as water penetrates the powder, and UKAEA carried out the criticality analysis on the powder fuel based on the experiment data⁽²⁵⁾.

Imperial College has recently developed a finite element analysis code, FETCH, which is a combination of three-dimensional, time-dependent neutron transport codes and thermal fluid dynamics code, and has conducted benchmarking calculations against the CRAC and SILENE data⁽²⁶⁾.

(b) Germany

In Germany, two accident analysis codes using two different approaches have been developed. The first is a detailed analysis code FELIX⁽²⁷⁾ developed by Gesellschaft für Anlagen-und Reaktorsicherheit (GRS) mbH. This code solves a time dependent one-dimensional neutron transport equation, which incorporates the negative reactivity feedback effect of the fuel solution systems. Radiolytic gas void parameters are based on the CRAC experiment data. Calculations using this code show good agreement with the CRAC experiment data in many benchmarking analyses.

Dornier System developed a code⁽²⁸⁾ to calculate average power as a function of time without accounting for power oscillations from radiolytic gas voids, which is considered by the FELIX. This code is based on a quasi-steady state modeling. The behavior of radiolytic gas voids is analyzed through a combination of basic empirical equations and parameters from the CRAC experiment data. Some CRAC experiment analyses have been conducted and calculations by the code agreed well with the experimental data.

(c) The United States of America

University of Arizona in the United States of America developed a code by combining state equations of fuel solution including radiolytic gas voids, energy balance equations, and point reactor kinetics equations.⁽²⁹⁾ This code can calculate power and pressure changes at the first burst of an accident, but cannot calculate power oscillations since void movements and surface releases are not considered. Parameters needed for calculating voids were derived from the KEWB and CRAC experiment data. Calculated data by this code showed good agreements with the CRAC and SILENE experiment data. The current version of this code models the fuel as single region, but a new version, which models multiple fuel regions, is being developed. In addition, University of Tennessee has conducted criticality accident analyses of the multiple unit fuel solution based on the point reactor kinetics approximation.⁽³⁰⁾

(d) Japan

Japan developed criticality accident analysis codes AGNES^{(31),(32)} and CREST⁽³³⁾ based on point reactor kinetics equations incorporating a radiolytic gas void model. Void parameters used in these code are ones deduced from the CRAC experimental data. In addition, based on the void model of the AGNES, JAERI developed a code TRACE which handles in more detail thermal fluid-dynamics of the solution.⁽³⁴⁾

The outline of the AGNES and CREST is discussed in the next section.

6.4.2 Outline of AGNES and CREST Codes⁽³¹⁾⁻⁽³³⁾

Both codes are based on a combination of the single channel thermal fluid-dynamics analysis

model and the nuclear model by the point reactor approximation kinetics equations (delayed neutron 6 group approximation). These codes account for the reactivity feedback effects of solution temperature and void volume changes. For production of radiolytic gas voids, these codes assume that only those gasses above a certain threshold value grow to voids, while those gasses below the threshold dissolve in the solution.

The AGNES code, developed by JAERI, uses the "pressure model" and the "modified energy model" to calculate the radiolytic gas volume. The "pressure model" calculates radiolytic gas void volume based on pressure changes (empirical equation) occurring at the time of radiolytic gas formation in the first burst stage. This model, however, does not take account of the movement and release of voids from surface, and cannot calculate the gas volume during the power oscillation stage or anytime later. Thus, by modifying the "energy model" that calculates the void volume based on generated energy, the "modified energy model" was developed. This model treats the void production and disappearance microscopically and can calculate formation, movement, and release of voids from surface in separated regions. The code with the "pressure model" shows good agreements with the first power burst of the KEWB and CRAC experiment (Fig. 6.4). Also, the "modified energy model" was used to simulate the power oscillation stage of the CRAC experiment, and the calculations show good agreements with the experiment data (Fig. 6.5).

The CREST code, developed by Mitsubishi Atomic Power Industries, Inc.(now, Mitsubishi Heavy Industries Corporation), assumes that the radiolytic gas void production rate is proportional to the fission rate, and this code accounts for steam voids produced at the boiling temperature. This code calculates the void volume by assuming that each new void is released from the solution surface with a first order lag. The lag time constant is the time required for the void to travel one half of the solution height. The code was used to simulate the long time period till the termination of criticality experiments at the CRAC, and the calculations show good agreement with the experiment data (Fig. 6.6).

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Table 6.1 Criticality accidents at nuclear fuel facilities in USA and Europe

| Location Date | Cause of criticality accident & total fissions | Fuel inventory & criticality situation | Alarm & Evacuation | Dose rate at site | Criticality monitor type & alarm set value | Dose |
|--|---|---|--|--|--|--|
| ORNL Y-12 (Oak Ridge, Tenn.) 1958.6.16 | Enriched U solution was combined with water for leak testing in a 208 L drum. 1.3×10^{18} | Became critical by ^{235}U 2.1 kg in U solution of 56.2 L, power spike 10^{16} , plateau power 1.29×10^8 (about 3 min.), duration 18 min, no shielding. | 8 persons were at site. Alarm was activated, and promptly evacuated. | 100mR/hr at 107m from site. 3hrs from accident, 60mR/hr at 30.5m from drum. | GM type. 8 monitors about 24.4m from center of plant. Alarm set at 1mR/hr. Changed to 3mR/hr after accident. | 8 persons irradiated at 28-461 rem |
| LASL (Los Alamos, N.Mexico) 1958.12.30 | At Pu recovery process, to measure Pu content, Pu containing solution was transferred to tank. Stirrer changed geometry to supercritical. 1.5×10^{17} | 96.5cm ϕ , 850 L tank, Pu3.27kg in 160 L Pu solution, Pu distributed to solvent, became critical when agitated with stirrer. Power spike 1.5×10^{17} , duration 2 sec, no shielding. | Alarm activated instantly. All personnel evacuated. Two persons nearby conducted rescue operation. | Right after accident, alarm 53.3m from site was activated. 20R/hr at 7.6m from tank. | Ionization chamber type, alarm activated with 2 or more signals, error calibrated with internal source. Alarm set at 1 and 10 mR/hr. | 1 person irradiated at 12000 rem and died. Others 53-134 rem |
| ICPP (Idaho Falls, Idaho) 1959.10.16 | Air-sparging operation for sampling initiated, siphoning action and transferred uranyl nitrate solution containing enriched U to nonsafe geometry tank. 4×10^{19} | 200 L of solution containing $170\text{g } ^{235}\text{U}$ siphoned to 18900 L tank. Total solution of 800 L and ^{235}U 34.0kg reached criticality. Power spike 10^{17} , shielded operation. | Airborne fission products passed through vent line and contaminated building. Radiation alarm was activated. Evacuation orders were communicated via telephone and verbal command. | Above 5R/hr outside of building at evacuation site, and 119m west from entrance (due to radioactive gas). | Criticality alarm device with 20 ionization chambers was installed. Alarm set at 20 mR/hr. | 11 persons irradiated at 2-50 rem |
| ICPP (Idaho Falls, Idaho) 1961.1.25 | High-pressure air was used to unplug a clogged pump pipe, forcing solution into vapor-disengagement cylinder of nonsafe geometry. 6×10^{17} | Top diameter of vapor-disengagement cylinder was 61cm (non-favorable geometry). 40 L of U solution containing $8\text{kg } ^{235}\text{U}$ was forced into cylinder. Power spike 6×10^{17} , shielded operation. | Radiation alarm was activated instantly, and evacuation alarm was activated manually. | Dose rate in building right after excursion was background. 30mR/hr at a downwind point due to radioactive fume. | Same as above. | Gamma ray dose only. 0-55 mrem |
| Recuplex (Richland, Wash.) 1962.4.7 | While cleaning up the facility, Pu solution in floor sump was vacuum transferred into an nonsafe geometry. 8×10^{17} | 69 L capacity cylinder of 46cm in diameter. 46 L of Pu solution containing 1.5kg Pu caused criticality. Power spike 10^{16} , duration 37hr, no shielding. | Radiation alarm was activated. Evacuation was prompt. Distance from site to nearby personnel was 1.5-8m. | Dose rate of accident is not known. 1200Ci of noble gas were released from stack. | Criticality monitor consisting of 10 NaI detectors was available in 234-5 building. Alarm set at 500mR/hr. | 3 persons 19-110 rem |
| UNC Wood River Junction 1964.7.24 | Containers were not correctly labeled. Enriched U solution was poured into a makeup tank of nonsafe geometry. 1.2×10^{17} | Cylinder 45.7cm in diameter, 66cm in depth. 10 L solution containing ^{235}U 2.6kg was poured into 41L of 0.54M Na_2CO_3 . Power spike 10^{17} , no shielding. | Alarm activation is not known. Operators evacuated after they observed a flash of blue-white light and splashing of solution. | Full scale on max. 100mR/hr radiation monitor right after accident. Detail dose rates are not known. | - | 1 person died. Pelvis 46000 rem Head 14000 rem |
| U.K.AEA Windscale 1970.8.24 | At a Pu recovery process, solution was transferred to a tank of nonsafe geometry. 1×10^{15} | Tank 61cm in diameter, 68.6cm in depth. 50 L solution with 55gPu/L and 6-7gPu/L. Power spike 10^{15} , duration 5-10sec, shielded operation. | Radiation alarm was activated. Evacuation was prompt. Personnel received dose check. | 200mR/hr in control area 10 min after accident. About 5mCi was released from stack. | - | 2 persons 2 rad or less. |
| ICPP (Idaho Falls, Idaho) 1978.10.17 | A drop in aluminum nitrate concentration caused an increase in U concentration. 2.74×10^{18} | Aluminum nitrate concentration at first cycle scrub column of solvent extraction process dropped. When U concentration increased to 22.2gU/L, system became critical. Delayed critical. Shielded operation. | - | - | - | None. |

Table 6. 2 Criticality accidents in Russia (1/2)

| No. | Date Place | Situation Cause | Quenching mechanism | Number of fissions | Dose | Others |
|-----|---|--|--|--|--|---|
| 1 | 1953.3.15 Mayak Enterprise, Urals Pu solution receiving tank | Transferred Pu nitric acid solution stored in two tanks in the cell to one tank outside the cell. Intended amount of the transfer was 26 L (650gPu), but the actual amount was 31 L. | Returned solution to the original tanks. | Burst 2.5×10^{17} | 1 person 1000rad (seriously rad damaged). 1 person 100rad | No radiation monitor. No in-advance instructions to personnel. No accident drills. No post-accident report. |
| 2 | 1957.4.21 Mayak Enterprise, Urals U purification vessel | 3.4kg of precipitates of high-enriched U-Oxalate accumulated in the vessel. Not known when driven to criticality. Operator found that filter was ballooning and gas was generated from the precipitates. | Discharged some solution to vacuum trap. | 2×10^{17} | 1 person died 12 days later. 5 person some rad. damaged | No radiation monitor. No periodic clean out. |
| 3 | 1958.1.2 Mayak Enterprise, Urals Criticality experiment device | After experiment with high-enriched U solution , fixing bolts were disconnected and the tank was tilted to discharge the solution, which caused criticality. 3 operators functioned as reflectors. | Splashing. | Spike 2.3×10^{17} | 3 persons died 5-6 days later. 1 person seriously rad. damaged and lost eyesights. (3m from the tank) | No compliance with rules. Not sufficient safety measures. The device was dismantled. |
| 4 | 1960.12.5 Mayak Enterprise, Urals Pu solution filter vessel | Precipitates containing 170g Pu accumulated in unfavorable geometry vessel. Solution including 830g of Pu was poured. Following first pulse, solution was pushed back to pipe and criticality terminated, but when vacuum system was shut down as emergency action, solution moved to vessel again, which caused second criticality. | | Total 10^{17} | Several persons Max. 5rad | Alarm system available. Poor recording of the process. Errors of Pu mass measurements exceeded 100%(specification: 20%) in many cases. |
| 5 | 1961.8.14 Siberian Chemical Combine UF ₆ enrichment, evaporation system. | UF ₆ accumulated in oil container (60L) of a pump connected with cooling vessel for 22.6% enriched UF₆ gas. Reached criticality at concentration of 400gU/L. Alarm activated, personnel evacuated. Radiation survey thereafter did not show abnormal radiation level and restarted operation, leading to another criticality. | Increased temperature. Discharged oil. | Total 10^{16} Two spikes, 5×10^{15} each. | 1 person 200rad | No monitor for process quantity. Insufficient UF ₆ gas cooling. Re-designed unit. Modified operating manual. |
| 6 | 1962.9.7 Mayak Enterprise, Urals Pu scrap dissolver tank | Following completion of the final dissolving operation in Pu dissolving tank (450mmD, 100L) (stopped stirring and heating), alarm was activated, and personnel evacuated. After the first spike, 2 spikes followed in next 40-50minutes. 1.32kg of Pu was left in the tank and some was undissolved. | Drained solution. | Total 2×10^{17} | No significant dose. | Dissolving tank had 5cm-lead shield. No persons nearby. |

Table 6.2 Criticality accidents in Russia (2/2)

| No | Date Place | Situation Cause | Quenching mechanism | Number of fissions | Dose | Others |
|----|--|--|-------------------------------------|--|---|--|
| 7 | 1963.1.30 Siberian Chemical Combine U scrap reprocessing | When dissolving high-enriched U metal pieces, misunderstood mass of 5% U as 5 gU/kg instead of 50gU/kg. The error was found by analysis after dissolving, and the solution was moved to safe geometry containers. But another analysis later was misunderstood again as 1/10 of actual mass. 40L(71gU/L) solution was moved to unfavorable geometry tank, which caused criticality. After 6 hours of 16 times power oscillations (solution draining and returning), the system became quasi-steady state condition. | Moved solution 10 hours later. | Total 7.9×10^{17} | 4 persons 6-17rad | First spike activated alarm, and personnel evacuated. |
| 8 | 1963.12.13 Siberian Chemical Combine U extraction process | Extraction solution went into vacuum system trap (100L) connected to high-enriched U transfer line. Contacted with overflowed U solution and extracted U. Trap filled with 33gU/L solution, caused criticality. First pulse and 16 power oscillations in next 6 hours. When stopped vacuum system, more extraction solution mixed with U solution, causing another spike and power oscillations and the system became quasi-steady state condition thereafter. | Fed cadmium solution. | 1 st pulse 1.6×10^{15} Total(18hr) 2×10^{17} | No exposure. | Alarm activated and personnel evacuated. |
| 9 | 1965.11.13 Electrostat Fuel Fabrication Plant, U conversion process | Double filters for 6.5% enrichment UO_2 powder were perforated, powder (slurry) escaped and accumulated 157kg (U: 51kg) in water reservoir (300mmDx650mmH) of vacuum system, creating criticality. | | Spike 10^{15} | 1 person 3.5rad | Filters were rarely inspected. No NDA. Alarm activated and personnel evacuated. Unit was dismantled. |
| 10 | 1965.12.16 Mayak Enterprise, Urals U scrap dissolving | Charged 2.2kg of high-enriched U scraps into dissolving tank (450mmD). Stopped dissolving operation in 40 min against standard practice of 1.5 hours (for planned room cleaning). 10 min later, alarm was activated and 11 spikes in 7 hours occurred. | Fed cadmium solution. | Total 7×10^{17} | Max. 0.03rad | No compliance with standard practices. U scrap mass was not controlled. Critical mass for dissolving tank was less than 2kg U scrap. |
| 11 | 1968.12.10 Mayak Enterprise, Urals Pu extraction | Some organic solvent went into 4000l tank containing low enriched Pu solution . While trying to remove organic solvent, transferred 40L solution to 60L unfavorable geometry, causing criticality. High concentration of Pu existent in organic solvent. Shift supervisor tried to drain leading to another criticality. | Draining | First 10^{16} Second 5×10^{16} | 1 person died (shift supervisor) 1 person seriously rad. damaged (both legs amputated) | Alarm activated and operators evacuated, but shift supervisor tried to drain solution in tank and received radiation. |
| 12 | 1978.12.13 Siberian Chemical Combine Pu metal storage | Inserted three Pu metal ingots in storage container, and when fourth ingot was inserted, criticality was achieved. Fourth ingot was ejected. | Operator removed ingots with hands. | 3×10^{15} | 1 person whole body 250rad hands 2000rad 7 persons 5-60rad | Container had spaces for polyethylene and cadmium, but they were shaped in such that Pu ingots could be inserted, which caused multiple inserted ingots. |

Table 6.3 Examples of criticality accident scenarios in safety analysis of reprocessing facility (1/2)

| Place | Barnwell | Exxon | West Valley | | PNC Tokai Plant | Rokkasho Reprocessing Plant of JNFL |
|-----------------------|---|--|--|---|---|--|
| | | | Design Basis Accident | Design Basis Accident | | |
| Purpose of evaluation | Design Basis Accident | Major Accident | Fuel storage pool at the fuel receiving facility | A solution-bearing vessel in the processing cells | Accident Evaluation | Design Basis Accident, Accident for Site Evaluation |
| Postulated Process | Dissolver tank | A solution-bearing vessel in the processing cells | | | Enriched U dissolver tank | Dissolver tank |
| Scenario | <p>(1) A criticality accident is postulated despite of the preventive actions below:</p> <ul style="list-style-type: none"> -1. Add 3g/ L of soluble neutron absorber (Gd) to nitric acid feed. -2. Install two independent Gd concentration monitors for each hull rins and seal Solution make-up tank and dissolver tank. -3. Low Gd concentration activates interlock for stopping nitric acid feed (two independent systems). -4. Hull Rins and seal solution make-up tank stores nitric acid containing three times more Gd concentration than that of dissolver tank for terminating the criticality. | <p>(1) When geometry control and fixed neutron absorber are in use, assume that these static safeguards were lost by mechanical damages or outside forces.</p> <p>*Slab tank distorted by hydrogen explosion was quoted as an example.</p> <p>(2) When concentration control and soluble neutron absorber are in use, assume that a mass control system failure or an operator error took place. Two or more independent but improbable abnormalities are assumed to be coincided.</p> | <p>(1) Due to a design basis tornado, the superstructure may collapse onto the stored fuel or a wind-generated missile may damage a portion of a fuel storage rack. Since the possibility of a criticality is not completely precluded, an evaluation has been made of the potential off-site radiological consequences of this highly improbable incident.</p> <p>(2) Total yield of 3×10^{19} is based on reported yields for heterogeneous metal incidents in water.</p> <ul style="list-style-type: none"> - Initial burst 3×10^{18} (0.1 sec) - Subsequent bursts 7×10^{18} (burst rate: 10^{17} fis./s) | <p>(1) Total yield of 3×10^{19} is based on reported yields for past solution fuel incidents.</p> <ul style="list-style-type: none"> - initial burst 1×10^{18} fissions (0.1 sec) - subsequent bursts 2.9×10^{19} | <p>(1) Total yield of 10^{20} and spike yield of 10^{19} are based on reported past solution fuel incidents and analyses.</p> | <p>(1) A criticality accident at the dissolver tank of the chopped and dissolving facility due to a decrease in acid concentration is postulated. The super criticality is detected by emergency soluble neutron absorber feeding system of the safety protection system, and quenched by addition of nitric gadolinium.</p> <p>(2) Total yield analyzed was 2.3×10^{17}, but, 10^{19} as DEB, 10^{20} as SEA are assumed based on past accident examples.</p> |

Table 6.3 Examples of criticality accident scenarios in safety analysis of reprocessing facility (2/2)

| Place | Barnwell | Exxon | West Valley | | PNC Tokai Plant | Rokkasho Reprocessing Plant of JNFL |
|----------------------|---|---|--|---|---|--|
| Scenario (cont'd) | <p>(2) Notified by cell radiation monitor alarm, the operator adds high concentration Gd (stops fuel shearing and acid feeding), and terminates the criticality within 7 minutes from the alarm.</p> <p>(3) Total yield is assumed to be 2×10^{19}.</p> <p>Initial burst 1×10^{18} fissions Average power 0.4×10^{17} fis./s</p> | <p>(3) The total yield of 2×10^{19} was assumed based on past incidents. One burst of 0.5-sec duration was assumed to occur every 10 minutes over the 24-hour period.</p> <p>0-30 sec: $5 \times 10^{17} \times 3$ (times) 30 min-8hrs: $2 \times 10^{17} \times 45$ (times) 8hrs-24hrs: $1 \times 10^{17} \times 95$ (times)</p> | | | | |
| Radiological effects | <p>(1) All of the noble gases and 50% of the iodine were assumed to be released from the stack.</p> <p>(2) in-site exposure Whole body: 3.5mrem Thyroid: 130mrem</p> | <p>(1) All of the noble gases and 25% of the iodine were assumed to be released from the stack.</p> <p>(2) Off-site exposure Whole body (external): 25mrem Whole body (inhalation): 0.2mrem Thyroid: 130mrem</p> | <p>(1) Assume dispersion on ground.</p> <p>(2) Off-site exposure Whole body (external): 1mrem Thyroid: 5mrem</p> | <p>(1) All of the noble gases and 50% of the iodine were assumed to be released from the stack.</p> <p>(2) Off-site exposure Whole body (external): 1.3rem Thyroid: 31rem</p> | <p>(1) All of the noble gases and 10% of the iodine were assumed to be released from the stack.</p> <p>(2) Off-site exposure Whole body: 0.1rem Thyroid: Child 2.1rem Adult about 0.47rem</p> <p>(3) National genetic dose 48,000 man-rem</p> | <p>(1) All of the noble gases and the iodine were assumed to be released to the atmosphere.</p> <p>(2) Off-site exposure DBE 0.57mSv SEA 6.1mSv</p> <p>(3) Population dose 3,600 man-Sv</p> |

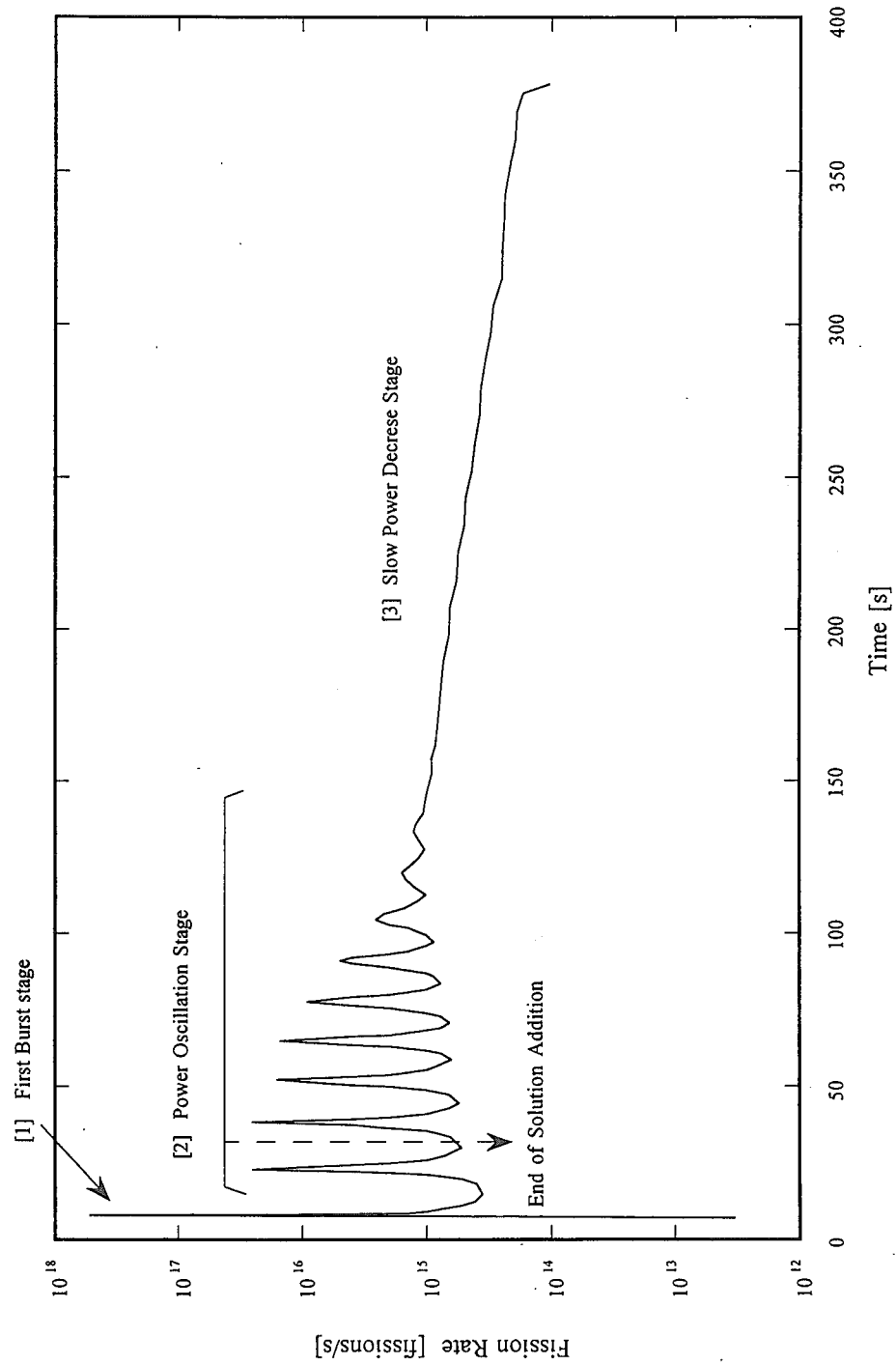


Fig. 6.1 Power (fission rate) changes in time at a CRAC experiment (CRAC13)

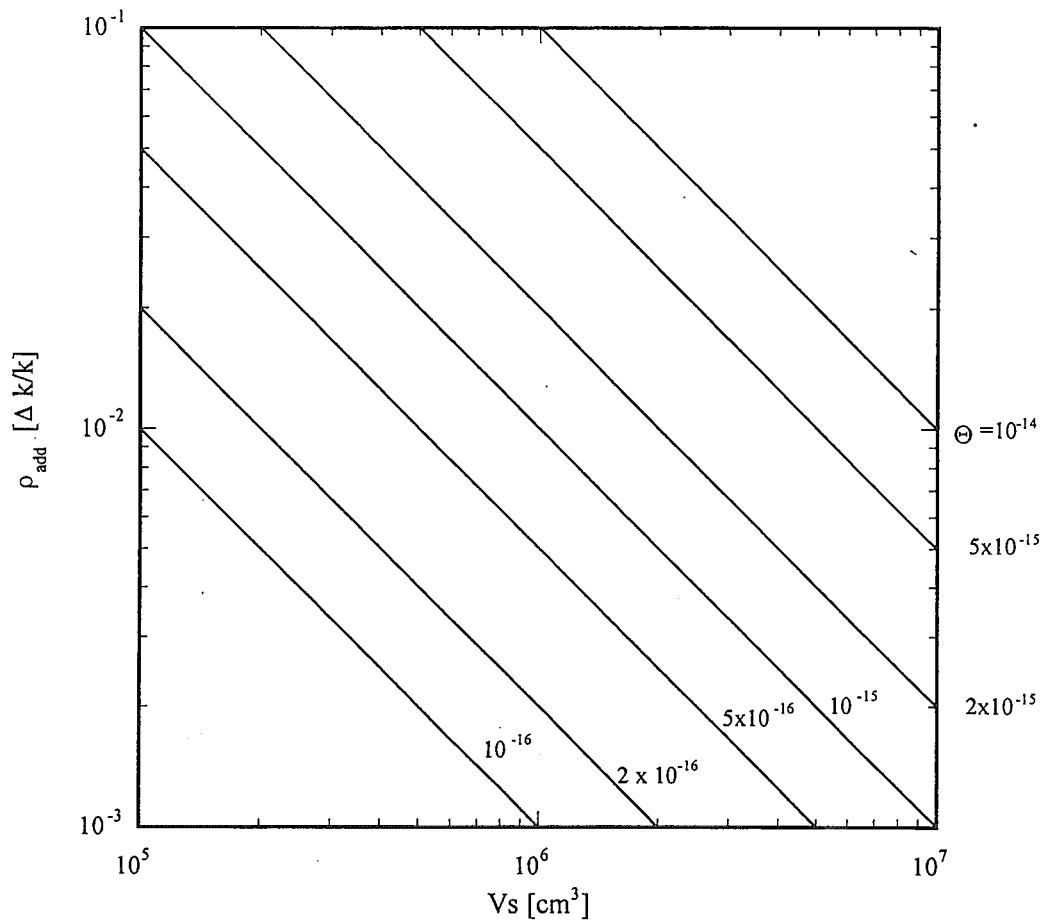


Fig.6.2 Correlation between ρ_{add} and V_s for various fuel solutions parameterized in fuel composition Θ ($F=10^{19}$ fissions)

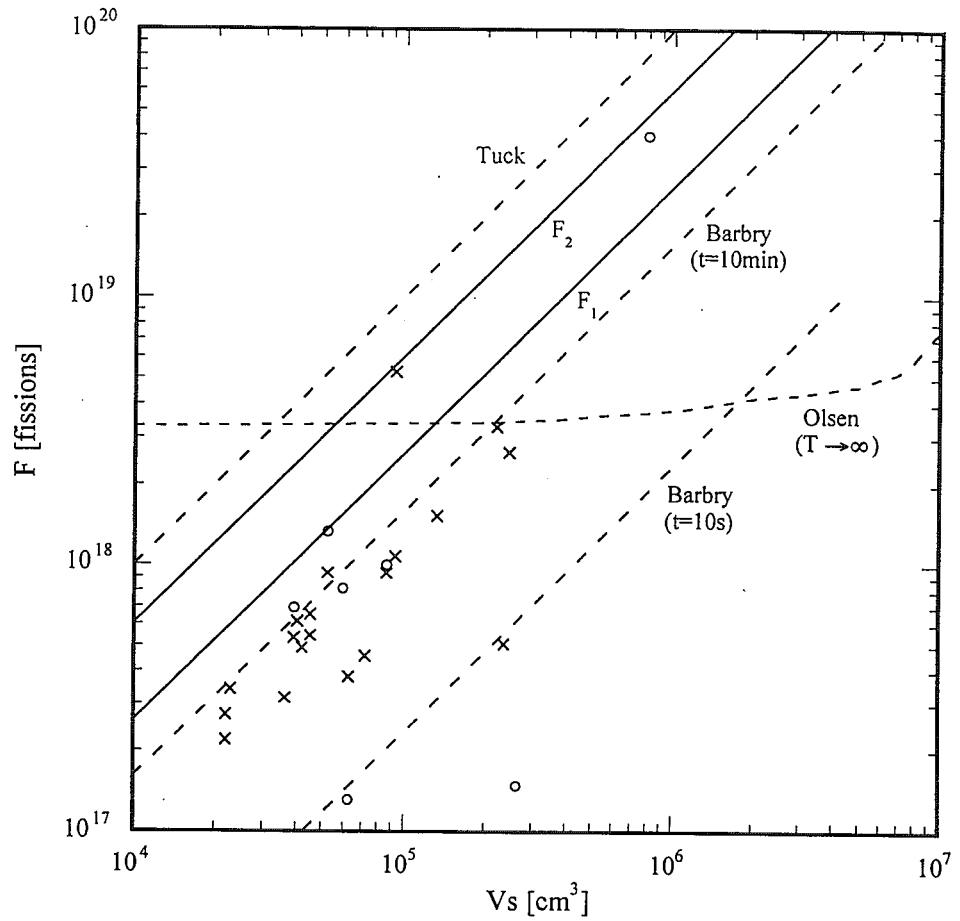
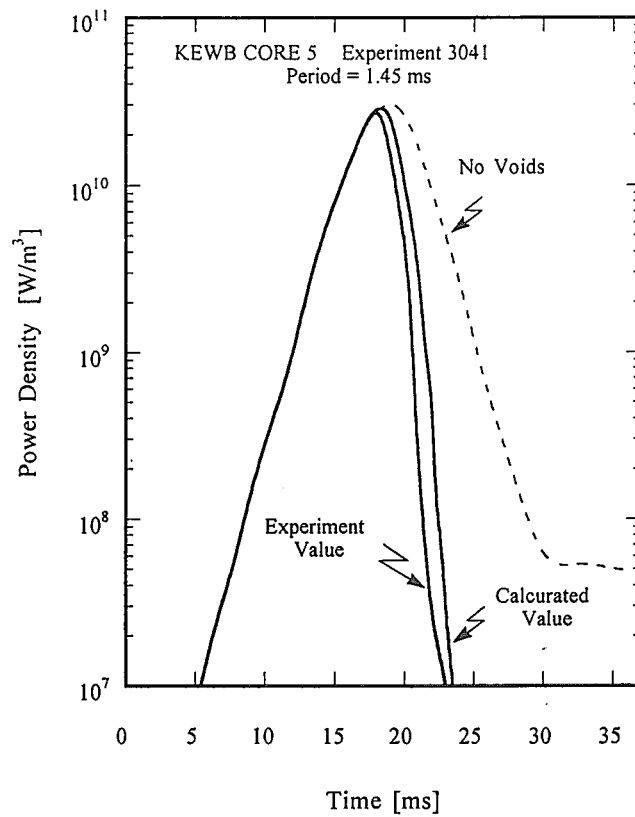
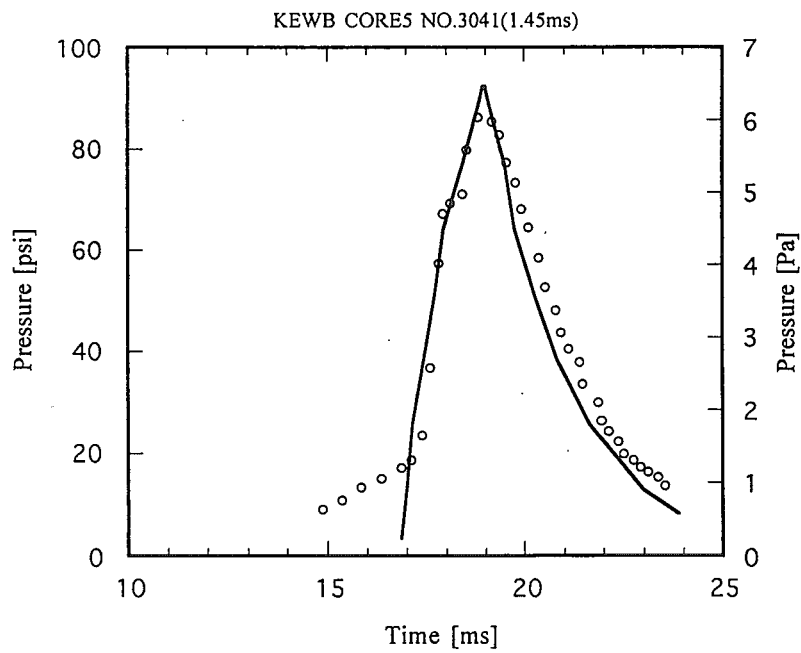


Fig. 6.3 Comparison among various simple analysis formula, $F_1 = 2.6 \times 10^{13} \cdot V_s$, $0 < V_s < \infty$, $F_2 = 6.0 \times 10^{13} \cdot V_s$, $0 < V_s < \infty$, CRAC experiment data (symbol x) and past accident data (symbol o)

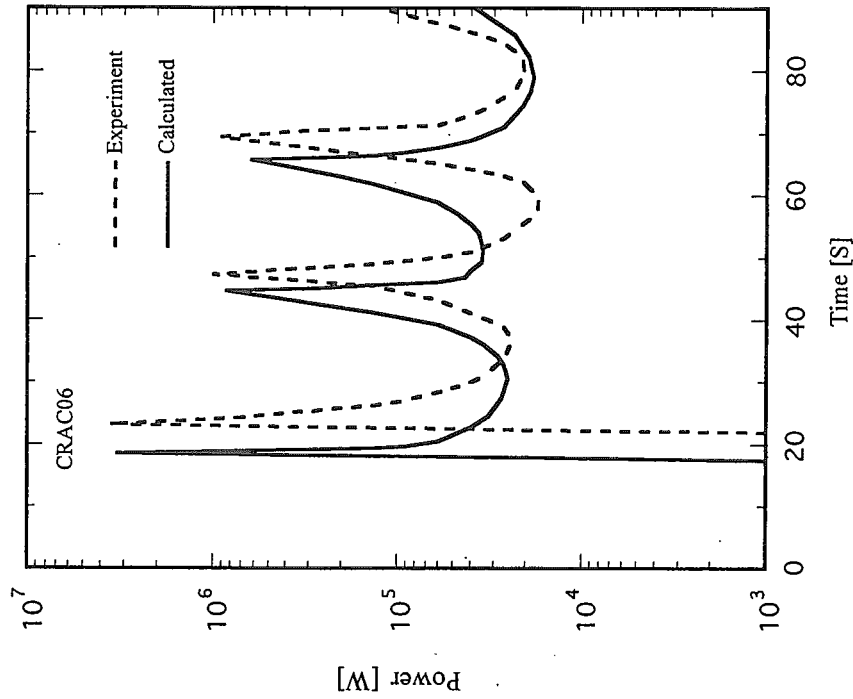


(a) Power Change in Time

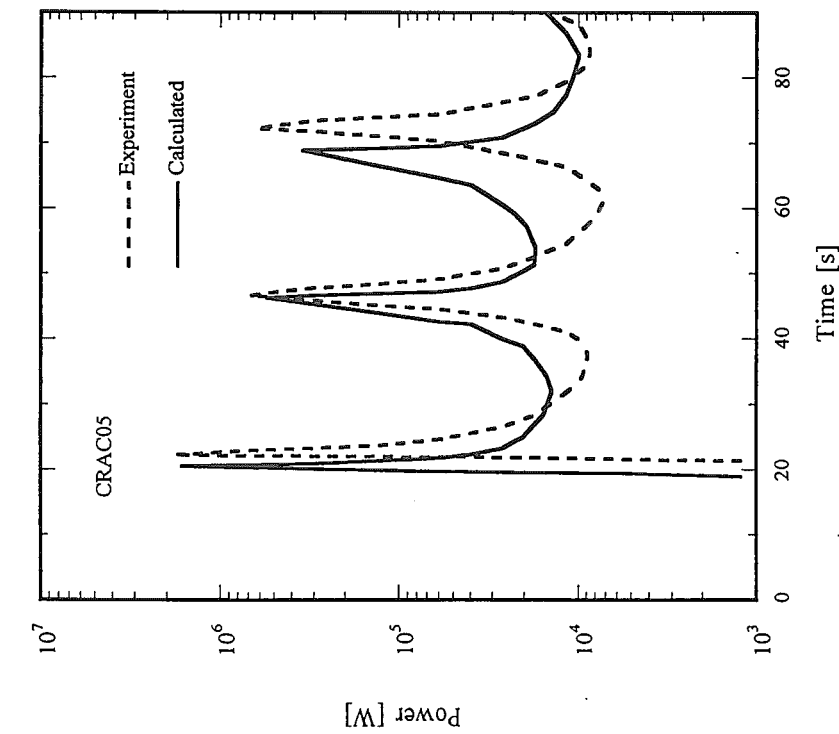


(b) Pressure change in time

Fig. 6.4 Analysis of KEWB experiments by AGNES code "pressure model"

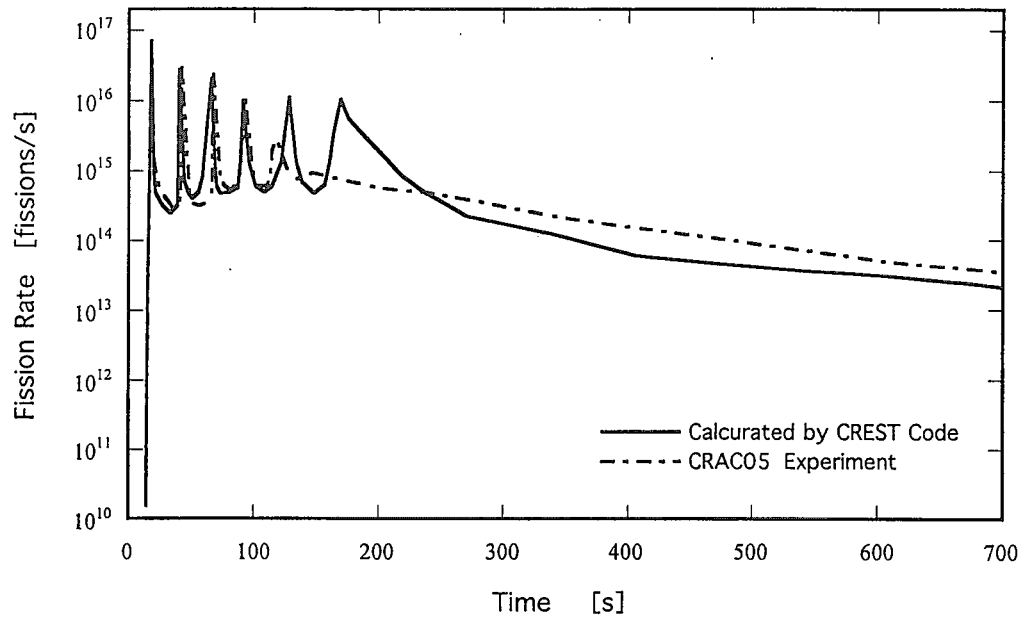


(a) CRAC05 experiment

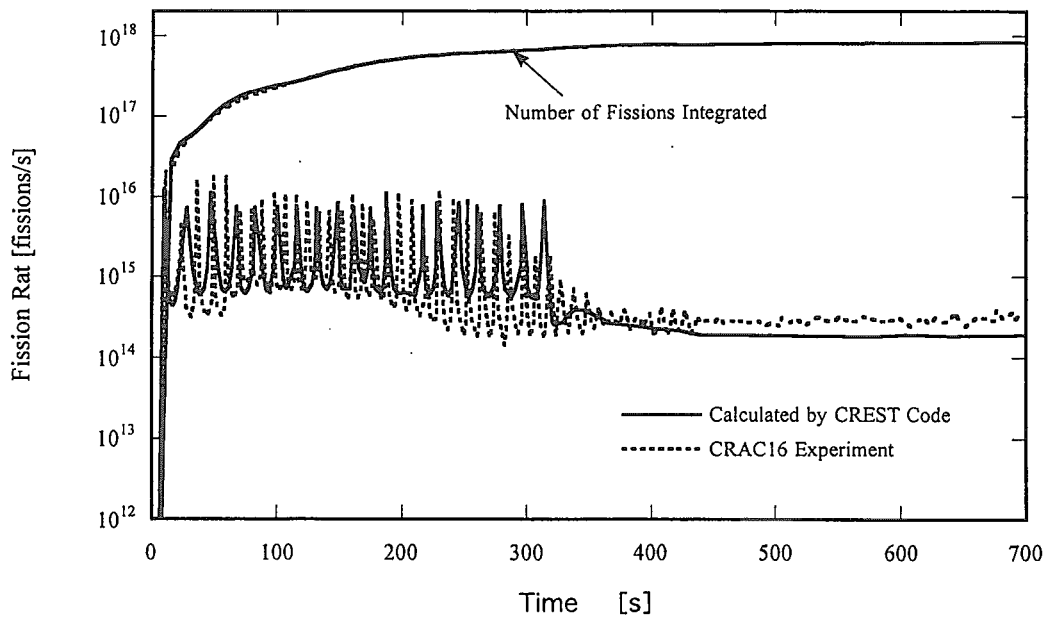


(b) CRAC06 experiment

Fig. 6.5 Analysis of CRAC experiments by AGNES code “modified energy model”



(a) CRAC05 Experiment



(b) CRAC16 experiment

Fig. 6.6 Analysis of CRAC experiments by CREST code

7. BASIC PRINCIPLES ON DESIGN AND INSTALLATION OF CRITICALITY ALARM SYSTEM

7.1. Basic Principles on Design and Installation

The Regulatory Basic Guides for Licensing Safety Review of Nuclear Fuel Facility⁽¹⁾ includes the Item 12 entitled *Consideration Against Criticality Accidents*. The Item 12 states; "For nuclear fuel facilities in which a criticality accident might happen for any cause such as an operator error, appropriate measures shall be provided against the very rare potential criticality". *The Regulatory Guides for Licensing Safety Review of Reprocessing Facilities*⁽²⁾ includes also Item 12 stating that the criticality alarm system shall be designed to detect a criticality accident, regardless of the low probability of an occurrence. Here, the criticality alarm system (CAS) is defined to be a system sounding an alarm signal detecting the accident.

The objective for installation of the CAS is to alert workers of a criticality accident and evacuate the affected area immediately to minimize the radiation exposure. Relevant international standards⁽⁶⁾ and/or standards in other countries such as the United States⁽³⁾, United Kingdom⁽⁴⁾ and France⁽⁵⁾ include identical objectives. The following items must be carefully considered in defining the requirements for a specific CAS.

- (a) Installation Regions Requiring CAS
- (b) Characteristics of Postulated Criticality Accident
- (c) Specifications and Arrangement of CAS

7.2. Selection of Installation Place

7.2.1. Regulations of USA and UK for Selection of Installation Place

(1) United States

The US Code of Federal Regulations 10 CFR⁽⁷⁾ Part 70 section 70.24(a) requires any facility processing more than 450 g of plutonium or 700 g of uranium-235 to have a CAS capable of detecting an absorbed dose of 20 rad (0.2 Gy) at a distance of 2 meters from the radiation source as described in the relevant section.

ANSI/ANS 8.3⁽³⁾ Section 4.2 requires evaluating the necessity of installing a CAS in any nuclear facility where there is more than 450 g of plutonium or 700 g of uranium-235. In addition, the standard stipulates that a CAS is not required in regions where the maximum adsorbed dose in air is less than 12 rad (0.12 Gy), equivalent to a maximum of 2×10^{19} total fissions in estimating the dose. The US Nuclear Regulatory Commission Guide 8.12⁽⁸⁾ states that the bases defined in ANSI/ANS 8.3 are appropriate for an exemption from CAS installation because of the quantity, chemical state and configuration of the nuclear fuel involved.

(2) United Kingdom

The United Kingdom Atomic Energy Authority (UKAEA) Advisory Committee for Radiation Protection and Measurement issued Guidance Report SRD R309⁽⁴⁾ "Design Criteria and Principles for Criticality Detection and Alarm Systems" in 1984. The Report recognizes criticality detection and alarm systems shall be provided at all places where fissile material may be used or stored, unless it is confidently judged that criticality would not reasonably be expected having regard to the nature of the particular operations and facility concerned. Also, in cases where the maximum allowable emergency dose of 12 rad (0.12 Gy) in air can not be exceeded, special consideration is necessary, which referred to ANSI's description.

7.2.2. Selection Criteria for Installation Place

The requirement for installation of a CAS must be evaluated in any area where the quantity of Special Nuclear Material (SNM) equals, or exceeds, the Minimum Estimated Lower Criticality Value (MELCV)* or where the value may be exceeded in event of a single equipment failure or operator error even if quantity of SNM is less than MELCV in normal operation. The evaluation must consider the probability of a Criticality event and the resultant maximum operator dose.

* This is not the minimum critical nuclear mass (450g of plutonium, 700g of uranium-235, etc.), described in US Standards, but the minimum estimated lower criticality specified in the this Handbook, considering the most severe chemical composition, configuration, reflection, etc.

(1) Probability of Criticality Accident

Criticality accident probability might be low in present operations where safety measures are implemented with sufficient margin to allow for single failure criteria. Selection of regions for CAS must be based on a careful examination of experience in previous accidents and the accumulated knowledge with criticality safety design. The "Criticality Accident Counterplan Facility Examination," described in "Criticality Safety Control Facility" Section 2.2, provides important guidance.

(2) Probability of Excessive Exposure to Workers

After selection of a region based on the probability of criticality accident has been completed, there must be an evaluation of the probability of workers exposed to excessive radiation from the event. Allowable radiation exposure for workers should be determined by using national limits (100mSv for emergency situations described in Section 7.4.3), with appropriate design or administrative reductions based on managerial philosophy. On the other hand, the excessive level of radiation exposure should be determined not only by referring to the national decree limiting effective

equivalent dose of 100 mSv for emergency works, but also by the design and operation philosophy adopted by the management. If the operator exposure during a criticality accident is within the specified allowable limits, installation of the CAS is not required.

The workers who are assumed low effective dose equivalent exposure due to sufficient distance from the criticality accident region shall be alerted by verbal messages throughout the facility to evacuate or to stay clear of the accident region.

7.2.3. Consideration in Maximum Assumed Number of Total Fissions

ANSI/ANS-8.3⁽³⁾ specifies 2×10^{19} fissions as the maximum from a credible criticality accident. This estimate might be based on experience of the past criticality accidents involving nuclear fuel in solution systems, together with the results of the French CRAC experiments (Section 6.1 in reference). In selection of the regions for installation of the CAS, the fuel conditions, i.e., kind, quantity, concentration, distribution, volume, and operating procedures must be considered.

7.3. Kinds of Radioactive Rays Detected

7.3.1. Radiation Field Surrounding a Critical Assembly

The majority of radiation from a criticality accident consists of gamma and neutrons produced directly in the fission process. Secondary gamma radiation, distinctly different from the primary radiation, is emitted as a result of neutron capture reactions with structural materials, shielding, etc. surrounding the criticality source. Both primary and secondary radiation are considered prompt radiation, emitted instantaneously in a criticality accident. Delayed neutrons, beta, and gamma, which are emitted as the fission products decay, are not significant compared to the prompt.

Neutron and gamma ray doses in free air, from prompt radiation observed in various critical assemblies including aqueous solution, metal, and shielding, are shown in Table 7.1^{(4),(9)-(15)}. The values are normalized to 10^{15} fissions at a distance of 1 meter from the critical assembly surface. The table indicates that the normalized absorbed dose depends largely on the composition and size of the critical assembly, and the size and kind of shielding materials surrounding the source. The ratio of neutron to gamma D_n/D_γ may vary by factors of 30 depending upon the specific parameters. Details of differences in various systems are summarized below:

(1) Systems without Shielding

The metal system without shielding exhibits higher D_n/D_γ ratio than the hydrogen moderated solution system without shielding. This is attributed to a higher gamma decay in the metal system and a lower neutron energy spectrum in the moderated solution. JEZEBEL ($D_n/D_\gamma = 10$) is an unmoderated metal system with the minimum quantity of ^{239}Pu to achieve criticality. The average energy of the

leaked neutron spectra emitted from this assembly is high. Conversely, in the hydrogen moderated system, with the large reduction in neutron energy, the D_n/D_γ is small. In an Oak Ridge Y-12 mock-up system, consisting of enriched uranyl nitrate solution in a 25cm radius cylindrical configuration, the D_n/D_γ was 0.36. In a CRAC experimental system, with similar uranyl nitrate solution in a smaller 15 cm radius cylindrical arrangement, the higher $D_n/D_\gamma = 0.76$ was attributed to higher neutron leakage spectra.

(2) Systems with Shielding

In shielded systems, the D_n/D_γ ratio varies with the size and composition of the shield. Iron and steels are very effective shielding for gamma radiation, with minor impact on neutrons; with a resultant higher D_n/D_γ . Graphites, concrete, and materials with high hydrogen content, with relatively little gamma shielding capability compared with steels, exhibit high neutron moderation effects, with a resultant decrease in the D_n/D_γ . The D_n/D_γ for concretes and other hydrogen shielding materials is further reduced because of the contribution of secondary gamma rays emitted by thermal neutron capture.

7.3.2. Kinds of Radioactive Rays Detected

As discussed in the previous section, the D_n/D_γ of prompt radiation emitted during a criticality accident is strongly dependent on the specific characteristics of the critical system and shielding. Because of this dependency, one must consider for selection of design conditions of the subject facility, and the development status of applicable radiation detectors.

Because the generation of prompt neutrons is more specifically associated with nuclear fission, compared with gamma rays, neutron measurement may be used to distinguish a criticality accident from other nuclear physics phenomena, especially in high gamma fields.

7.4. Minimum Level for Detection

7.4.1. Ideas Found in European and American Regulations

Standards for CAS in the US, UK, and France specify the threshold detection value as the dose at worker's location for the minimum credible criticality accident to be detected (Table 7.2). In the ANSI of US and France, the detected dose is based on the absorbed dose in air; in the UK and USNRC the absorbed dose in human tissue is used. The doses in these standards range from 0.125 to 0.25 Gy. The US standard employs a dose rate limit, while the UK and France use an integrated dose in consideration of relatively slow transients that may be involved. In a simulation experiment of a criticality accident using SHEBA in the US and SILENE in France, large individual exposures, resulting from gradually increasing radiation intensity, could not be avoided by using the dose rate for

detection⁽¹⁶⁾.

7.4.2. Criteria for Deciding Minimum Level for Detection

The threshold value is the radiation dose to be detected that is necessary for CAS to sound the evacuation alarm. This value is determined as the detected dose at the worker's position defined in the standards of the US, UK, and France. The relationship between the threshold value, CAS sensitivity, and dose rate for initiating alarm is discussed in section 7.5.

The primary functions of the detection system are to 1) alert workers of the minimum credible criticality accident with an adequately high reliability and 2) have an adequately low probability of false alarms. Optimization of the detector system design and arrangement should consider all aspects of the radiation field characteristics (type and intensity), detector type (integrated dose or dose rate), safety factor (Section 7.5.1) and sensitivity of alarm sounding. Detector arrangement considerations are discussed in Section 7.5.

7.4.3. Remarks on Deciding Minimum Level Based on Effective Dose Equivalent Limits

Japan has a number of regulations relating to public safety assurance and the minimization of radiation hazards to worker in research reactors, commercial reactors, RI handling facilities, and other nuclear fuel handling facilities. Major regulations addressing these issues include 1) "*the Law for the Regulations of Nuclear source Material, Nuclear Fuel Material and Reactors*," 2) "*the Law of Prevention from Radiation Hazard Caused by Various Radioactive Isotopes*," and 3) "*the Decree of Prevention from Ionizing Radiation Hazard*" coming from "*the Law of Safety and Health for Workers*". Effective dose equivalent is essentially used as unit of the limit of dose equivalent for radiation workers and the surrounding environment.

To date, this dose has been based on the dose equivalent (rem) is expressed by quality factor showing magnitude of effect of each radiation type such as gamma, beta, or neutrons, multiplied by the irradiated dose R or the absorbed dose (rad) in air or human equivalent tissue. Since April 1989, present regulations have utilized the effective dose equivalent (Sv) in accordance with the ICRP Publication No.26⁽²⁰⁾. The effective dose equivalent (Sv) is summation of the dose equivalents of the external radiation exposure to each of human body's organ tissues considering the magnitude of the stochastic effect of carcinogens in each organ tissue. Eventually, the effective dose equivalent represents dose equivalent proportional to incidence of stochastic effect on the whole body. The effective dose equivalent for external exposure is managed by the ICRU sphere 1 cm dose equivalent (Sv). The quality factor of gamma radiation is 1 while that of neutron, depending upon the energy level, is considered to be 2 to 10. There is a potential for increasing quality factor of neutron by a factor of 2 in the future.

In the determination of the threshold value for criticality detection in Japan, it may be

appropriate to use the effective dose equivalent (Sv) as currently specified in the national radiation exposure control regulations. The adoption of 0.1 Sv (12 rem in the past, altered by revision of regulation), which is the limit applicable to emergency operation, is readily understandable. However, the dose equivalent is not recommended for use in evaluation of non-stochastic health effects caused by highly absorbed dose in accident situations, where the quality factor is not determined, according to the 1977 ICRP recommendation ⁽²⁰⁾. The maximum permissible emergency dose could be defined as absorbed dose as is used in some countries. To estimate the dose equivalent for the human body from the maximum credible fissions, additional transient criticality experiments may be required, simulating nuclear material systems encountered in practical facilities.

7.5. Sensitivity and Installation of Detectors

7.5.1. Sensitivity of Gamma-ray Detector for Alarming Criticality

Duration of gamma radiation produced during a criticality accident depends primarily upon whether the medium is metallic or aqueous solution, and the inserted reactivity of the system. Based on previous experience and power excursion experiments, the duration is expected to range from about 1 millisecon to more than 1 minute. If the electronic circuit time constant of the gamma ray CAS is more than the gamma pulse width, the alarm may not be activated even though the radiation level exceeds the alarm setting dose rate. Fig. 7.1 shows the relation of incident gamma pulse width with alarm output for a typical gamma CAS (response :1.5 ms). This indicates that for an incident pulse width of less than 1 millisecon the alarm will not be activated.

The dose sensitivity to generate alarm signal is defined as the gamma radiation pulse dose rate that will be detected and generate the alarm with a pulse width of 1 ms. The alarm safety factor is the multiple between the dose rate set for alarm generation and the dose sensitivity to generate alarm signal. The value, which is dependent upon the electronic circuit time constant, is determined by simulating detector current with electrical pulses.

As an example, with a electronic circuit time constant of 1 millisecon and a safety factor of 10, and dose rate set for alarm generation of 20 pC/(kg·s), the dose sensitivity to generate alarm signal is evaluated to be 0.2 pC/kg by using the following equation:

$$20[\text{pC}/(\text{kg}\cdot\text{s})]\times 10\times 10^{-3}=0.2[\text{pC}/\text{kg}] \quad (7.1)$$

The result shows that the gamma CAS will generate an alarm within 1ms after exposure to a gamma dose of 0.2 pC/kg.

7.5.2. Sensitivity of Ionization-Chamber-Typed Detector for Alarming Criticality

Behavior of the integrated dose response for a pulse exposure of a typical ionization chamber detector is shown in Fig. 7.2. Since the dose sensitivity of an ionization chamber to generate alarm signal may be measured experimentally, the safety factor may be determined by the following equation, similar to (7.1).

$$D_A[\text{pC}/(\text{kg}\cdot\text{s})]\cdot f_1 \times t[\text{s}] = D_M[\text{pC}/\text{kg}] \quad (7.2)$$

where

- D_A : Dose rate set for alarm generation [$\text{pC}/(\text{kg}\cdot\text{s})$],
- f_1 : Safety factor [-],
- t : Duration of burst [s], and
- D_M : Dose sensitivity to generate alarm signal [pC/kg].

Assume the dose rate set for alarm generation (D_A) for a logarithmic ionization detector is $10 \text{ pC}/(\text{kg}\cdot\text{s})$, the gamma pulse duration is 1 millisecon, and the experimentally determined dose sensitivity to generate alarm signal (D_M) is $22.5 \text{ pC}/\text{kg}$. Using equation 7.2, the safety factor (f_1) becomes 2,250. Based on this calculation, ionization chambers will generate a criticality alarm for 1 millisecon gamma pulses of $22.5 \text{ pC}/\text{kg}$ with a safety factor of 2,250. This safety factor is consistent with the value $f_1 = 2500$, which is shown in the appendix of ANSI/ANS 8.3⁽³⁾. However, it is unrealistic to assume a safety factor in excess of 2,000 for a logarithmic ionization chamber CAS. Using a different type of radiation detector with a fast response is recommendable for a criticality accident with a pulse duration in the range of 1 millisecon. It may also increase the detection range.

7.5.3. Planning for Installation of Detectors

The US Code of Federal Regulations 10 CFR⁽⁷⁾-70.24 (a)(1) requires use of redundant criticality detectors for all equipment or systems where there is a possibility of a criticality accident. Use of a single, highly reliable, detector to simultaneously monitor multiple locations is permissible. In such cases, the required detection threshold dose sensitivity, location and number of detectors must be determined based on the dose sensitivity to generate alarm signal.

The method to determine the detection range is shown by equation 7.3 based on the appendix of US standard ANSI/ANS 8.3. The arrangement between a radiation source, a worker, and a detector is shown in Fig. 7.3.

$$r^2 \cdot D_M = d^2 \cdot D_P \cdot f_2 \cdot 1/K, \quad (7.3)$$

where

D_M : Dose sensitivity to generate alarm signal[Gy] (determined by the last section) ,
 D_p : Exposure dose detection threshold value[Gy] (this value is 0.2Gy in ANSI standard),
 r : Distance between a source and a detector [m] ,
 d : Distance between a source and a person [m] ,
 K : Damping ratio by air (1/3 in ANSI) , and
 f_2 : Detector correction factor defined by a ratio of gamma rays and neutrons in the radiation field depending on the property of source material; for example of gamma ray detector,

Metallic system (gamma ray : neutrons = 1 : 12)

$$f_2 = 1/(1+12) = 0.077$$

Aqueous solution system (gamma ray : neutrons = 1 : 0.3)

$$f_2 = 1/(1+0.3) = 0.77$$

Using Eq. (7.3), the radius of the detectable range is given by the following equation.

$$r^2 = d^2 \cdot D_p / D_M \cdot f_2 \cdot (1/K) \quad (7.4)$$

7.5.4. Checking Detectors in Over-loaded States

The maximum fission rate and the total number of fissions during a maximum credible accident must be determined to evaluate the operability of a detector in an overloading state. UK Standard⁽⁴⁾ recommends the use of 10^{21} fissions/sec as the maximum credible fission rate.

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Table 7.1 Prompt radiation absorbed dose of tissue in open air at distance of 1 m from a criticality assembly with a fission number of 10^{15} (4),(9)-(15)

| Criticality system | Neutron dose D_n (mGy) | Gamma dose D_γ (mGy) | Total dose $D_n + D_\gamma$ | Dose ratio D_n/D_γ | Ref |
|---|-----------------------------|--------------------------------|--------------------------------|------------------------------|------|
| Bare metal system | | | | | |
| (1) JEZEBEL 17kg ^{239}Pu | ~440 | ~44 | ~480 | 10 | (9) |
| (2) HPRR 117kg $^{235}\text{U-Mo}$ | 360 | 59 | 420 | 6.1 | (10) |
| (3) VIPER | 49 | 23 | 72 | 2.2 | (11) |
| (4) GODIVA ^{235}U R=8.7cm | 350 | 77 | 430 | 4.5 | (12) |
| (5) ^{239}Pu sphere R=5cm | 400 | 34 | 430 | 12 | (13) |
| Bare hydrogen moderating system | | | | | |
| (1) ORNL UO_2F_2 solution | 160 | ~250 | ~410 | ~0.63 | (12) |
| (2) CRAC ^{235}U solution | 210 | 280 | 490 | 0.76 | (14) |
| (3) Y-12 ^{235}U solution | 67 | 190 | 260 | 0.36 | (15) |
| Shielded system | | | | | |
| (1) 13mm thickness steel | 130 | 18 | 150 | 7.3 | (10) |
| (2) 20cm thickness graphite | 130 | 40 | 170 | 3.2 | (10) |
| (3) 20cm thickness concrete | 84 | 35 | 120 | 2.4 | (10) |
| (4) 12cm thickness lucite | 74 | 65 | 140 | 1.1 | (10) |
| (5) 23cm thickness lucite | 27 | 39 | 66 | 0.7 | (10) |

Table 7.2 Comparison and examination of criticality accident detection standards

| Ref. | Detection threshold value | Principle for establishment | The problem |
|---|--|---|---|
| ANSI/ANS 8.3-1986 ⁽³⁾ (USA, ISO ⁽⁶⁾) | With conventional shielding position of 2m from the source, absorbed dose in the air is 0.2Gy, 60sec. | Based on slow excursion in the CRAC experiment of highly enriched uranium system. | Difference between this and maximum permissible dose of 0.12Gy during emergency. Applicable to slightly enriched uranium. |
| SRD R-309 (1984) ⁽⁴⁾ (UK) | Minimum criticality phenomenon detected should be 1×10^{15} fissions, and decrease one order in proximate work. At the position of 2 m from a source without shielding, the absorbed dose corresponds to 0.125 Gy of human tissue in the air. | Based on mechanism of excursion and studies of past phenomena. In proximate work, assume smaller minimum criticality phenomenon in order to avoid extra exposure. | The process to induce minimum criticality phenomenon is complicated, depending on the principle for limiting exposure. Applicable to slightly enriched uranium. |
| IAEA-SM-215/34 ⁽⁵⁾ (France) | At the position of 1 m from a source, the absorbed dose is 0.25 Gy in the air. | At the half limit of a lethal dose of 4 Gy, sound the alarm when dose reaches 1 Gy at the distance 50 m. | The evidence is not clear for estimating dose limit. |

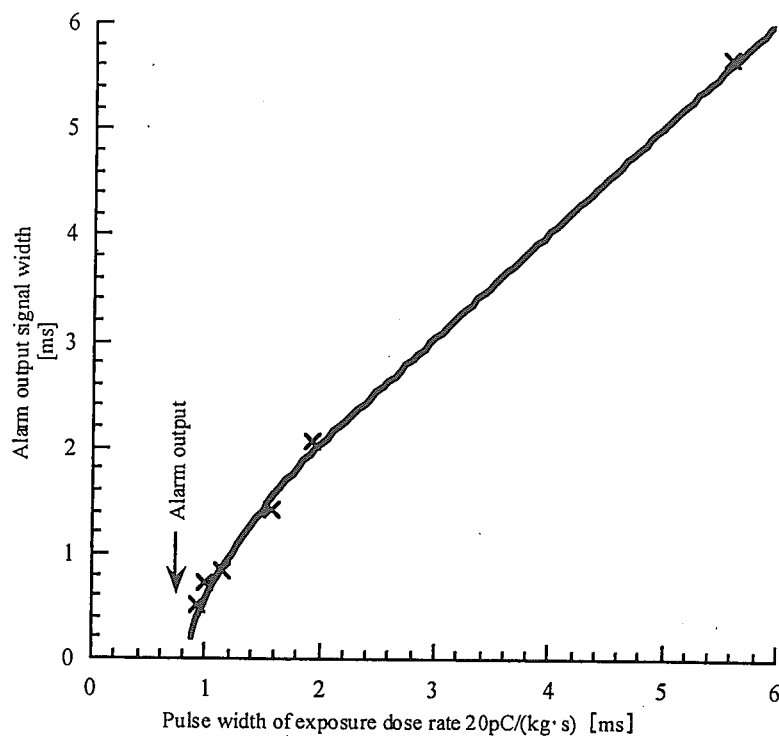


Fig. 7.1 Relation between incident gamma ray pulse width and alarm output width (pulse scintillation counter of current measure type)

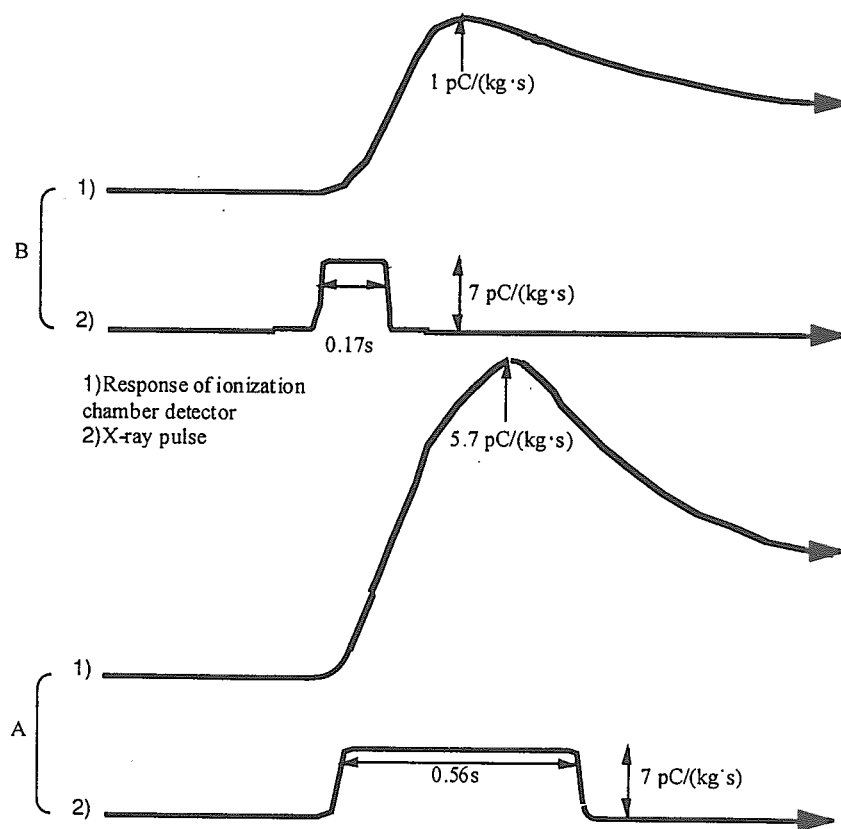


Fig. 7.2 Relation of ionization chamber detector irradiated width pulse X-ray up to 7pC/(kgos)

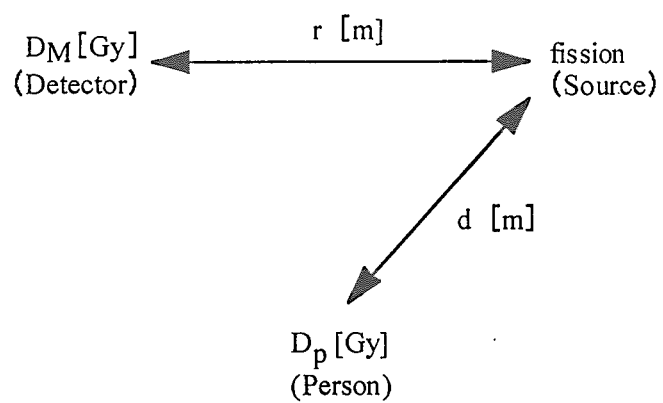


Fig. 7.3 Relation of position between source, person and detector

Appendix of Chapter 7 Recent Trends in Dose and Dose Limits for Radiation Protection

Table 7A.1 shows the International Commission on Radiological Protection (ICRP) recommendation (1977) (publication 26), which formed a foundation of the present regulation, in comparison with the updated recommendation (1990). The new recommendation which adopted the Paris statement of 1985 is characterized as follows: (1) Radiation weighting factor corresponding to the usual quality factor of neutron formerly used was changed. (2) In adopting new weighting factors for digestive system, weighting factors for other internal organs were also made smaller. (3) Annual dose limit was reduced from 50mSv to 20mSv (More strictly stating, 100mSv over five years. With the condition that, dose limit cannot exceed 50mSv any one year). In the future, Japan will expedite the implementation of these new recommendations to existing regulations.

Table 7A.1 Comparison of 1990 and 1977 ICRP recommendation (1/2)

| ICRP'77 recommendation | | ICRP'90 recommendation | |
|--|-------|---|-------|
| $D_T = d\epsilon_T/dm_T$ (Gy) | | $D_T = \epsilon_T/m_T$ (Gy) | |
| D_T : Absorbed dose in tissue (organ) T (Gy) | | D_T : Average absorbed dose in tissue (organ) T (Gy) | |
| $d\epsilon_T$: Mean energy imparted by ionizing radiation to matter in a volume element (J) | | ϵ_T : Total energy imparted in tissue (organ) T (J) | |
| dm_T : Mass of matter in this volume element (kg) | | m_T : mass of that tissue (organ) T (kg) | |
| $H_T = Q D_T N$ (Sv) | | $H_T = \sum w_R D_{T,R}$ | |
| H_T : Dose equivalent in tissue (organ) T (Sv) | | H_T : Equivalent dose in tissue (organ) T (Sv) | |
| Q : Quality factor | | $D_{T,R}$: Average absorbed dose of the tissue (organ) T (Gy), due to radiation R | |
| N : Product of all modifying factors with a value of 1 | | w_R : Weighting factor for radiation R | |
| $H_E = \sum w_T H_T$ (Sv) | | $E = \sum w_T H_T$ (Sv) | |
| H_E : Effective dose equivalent (Sv) | | E : Effective dose (Sv) | |
| w_T : Weighting factor for tissue (organ) T (fatal cancer, incidence of hereditary effect) | | w_T : the weighting factor for tissue (organ) T (fatal cancer, weighting non-fatal cancer, weighting serious hereditary effect, relative period of lifetime loss) | |
| | w_T | | w_T |
| Gonads | 0.25 | Gonads | 0.20 |
| Bone marrow (red) | 0.12 | Bone marrow (red) | 0.12 |
| Colon | - | Colon | 0.12 |
| Lung | 0.12 | Lung | 0.12 |
| Stomach | - | Stomach | 0.12 |
| Bladder | - | Bladder | 0.05 |
| Breast | 0.15 | Breast | 0.05 |
| Liver | - | Liver | 0.05 |
| Oesophagus | - | Oesophagus | 0.05 |
| Thyroid | 0.03 | Thyroid | 0.05 |
| Skin | - | Skin | 0.01 |
| Bone surface | 0.03 | Bone surface | 0.01 |
| Remainder | 0.30 | Remainder | 0.05 |

Table 7A.1 Comparison of 1990 and 1997 ICRP recommendation (2/2)

| ICRP'77 recommendation | | ICRP'90 recommendation | |
|---|-----|--|-------|
| Quality factor, Q | | Radiation weighting factor, w_R | |
| Type of radiation | Q | Type and energy range | w_R |
| X rays, γ rays and electrons | 1 | Photons, all energies | 1 |
| Thermal neutrons | 2.3 | Electrons, mesons, all energies | 1 |
| Neutrons, protons and single-charged particles at a rest mass greater than one atomic mass unit of unknown energy | 10 | Neutrons, energy | |
| | | < 10keV | 5 |
| | | 10 to 100keV | 10 |
| | | 0.1 to 2MeV | 20 |
| | | 2 to 20MeV | 10 |
| | | > 20MeV | 5 |
| | | (or $w_R = 5 + 17\exp[-(\ln 2E)^2/6]$) | |
| Alpha particles and multiple-charged particles (and particles of unknown charge), of unknown energy | 20 | Protons, other than recoiled protons, energy > 2MeV | 5 |
| | | Alpha particles, fission fragments, heavy nuclei | 20 |
| (However, by the statement of ICRP Paris conference in 1985, it is recommended that Q value of neutrons be doubled) | | Others : calculation value of \bar{Q} at a depth of 10mm in ICRU sphere. | |

8. CONCLUDING REMARKS

A total of ten years, seven years preparing documents and three years subsequent reviewing, has finally led to the publication of the Nuclear Criticality Safety Handbook, second version. It is expected that the second version will be used for the criticality safety control of the Rokkasho reprocessing plant. In addition, this document can be used to conduct criticality safety analyses of other nuclear fuel facilities such as fuel transportation, storage, and fuel fabrication (MOX fuel fabrication in particular) facilities. Furthermore, this handbook can be used to promote technical cooperation with other countries organized through the Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA), to help establish international standards through the International Standardization Organization (ISO), and to conduct future technical cooperation with developing countries.

Although the second version of the Nuclear Criticality Safety Handbook contains much new information, there are still many improvements anticipated for future version. Due to various constraints, we were not able to incorporate the most recent nuclear data in the Nuclear Criticality Safety Handbook, second version. That is, although the JENDL-3.2 was published in Japan in 1994 as evaluated nuclear data, the criticality data in this second version were calculated based on the ENDF/B-IV (some criticality data were calculated based on the JENDL-3.1). In the future version of the Nuclear Criticality Safety Handbook, we intend to calculate criticality data more accurately based on the most recent nuclear data available. In doing so, however, we must review the dependency of the criticality calculations with respect to the particular nuclear data set used. In 1995, the Japan Atomic Energy Research Institute (JAERI) has started criticality experiments at the Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF) and has been accumulating the criticality data of cylindrical and slab shape uranium fuel solution systems. Near future, it plans to conduct criticality experiments for plutonium fuel solution systems. In the future version, based on these accumulated data, we intend to improve the accuracy of the criticality calculations of uranium and plutonium fuel solution systems, so that more reasonable criticality safety analyses can be conducted. Furthermore, at the advent of the extended life of light water reactors in recent years, it has been demanded to conduct reasonable safety analyses for the storage and transport of high burnup UO_2 and MOX spent fuels. In the future version, we need to incorporate burnup credit evaluation methods and relevant data for the safety analyses of the high burnup spent fuels. In addition to absorbing and incorporating new knowledge shown above in the Handbook, summarizing the past expertise and clearly specifying the technical bases for them are important as the original role of the Handbook. We shall continue our efforts to resolve these remaining tasks and intend to incorporate the improvements in the future third version.

Many people concerned with nuclear related education, research, and industry provided valuable comments and suggestions for the published version of this document. Although this document was issued under the Working Group on Nuclear Criticality Safety Data, many members in

the Special Committee on Nuclear Criticality Safety Nuclear Fuel Facility Safety Research Committee at the Japan Atomic Energy Research Institute provided both direct and indirect assistance during the various preparatory stages of this document. We wish to express our deepest gratitude to them.

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国際単位系 (SI) と換算表

表 1 SI 基本単位および補助単位

| 量 | 名 称 | 記 号 |
|-------|-----------|-----|
| 長 度 | メ ー ト ル | m |
| 質 量 | キ ロ グ ラ ム | kg |
| 時 間 | 秒 | s |
| 電 流 | ア ン ペ ア | A |
| 熱力学温度 | ケ ル ビ ン | K |
| 物 質 量 | モ ル | mol |
| 光 度 | カ ン デ ラ | cd |
| 平 面 角 | ラ ジ ア ン | rad |
| 立 体 角 | ステラジアン | sr |

表 3 固有の名称をもつ SI 組立単位

| 量 | 名 称 | 記号 | 他の SI 単位 による表現 |
|---------------|-----------|----|---------------------|
| 周 波 数 | ヘ ル ツ | Hz | s ⁻¹ |
| 力 | ニ ュ ー ト ン | N | m·kg/s ² |
| 圧 力, 応 力 | パ ス カ ル | Pa | N/m ² |
| エネルギー, 仕事, 熱量 | ジ ュ ー ル | J | N·m |
| 工 率, 放 射 束 | ワ ッ ト | W | J/s |
| 電 気 量, 電 荷 | ク ー ロ ン | C | A·s |
| 電位, 電圧, 起電力 | ボ ル ト | V | W/A |
| 静 電 容 量 | ファラド | F | C/V |
| 電 気 抵 抗 | オ ー ム | Ω | V/A |
| コンダクタンス | ジーメンズ | S | A/V |
| 磁 束 | ウェーバ | Wb | V·s |
| 磁 束 密 度 | テ ス ラ | T | Wb/m ² |
| インダクタンス | ヘ ン リ ー | H | Wb/A |
| セルシウス温度 | セルシウス度 | °C | |
| 光 束 | ル ー メ ン | lm | cd·sr |
| 照 度 | ル ク ス | lx | lm/m ² |
| 放 射 能 | ベ ク レ ル | Bq | s ⁻¹ |
| 吸 収 線 量 | グ レ イ | Gy | J/kg |
| 線 量 当 量 | シーベルト | Sv | J/kg |

表 2 SI と併用される単位

| 名 称 | 記 号 |
|---------|-----------|
| 分, 時, 日 | min, h, d |
| 度, 分, 秒 | °, ', " |
| リットル | l, L |
| トン | t |
| 電子ボルト | eV |
| 原子質量単位 | u |

$$1 \text{ eV} = 1.60218 \times 10^{-19} \text{ J}$$

$$1 \text{ u} = 1.66054 \times 10^{-27} \text{ kg}$$

表 4 SI と共に暫定的に維持される単位

| 名 称 | 記 号 |
|----------|-----|
| オングストローム | Å |
| バ ー ン | b |
| バ ー ル | bar |
| ガ ル | Gal |
| キ ュ リ ー | Ci |
| レンツゲン | R |
| ラ ヌ ム | rad |
| レ ム | rem |

$$1 \text{ Å} = 0.1 \text{ nm} = 10^{-10} \text{ m}$$

$$1 \text{ b} = 100 \text{ fm} = 10^{-28} \text{ m}^2$$

$$1 \text{ bar} = 0.1 \text{ MPa} = 10^5 \text{ Pa}$$

$$1 \text{ Gal} = 1 \text{ cm/s}^2 = 10^{-2} \text{ m/s}^2$$

$$1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$$

$$1 \text{ R} = 2.58 \times 10^{-4} \text{ C/kg}$$

$$1 \text{ rad} = 1 \text{ cGy} = 10^{-2} \text{ Gy}$$

$$1 \text{ rem} = 1 \text{ cSv} = 10^{-2} \text{ Sv}$$

表 5 SI 接頭語

| 倍数 | 接頭語 | 記 号 |
|-------------------|------|-----|
| 10 ¹⁸ | エクサ | E |
| 10 ¹⁵ | ペタ | P |
| 10 ¹² | テラ | T |
| 10 ⁹ | ギガ | G |
| 10 ⁶ | メガ | M |
| 10 ³ | キロ | k |
| 10 ² | ヘクト | h |
| 10 ¹ | デカ | da |
| 10 ⁻¹ | デシ | d |
| 10 ⁻² | センチ | c |
| 10 ⁻³ | ミリ | m |
| 10 ⁻⁶ | マイクロ | μ |
| 10 ⁻⁹ | ナノ | n |
| 10 ⁻¹² | ピコ | p |
| 10 ⁻¹⁵ | フェムト | f |
| 10 ⁻¹⁸ | アト | a |

(注)

- 表 1-5 は「国際単位系」第 5 版, 国際度量衡局 1985 年刊行による。ただし, 1 eV および 1 u の値は CODATA の 1986 年推奨値によった。
- 表 4 には海里, ノット, アール, ヘクトールも含まれているが日常の単位なのでここでは省略した。
- bar は, JIS では流体の圧力を表わす場合に限り表 2 のカテゴリーに分類されている。
- EC 閣僚理事会指令では bar, barn および「血圧の単位」mmHg を表 2 のカテゴリーに入れている。

換 算 表

| 力 | N (=10 ⁵ dyn) | kgf | lbf |
|---|--------------------------|----------|----------|
| | 1 | 0.101972 | 0.224809 |
| | 9.80665 | 1 | 2.20462 |
| | 4.44822 | 0.453592 | 1 |

$$\text{粘 度 } 1 \text{ Pa} \cdot \text{s} (\text{N} \cdot \text{s/m}^2) = 10 \text{ P (ポアズ)} (\text{g}/(\text{cm} \cdot \text{s}))$$

$$\text{動粘度 } 1 \text{ m}^2/\text{s} = 10^4 \text{ St (ストークス)} (\text{cm}^2/\text{s})$$

| 圧 | MPa (=10 bar) | kgf/cm ² | atm | mmHg (Torr) | lbf/in ² (psi) |
|---|----------------------------|----------------------------|----------------------------|---------------------------|----------------------------|
| | 1 | 10.1972 | 9.86923 | 7.50062 × 10 ³ | 145.038 |
| 力 | 0.0980665 | 1 | 0.967841 | 735.559 | 14.2233 |
| | 0.101325 | 1.03323 | 1 | 760 | 14.6959 |
| | 1.33322 × 10 ⁻⁴ | 1.35951 × 10 ⁻³ | 1.31579 × 10 ⁻³ | 1 | 1.93368 × 10 ⁻² |
| | 6.89476 × 10 ⁻³ | 7.03070 × 10 ⁻² | 6.80460 × 10 ⁻² | 51.7149 | 1 |

| エネルギー・仕事・熱量 | J (=10 ⁷ erg) | kgf·m | kW·h | cal (計量法) | Btu | ft·lbf | eV |
|-------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|----------------------------|
| | 1 | 0.101972 | 2.77778 × 10 ⁻⁷ | 0.238889 | 9.47813 × 10 ⁻⁴ | 0.737562 | 6.24150 × 10 ¹⁸ |
| | 9.80665 | 1 | 2.72407 × 10 ⁻⁶ | 2.34270 | 9.29487 × 10 ⁻³ | 7.23301 | 6.12082 × 10 ¹⁹ |
| | 3.6 × 10 ⁶ | 3.67098 × 10 ⁵ | 1 | 8.59999 × 10 ⁵ | 3412.13 | 2.65522 × 10 ⁶ | 2.24694 × 10 ²⁵ |
| | 4.18605 | 0.426858 | 1.16279 × 10 ⁻⁶ | 1 | 3.96759 × 10 ⁻³ | 3.08747 | 2.61272 × 10 ¹⁹ |
| | 1055.06 | 107.586 | 2.93072 × 10 ⁻⁴ | 252.042 | 1 | 778.172 | 6.58515 × 10 ²¹ |
| | 1.35582 | 0.138255 | 3.76616 × 10 ⁻⁷ | 0.323890 | 1.28506 × 10 ⁻³ | 1 | 8.46233 × 10 ¹⁸ |
| | 1.60218 × 10 ⁻¹⁹ | 1.63377 × 10 ⁻²⁰ | 4.45050 × 10 ⁻²⁶ | 3.82743 × 10 ⁻²⁰ | 1.51857 × 10 ⁻²² | 1.18171 × 10 ⁻¹⁹ | 1 |

$$1 \text{ cal} = 4.18605 \text{ J (計量法)}$$

$$= 4.184 \text{ J (熱化学)}$$

$$= 4.1855 \text{ J (15 °C)}$$

$$= 4.1868 \text{ J (国際蒸気表)}$$

$$\text{仕事率 } 1 \text{ PS (仏馬力)}$$

$$= 75 \text{ kgf} \cdot \text{m/s}$$

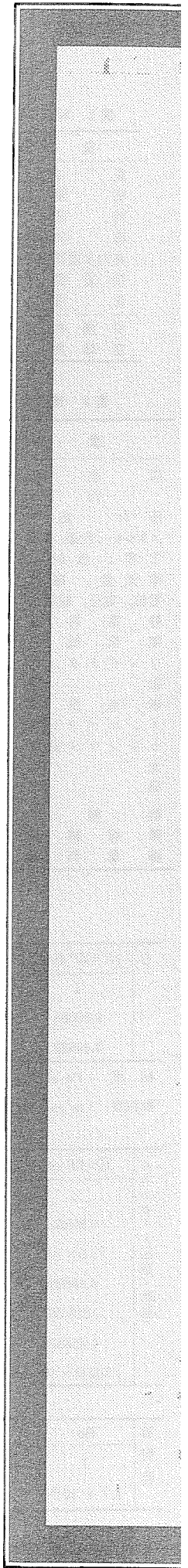
$$= 735.499 \text{ W}$$

| 放射能 | Bq | Ci |
|-----|------------------------|-----------------------------|
| | 1 | 2.70270 × 10 ⁻¹¹ |
| | 3.7 × 10 ¹⁰ | 1 |

| 吸収線量 | Gy | rad |
|------|------|-----|
| | 1 | 100 |
| | 0.01 | 1 |

| 照射線量 | C/kg | R |
|------|-------------------------|------|
| | 1 | 3876 |
| | 2.58 × 10 ⁻⁴ | 1 |

| 線量当量 | Sv | rem |
|------|------|-----|
| | 1 | 100 |
| | 0.01 | 1 |



古紙配合率100%
白度70%の再生紙を使用しています