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RESEARCH PROGRAM (VEGA) ON THE FISSION PRODUCT RELEASE FROM IRRADIATED FUEL

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Research Program (VEGA) on the Fission Product Release from Irradiated Fuel

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Fission product release from overheated fuel is one of the key phenomena controlling the source terms in hypothetical severe accidents. In-pile and out-of-pile heating tests of the fuel have been conducted to study the release behavior in the world. In-pile tests covered a wide range of accident conditions up to fuel melting under high pressure. The uncertainty and variation of the test conditions in the in-pile test, however, made the interpretation very difficult. The out-of-pile tests under better controlled conditions, on the other hand, was limited below 2425℃ under the atmospheric pressure. Thus, considerable uncertainties still remain, especially in the areas under high temperature and/or high pressure conditions.

An experimental and analytical program, Verification Experiment of Gas/Aerosol release (VEGA), was initiated at JAERI to study the fission product release behavior from light water reactor fuels irradiated in Japanese power reactors. A short fuel segment will be inductively heated to high temperatures in a hot cell under simulated accident conditions. In the program, less understood release and transport behavior of low volatile and short life fission products will be studied,

1. under high temperature conditions, covering melting temperature of the fuel, and,
2. at high pressure conditions up to1.0 MPa.

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The program will be carried out keeping a close corporation with the NSRR RIA program, sharing the test fuel specimens and examination results. This way, irradiated fuel behavior and characteristics under various accident conditions are studied comprehensively. This report describes background, status and outlines of the VEGA program.

Keywords: Fission Product Release, Severe Accidents, Induction Heating, Source Term,

照射済燃料からのFP放出実験計画(VEGA)

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原子炉のシビアアクシデント時のソースターム（環境に放出される核分裂生成物(FP)の量と化学形）を評価する上では、燃料からのFP放出挙動の評価が重要である。このため、炉内および炉外実験による研究が世界的に進められてきた。炉内実験では燃料溶融や高圧条件下を含む広範な事故条件での実験が行われたが、実験条件のばらつきが大きく精度に問題があり、評価が困難である。他方、実験条件が明確な炉外実験は2425℃以下の大気圧条件で実施されている。このため、燃料の融点を超える高温あるいは高圧条件の放出データを中心に大きな不確実性が残っている。

これらの点を明確にするため、原研では日本の発電用軽水炉で照射された燃料を用いてFPの放出・移行挙動を調べるVEGA実験計画を開始した。同実験では、ホットセル内で短尺の燃料を事故を模擬した条件で高温まで誘導加熱する。この計画では、下記の条件を中心にデータの少ない低揮発性および短半減期のFPに注目して、その放出及び移行挙動を調べる実験を行う。

1. 燃料の融点を超える高温条件、
2. 1.0 MPaまでの高圧条件

本実験は、出力暴走時の燃料挙動を調べるNSRR反応度事故実験計画と実験燃料および燃料検査結果等を共有し、密接な関係で行う。これにより、燃焼の進んだ燃料の広範な事故条件での特性および挙動を総合的に調べることが可能となる。本報告書はVEGA実験について、その背景、位置付け、計画の概要をまとめたものである。

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1. INTRODUCTION

1.1 Background

Numbers of hot cell experiments were conducted to investigate fission product (FP) releases from reactor fuels. Many of the tests, however, were conducted at relatively low temperatures to investigate fission gas behavior under normal operating conditions. After the accident at the Three Mile Island Unit 2 reactor, more attention was paid on the tests at higher temperatures under accidental conditions. In this context, HI/VI tests[1, 2] were started in Oak Ridge National Laboratory (ORNL) of USA with irradiated fuel segments of 15 to 20cm long under simulated accident conditions in steam, hydrogen and air atmosphere at temperatures up to 2425℃. Schematic configuration of the VI test facility is shown in **Fig. 1**. The fuel specimen was inductively heated in a furnace and decrease of radio activity was on-line monitored. Fission products from the fuel deposited on thermal gradient tubes (TGTs), and aerosols are filtered in a sequential way. Gaseous fission products were trapped in condenser or cooled charcoal traps. These TGTs, filters and traps were also on-line monitored by radiation detectors to measure release kinetic of the FPs. Measurable fission products by radioactivity, however, were limited, because the fuel had fairly long cooling time of a few to ten years before the heating tests.

A similar test program has been carried out at Centre D'Étude Nucléaire de Grenoble (CENG) of France in HEVA/VERCORS tests [3, 4]. This program is extended to VERCORS-HT to cover high temperatures of UO_2 melting. The first test of VERCORS-HT was conducted in 1996, though the test was not totally successful and the data were not published yet. In the HEVA/VERCORS tests, re-irradiation of the test fuel was conducted just before the heating tests to generate short life FPs. Other series of tests, such as UCT, UCE and HCE[5, 6] have been conducted at Chalk River Laboratories (CRL) of AECL Research of Canada focusing on the fuel oxidation effects by steam and air. Bare fuel fragments or short cladded fuel specimens of about 30g were used in the tests. These out-of-pile FP release test programs are summarized in **Table 1**.

In addition to these hot cell experiments, numbers of in-pile tests were conducted using test reactors. Examples of the experiments are listed in **Table 2**. Two source term (ST) tests[7, 8] were conducted at Annular Core Research Reactor (ACRR) of Sandia National Laboratories (SNL) of the U.S.A. The tests, ST-1 and 2,

were conducted under reducing atmosphere at temperatures around 2200℃ at different system pressures to examine the pressure effect on the fission product release. Schematic of the ST-1 test facility is shown in **Fig. 2**. The on-line kinetic measurement of the FP release was not available in the test ST-1. The kinetic data was obtained only from 7 TGT/filters (aerosols) and 5 grab sampler (gases). A hot cell test, VI-4[9] at ORNL in **Table 1**, was conducted under a duplicating test condition to ST-1 to examine the difference between the in-pile and the out-of-pile tests. Releases in the in-pile tests could be larger due to fissioning induced lager diffusivity of the FPs in the fuel matrix, though no significant difference was observed in these tests.

A comprehensive test program, to research not only the fission product release but also the core degradation behavior, with a fuel bundle of 32 rods was performed in severe fuel damage (SFD) tests[10] in the Power Burst Facility (PBF) at the Idaho National Engineering Laboratory (INEL) of the U.S.A. The last test, SFD 1-4, of the four test series was conducted with 26 irradiated fuel rod at burnups up to 42GWd/tU and 4 Ag-In-Cd control rods. The test atmosphere was limited steam at high pressure of 6.95MPa to cause 32% of the Zircaloy to be oxidized by the steam. Another comprehensive test program was initiated at Phebus reactor at Centre Étude Cadarache of Institut Sûreté Nucléaire (IPSN) in France. The Phebus-FP program[11] is intended to study all the phenomena associated to the source term, i.e. starting at fission product release from the degraded core, to transport of the FPs to the containment, and behavior in the containment. Test conditions of these in-pile experiments are listed in **Table 2**.

These in-pile tests cover high temperatures up to melting temperature of UO_2 fuel under various atmosphere and pressure conditions. However, fuel temperature and atmosphere varied locally in the test sections and also time-dependently during the progression of the core degradation in these integral tests. Thus, release correlation or models of a function of temperature and/or atmosphere etc. are hardly derived only from the integral tests. These tests would serve better for verification of the integral models or a code system. Thus, out-of-pile tests under controlled conditions are needed for developing the models.

1.2 Objectives of VEGA Program

An out-of-pile separate effect test program, Verification Experiment of Gas/Aerosol release (VEGA), was initiated to realize isothermal heating of small fuel specimen under well controlled atmosphere conditions beyond the conditions covered

by earlier out-of-pile tests. The program is focusing on the area where accurate release data are missing, i.e.

1. fission product releases under high temperature conditions, covering melting of fuel
   - up to 3000℃ under oxidizing and reducing atmosphere (using ThO_2 crucible),
2. fission product release and transport under high pressure conditions
   - up to 1MPa, and
3. release and transport of short life fission products
   - by re-irradiation of test fuel at the Nuclear Safety Research Reactor (NSRR).

In addition, the effects of other parameters such as, inert, oxidizing/reducing atmosphere, fuel burnup, etc. on the FP release behavior will be investigated. In the reducing atmosphere tests, fuel dissolution by molten cladding is one of other interesting subjects to be studied, which has not been well understood especially for the irradiated fuels. These data would be utilized for the modeling of FP release and transport models, which currently have uncertainties of a few orders of magnitudes especially for low volatile FPs.

Judging from the review of currently available data in the following section, semi-mechanistic models, which at least take account of diffusion release process in the fuel grains, chemistry and vaporization process on the fuel surfaces, seems to be needed for more accurate source term estimation. The current understanding of the FP release behavior and its controlling or affecting parameters are discussed and summarized in the following section.

2. REVIEW AND ANALYSES OF EARLIER STUDIES

2.1 Parameters Affecting Fission Product Release

The fission product release is believed to be primarily a function of temperature. In addition, other parameters such as, fuel oxidation, burnup, pre-transient conditions, are found to affect the FP releases considerably from the earlier studies. The fuel oxidation is well known to enlarge diffusivity of fission gases in the UO_2 matrix[12, 13]. Many of the Canadian tests are dedicated to clarify the effect, then the oxidation and release kinetics were fairly well understood[6]. In the realistic

geometry of the reactor core, however, Zircaloy cladding would serve as protecting boundary for the fuel against the oxidation. Thus, the fuel oxidation effect would be mitigated depending on the fuel damage progression. In this section, the parameters which affect the fission product releases are discussed.

In order to demonstrate these parameter effects, releases of kripton-85 (Kr-85) and cesium-137 (Cs-137) in ORNL/HI-VI tests are assembled in **Figs. 3** and **4** in terms of apparent diffusion coefficients as a function of temperature. The apparent diffusion coefficients are calculated by reversal use[14, 15] of the simple Booth diffusion model[16], which assumes intragranular atomic diffusion in a hypothetical spherical UO_2 grain. The fractional release, F, and diffusion coefficient, D[m^2/s], are expressed by following equations with grain radius, a[m], and time, t[s].

$$F = 6\sqrt{\frac{D't}{\pi}} - 3D't \cdots\cdots\cdots (D't \le 0.1)$$
$$F = 1 - \frac{6}{\pi^2}\exp(-\pi^2 D't) \cdots\cdots (D't \ge 0.1) \qquad (1)$$

where

$$D' = \frac{D}{a^2}.$$

Another way of expressing the release is release rate coefficient, k[min^{-1}], which is a fractional release to the current inventory during an unit time period. Then, k is expressed as,

$$k = \frac{dF}{dt} = \frac{-\ln[1-(F_2-F_1)/(1-F_1)]}{t_2 - t_1} \approx \frac{F_2 - F_1}{(1-F_1)(t_2 - t_1)}, \qquad (2)$$

where

F_1= fraction of original inventory released at start of the time period,

F_2= fraction of original inventory released at end of the time period,

t_1= time corresponding to F_1,

t_1= time corresponding to F_2.

In both two models, the diffusion coefficient and release rate coefficient are treated as a function of temperature in codes and models[15-20]. The diffusion model could fit better for volatile FPs, the release of which would be controlled by the diffusional process in the UO_2 matrix. On the other hand, the release rate coefficient could be a reasonably good approximation when the release was controlled by vaporization like of low volatile FPs. In addition, the release rate coefficient is easier to be used and be understood.

(1) Effect of fuel burnup

The apparent diffusion coefficients of Kr-85 and Cs-137 in **Figs. 3** and **4** are in good agreement with those of stage IIA and IIB in a review by Matzke[13], suggesting that the releases are dominated by the atomic diffusion in the grains and they are primarily a function of temperature. The variation of the coefficients, however, could indicate the effects of other parameters. **Figure 5** highlights difference in the coefficients between high burnup(40GWd/tU or higher) and low/intermediate burnups (30GWd/tU or less). The figure indicates larger releases for higher burnup fuels. Best fit diffusion coefficients for the two bumup groups in the HI/VI tests are shown in **Figs. 3** and **4**. Similar correlations for burnup dependence were used in other models such as, ANS5.4[17] and CORSOR-BOOTH[18] models. However, magnitude of the effect is different among the models, and none of the models has fully verified to high burnups above 50GWd/tU.

(2) Effect of atmosphere

The reactor fuel could be heated up in oxidizing atmosphere (steam or air) or in reducing atmosphere (hydrogen), depending on the sequences, time and location during severe accidents. This atmosphere variation could cause significant change of FP release and transport behavior. The oxidation of UO_2 can enhance diffusivity of FPs in fuel matrixes. The oxidation potential can alter the chemical forms of the reactive FPs, which subsequently change the mobility of the FPs significantly. On the other hand, the chemical reaction of UO_2 with core materials could cause fuel melting at lower temperatures than the melting point of UO_2, which changes core configuration and FP release drastically. In addition, metallic tellurium (Te) reacts with cladding metal and its release starts when all the cladding has been oxidized. In case of ruthenium (Ru), the release becomes significant when ruthenium is oxidized by air. In the similar way, releases of antimony (Sb), molybdenum (Mo) and silver (Ag) are enhanced in the oxidizing atmospheres. On the other hand, the releases of barium (Ba), strontium (Sr) and europium (Eu) are enhanced in reducing atmosphere. These difference on release rates, especially of the short life fission products like Ba, could make significant effects on decay heat source distributions of the core or the pipings, which could alter the accident scenario in some cases. However, these atmosphere effects are understood only qualitatively, due to the rack of quantitative data.

The effect of fuel oxidation on fission gas release is better understood and is illustrated in **Fig. 6** by comparing HI-2[19] and HI-3[20] tests, where identical fuel specimens were used. In HI-2 test, cladding had fracture opening during the heating

test. Then, considerable fuel oxidation was believed to take place in the test[21]. In the other HI/VI tests, however, the oxidation effect was not evident. Though, HI/VI test lasted only for 20 to 60min, fuel could have suffered considerable oxidation after all the cladding metal has consumed, if the tests lasted longer period. Further investigation seems needed to have proper modeling of cladding effect for long term heating on the fuel oxidation.

In addition to the fuel oxidation, chemical interaction of UO_2 to molten metals is pointed out to be important[21]. This interaction results in fuel dissolution at temperatures considerably below melting temperature of UO_2 itself. In addition to the UO_2 dissolution by metallic melts, recent Phebus test results suggested that the low temperature melting of UO_2 with oxidized cladding, ZrO_2, could also cause significant core degradation at temperature about 2500℃. These reaction and subsequent reactor core degradation could cause enhanced FP releases.

In order to cover all these parameters, numbers of tests and examinations are not sufficient enough to make comprehensive models for reliable source term estimation.

(3) Effect of steady state linear heat rate

Test HI-4[22] is compared to HI-3 in **Fig. 7**, in order to illustrate the effect of the pre-transient fuel temperature on the FP release behavior. The test fuel used in HI-4 had relatively large in-pile fission gas release of 10.2% before the heating test. Relatively high linear heat rate and subsequent high fuel temperature during irradiation of HI-4 fuel was responsible for the large release. This high temperature suggested larger accumulation of the fission gases in the grain boundaries to cause larger release during the heat-up stage of the test. This effect, however, was limited in the heat-up stage of the test and less important in high temperature conditions.

The grain boundary inventory is important for detailed mechanistic modeling of the fission gas release, in the codes such as FASTGRASS[23], FEMAXI-IV[24] and VICTORIA[25]. In addition, the grain boundary fission gas is believed to cause fuel expansion when the fuel was rapidly heated up in power burst type accidents such as reactivity initiated accident (RIA). However, very few grain boundary inventory measurements have been done so far. Only a few limited data are available from Canadian tests. In VEGA program, the grain boundary inventory is going to be measured in air ingress tests. With selective oxidation of UO_2 by air in the grain boundaries, the release of grain boundary fission gas occurs at relatively low temperature of 500℃ and can be separated from the release of gases in UO_2 matrix.

(4) Effect of ambient pressure

It was pointed out by Sandia National Laboratories (SNL) that the ambient pressure could affect the FP release rate from fuel. The recent calculations with the VICTORIA code[25] showed that the higher the ambient pressure becomes the lower the FP release rate is. The VICTORIA code calculates the FP release from a fuel rod basically using the diffusion theory. The code models the FP release process which consists of 1) diffusion inside the UO_2 grain (intragranular transport), and 2) diffusion or convection on the grain boundary (open porosity) which is finally connected to the fuel pellet surface.

Schematic of the VICTORIA model for FP release process is shown in **Fig. 8**. This figure indicates that the time for FP diffusion in fuel grain ($L^{2/D}$) at 2500 K and 3000 K are about 1 and 10 seconds, respectively. On the other hand, the time for FP diffusion at fuel pores ($L^{2/D}$) at 2500 K and 3000 K are equal to $0.4 \times$P(bar) = $\sim 10^4$ seconds and $0.3 \times$P(bar) = $\sim 10^4$ seconds, respectively. From the comparison of FP diffusion time between in the fuel grain and at the fuel pores (open porosity), it can be expected that the FP release process could be governed by the FP diffusion at fuel pores which may become a rate-determining step in these diffusion processes. Therefore, if the FP diffusion process at fuel pores can be described as a function of pressure as in the following VICTORIA model, the FP release could be affected by ambient pressure.

In the VICTORIA modeling, the intragranular FP transport is calculated using the Booth model or the FASTGRASS model[23] while the FP diffusion or convection at an open porosity is calculated using the Chapman-Enskog model[26] which is a function of ambient pressure. The averaged diffusion coefficient for a mixed gas (binary system, i, j) in the Chapman-Enskog model is given by Eq. (3) .

$$D_{i,j} = 0.018824 \frac{[T^3(\frac{1}{M_i} + \frac{1}{M_j})]^{0.5}}{P \cdot s_{j,j}^2 \cdot W_{i,j}}, \qquad (3)$$

where

$D_{i,j}$: mass diffusivity coefficient (m^2/s),

T : temperature (K),

M_i, M_j : atomic weight of gases i and j (g),

P : pressure (bar),

$s_{i,j}$: Lennard-Jones parameter (Å), and

$W_{i,j}$: dimensionless function of temperature and intermolecular potential field.

It is noted that the mass diffusivity coefficient is in inverse proportion to the pressure. This indicates that the FP gases diffused to the grain boundary could be transported slower in the high pressure open porosity than that in the low pressure open porosity. The preliminary calculations with VICTORIA by SNL[27] on the effect of pressure on the FP release rate is shown in **Fig. 9**. In the calculation, the FP release rates for different pressure were calculated by normalizing the FP release rate at 2.2 bar. The results showed that the effect of pressure for low volatile FPs is significant while not much for volatile FPs.

Since it is expected in typical severe accidents that the FP release occurs under high pressure condition, the investigation on the pressure dependence is considered to be important to evaluate precisely the source term.

2.2 Release of Low Volatile Fission Products and Actinides

The release behavior of volatile fission products, such as Cs, Kr and Xe, is fairly well studied in ORNL, CRL and CENG works. Still, the effects of the test conditions on the release have not enough understood as discussed above. Much less is understood for low volatile fission products and actinides, because only limited experimental data are available for them. The release rate coefficients, which is a fractional release rate in a minute, for low volatile strontium (Sr) and plutonium (Pu) are shown in **Figs. 10** through **13**, as examples[28]. The diffusion coefficients in the same figures are about 2 to 8 orders of magnitude smaller than those of volatile FPs. Strontium 90 is hazardous as a strong β emitter which has a half life of 29.12 year and has accumulated to a similar amount as Cs-137 or 134. The release rates coefficient of strontium measured in various test conditions in VI, ST and PBF tests scattered a lot by 2 to 3 orders of magnitude. The coefficients tends to be larger in reducing atmosphere, in which Sr can be released as a metal. The release rate coefficient of Sr could be smaller just by a factor than those of volatile FPs in the reducing atmosphere. While, it would be 2 to 3 orders of magnitude smaller in oxidizing atmosphere.

The fewer release rate measurements of Pu are available and have larger scatter of 3 to 4 orders of magnitude in **Figs. 12** and **13**. The release of Pu is believed to occur in a process of fuel vaporization. Thus, flow rate of the atmospheric gas in addition to the fuel temperature is important for the release.

2.3 Summary

From the review and analyses of the earlier studies discussed in this section, it was found that the atmosphere effect is significant for releases of many FPs. The release rates could be different by a few orders of magnitude. This difference is partly due to the unknown experimental conditions, such as the local temperature, the local atmosphere (oxygen/hydrogen partial pressure), and local flow rate. The experimental limitation on the accuracy of release measurements also contributed to the large uncertainties. In addition, release data at high temperatures from molten fuel and under high pressure conditions are not available practically. In order to reduce these uncertainties, a series of tests under well characterized conditions are needed, which are expected to be realized in the VEGA program. Relying on the test results, semi-mechanistic models, which take account of diffusional release process in the fuel grains, the atmosphere effects on the chemical forms of the FPs and vaporization process on the fuel surfaces, are needed to be developed for more accurate source term estimation.

3 VEGA TEST PROGRAM

An experimental facility with an induction furnace, thermal gradient tubes(TGTs), filter sets, cascade impactor and traps for gases, has been built in a hot cell No. 5 at Reactor Fuel Examination Facility (RFEF) in Tokai establishment of JAERI. **Figure 14** shows rough schematic of the VEGA experiment facility and procedure. In the RFEF, a comprehensive post irradiation examination (PIE) is conducted to commercial PWR/BWR fuels and to MOX fuel from the Advanced Thermal Reactor(ATR), Fugen. In course of the PIEs, segmentation of the reactor fuel will be conducted. The segmented test fuel will be transferred to the Nuclear Safety Research Reactor(NSRR) for re-irradiation. The re-irradiation will be made at a low linear heat rate of about 10 to 30 W/cm for a few hours by steady state operation of the NSRR at a power of 300kW. Isothermal heating test of the segment under controlled atmosphere of steam/hydrogen/helium/air will be conducted in a few days after the re-irradiation. Fission product release will be measured by on-line monitoring of gamma-rays from fuel segment, filters and traps. A set of comprehensive examination on the segment and deposition/leached samples from the system will take place in the connecting hot cells. The various test conditions and the comprehensive examinations are essential to clarify the effects on FP releases of the parameters discussed in Section 2.

Feasibility study and preliminary designing of the facility for the VEGA program was initiated in 1991. Heating tests and fabrication tests of the thoria components were conducted in the first 4 years. Starting in 1995, final designing of the facility and preparation for safety assessment were performed to start the in-cell experiment in 1998. A time table for the program is shown in **Table 3**.

The program will be carried out in close connection to the NSRR reactivity initiated accident (RIA) test program[29, 30] which studies the fuel performance under power burst conditions. The VEGA and the NSRR RIA programs share the test fuel specimens, pre-test examinations and test results. With the two programs, irradiated fuel performance under various accident conditions, i.e. at slow and very fast heat up rates and in gaseous and liquid water atmospheres, will be studied comprehensively. Characterization of the test fuel rods after the irradiation in the commercial reactor and after the tests simulating the accident conditions will enlarge our understanding on the irradiated fuel performance.

3.1 Test Facility

An induction furnace, which is capable to heat up a fuel specimen up to 3000℃, was built in a hot cell of Reactor Fuel Examination Facility (RFEF) as shown in **Fig. 14**. The test specimen will be heated in (1) steam + helium, (2) hydrogen + helium, (3) steam +hydrogen + helium or (4) air + helium, under pressures up to about 1.0MPa. The specification of the furnace is listed in **Table 4**. A set of thermal gradient tubes (TGTs), filter packages, traps for gaseous fission products (FPs) and gamma detectors for measuring the FPs will be facilitated in the VEGA apparatus. A set of cascade impactor will be operated for a certain period to measure aerosol size distribution. The filters, the traps and the furnace are equipped with gamma detectors separately for online measurements of fission product releases and accumulation on the each components.

In the furnace, a zirconia (ZrO_2) tube and crucible containing the fuel specimen will be surrounded by graphite (C) tube suceptor and graphite felt insulator to realize the isothermal heating. The zirconia compornents will be replaced by thoria (ThO_2) for the tests at temperatures of 2400℃ or higher. The outlet tubings from the furnace to the TGT will be kept above 750℃ to minimize fission product deposition on the tube. Three TGTs will be operated sequentially to obtain time dependent deposition behavior of the FPs on the liner tubes at temperatures from 750℃ to 200℃.

The cascade impactor could be operated at a temperature range from 750℃ to 200℃ at any time during a short period to measure aerosol size distribution at a range from 0.5 to 10 μ m. Fission product aerosols will be trapped by three filter packages at a temperature of 200℃ following the TGTs. Fission gas would be trapped by a packages of condenser, dryer and cooled charcoal trap. Online gamma measurement will be done for the furnace, the filters, the condenser and the charcoal trap, to obtain release kinetic data.

3.2 Test Procedure

Long size commercial fuel rods is going to be segmented in hot cells of RFEF. Gamma spectrometry and ion mass spectrometry, in combination with ORIGEN2[31] calculation, would be the primary source for estimating fission product inventories before and after the tests. A set of other examinations, such as metallography and Secondary Electron Micrography (SEM)/ Electron Probe Micro Analysis (EPMA), will also be conducted to reference samples to have better understanding of the fuel condition before and after the tests.

The short segment fuel will be seal welded for re-irradiation in the NSRR to create short live fission products. A short, for about 8 hours, irradiation at a low linear power level of 10 to 30W/cm is expected to be done in the NSRR. The re-irradiation would create 0.1 to 0.01 of I-131 in comparison with Cs-137. The re-irradiations expected to be performed in one out of four tests each year.

The assembling and disassembling works for the heating tests will be performed by remote handling, in order to perform four tests each year. The online measurements of gamma intensity, temperature, oxygen and hydrogen, etc. are going to be realized in the VEGA tests.

Comprehensive post test examinations are expected on the test segment after solidification by resin. Detailed gamma spectrometry, metallography, Secondary Ion Mass Spectrometry (SIMS), SEM/EPMA are expected for the segment. The fission product collection components will also be individually gamma measured. Basic and Acid leaching and solution analysis is to be conducted for low or non radioactive nuclide measurement. Release of actinides, such as U and Pu, is being evaluated with the solution analyses. The outline of the test procedure is illustrated in **Fig. 15**. Main items and techniques for the measurements are listed below.

Pre-test Examination

Gamma Spectrometry (Axial and radial scanning)
Metallography (Optical examination)
Secondary Ion Mass Spectrometry (SIMS)
SEM/EPMA

Re-irradiation in the NSRR

Steady state irradiation for a day

Heating Test

Online gamma spectrometry
Temperature measurement
Oxygen measurement
Hydrogen measurement

Post-test Examination

Gamma Spectrometry (Axial and radial scanning, each components, solution)
Metallography (Optical examination)
Secondary Ion Mass Spectrometry (SIMS)
SEM/EPMA
Ion Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) (for solutions)

4 TEST SCHEDULE AND MATRIX

For the test fuel rods, 5 types of PWR rods and 3 BWR rods with burnups raging from 26 to 56GWd/tU are ready to be used for the VEGA program. Other fuels at extended burnups are expected to be available in a few years in connection to the NSRR RIA test program. Basic designing of the facility, mockup test of the furnace, scoping tests for fabricating the Thoria tube have completed by 1992. Detailed designing and fabrication of the facility are going to start in 1995 and will finish in 1998. In parallel to this, development of the thoria components for the furnace will also be conducted by the time. Preliminary test schedule and test matrix from year 1998 to 2000 are shown in **Table 3**.

Location of volatile fission products, such as Kr, Xe, Cs, I, would be different depending on the irradiation condition prior to the transient heating tests. Some information could be obtained by combination of micrography, EPMA and X-Ray Fluorescence (XRF) analyses. However, analysis by XRF to obtain intra- and inter-granular Xe distribution is not currently available at JAERI. These information is

important to estimate larger releases at early stages of the heating tests at lower temperatures, which sometimes is called burst release. Grain boundary fission gases which have migrated through the grains during the steady state irradiation is believed to cause this burst release. For quick heat up conditions like in NSRR RIA tests, the grain boundary inventory is important for the gas release and fuel deformation. However, very limited data is available for the grain boundary inventory. A few VEGA tests will serve for the evaluation of the grain boundary inventory. Relatively low temperature heating of UO_2 pellet at 500℃ in air will cause selective oxidation at the grain boundaries and subsequent gas release in the tests.

In addition to the heating tests of the fuel segments, re-heating test of fuel debris from TMI-2 core is planned, in order to examine FP release from once molten debris at higher temperatures.

5. SUMMARY

Preparation for a series of fission product release and transport experiments, VEGA, is in progress at the Tokai Establishment of Japan Atomic Energy Research Institute. Preliminary heating tests and facility construction are on going to start the first test in early 1999. In the program, fission product release behavior under high pressure (≦1MPa) and high temperature (≦3000℃) conditions will be studied in well controlled atmospheres. The program will provide comprehensive data base on fission product release and transport behavior to support the modeling for the source term evaluation. The program will focus on less understood part of fission product release behavior, such as, pressure effect and high temperatures releases, specially of low volatile FPs and actinides. Re-irradiation of the fuel specimen in the NSRR would enlarge measurable FPs of short lives. A few tests would be performed to evaluate grain boundary fission gas inventory, which has hardly been measured and is important for the modeling of the release mechanisms both for the fast transient like RIAa and the severe accidents.

ACKNOWLEDGMENTS

The facility designing and fabrication is organized in corporation with a project team from Mitsubishi Kakoki Kaisha, Ltd lead by Mr. S. Goto. Noticeable support

and suggestions have been made to make the facility well adapted in the hot cell by Messrs. T. Kodaira, T. Yamahara, T. Sukegawa, H. Amano, F. Kanaizuka and their staff members in Department of Hot Laboratories. The program has been realized in Department of Reactor Safety Research headed formerly by Drs. Fujishiro and Akimoto and currently by Dr. K. Abe. Contribution from them and their staff members for supporting the program is highly acknowledged. Valuable suggestions and comments were made by Mr. M. F. Osborne of Oak Ridge National Laboratory for the basic designing of the facility. His support is acknowledged.

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Table 1 Outlines of out-of-pile fission product release experiments

Test series	Atmosphere	Test fuel	Temperature	Main results	Status
HI/VI (ORNL, USA)	steam hydrogen air (0.1MPa)	segment (15-20cm) (10-47GWd/t)	1400-2425C	・Comprehensive release data on Kr, Cs. ・Limited amount of I in gaseous form. ・Less volatile releases depending on atmosphere.	terminated
HEVA/ VERCORS (Grenoble, FRANCE)	steam hydrogen (0.1MPa)	segment (3 pellets) (0-55GWd/t)	1630-2350C	・Release kinetics of I, Ba, Te, etc. by re-irradiation. ・Aerosol size measurements.	VERCORS-HT to be started up
UCT/ UCE/ HCE (CANADA)	air, steam argon (0.1MPa)	fragments segment (<30g)	400-2600C	・Comprehensive study of effect of oxidation by steam and by air.	on going

Table 2 Outlines of in-pile fission product release experiments

Test series	Atmosphere	Test fuel	Temperature	Main results	Status
ST-1 ST-2 (SNL, USA)	H_2 (0.2MPa) H_2 (2MPa)	bundle: 4 irradiated segments(15cm, 47GWd/t) on top of fresh fuel	2215C 2140C	・Comparison of in-pile (ST-1) and out of pile (VI-4) experiment. ・Pressure effect study (ST-1 vs ST-2).	terminated
PBF 1-4 (INEL, USA)	steam (7MPa)	bundle : 26 irradiated +2 fresh+ 4cr (100cm) (29-42GWd/t)	< 2725C (estimated with SCDAP/ RERAP5)	・Overall views including, fuel degradation, transportation, locally dependent conditions. ・FP releases comparable to other tests and models.	terminated
Phebus-FPT0	steam (0.2MPa)	bundle: 20 fresh + 1 cr (100cm) (trace irradiation)	< 2800C (estimated with ICARE2)	・Overall views including transportation in pipes and the containment. ・Considerable fuel degradation and gaseous I release into the containment.	on going.

irradiated: irradiated fuel rod

fresh: fresh (unirradiated) fuel rod

cr: control rod

Table 3 Time table and test schedule of VEGA project.

FY*1	1991	92	93	94	95	96	97	98	99	2000
Designing	Pre.	Basic				Detail				
Furnace	Mockup	Scoping Test			Fabrication			Hot Cell Test		
ThO_2 Fabrication		Scoping Test			Development of the Fabrication Technique				Fabrication for Experiments	
Construction							Cold*2	Hot Cell		
Number of Tests								1	4	4
Test Conditions Temperature (Atmosphere/ Pressure/ Remarks)								2025° C (st/LP)	2400° C(St/LP) 2400° C(St/HP) 500/1200° C(Air/LP) 2800° C(He/LP/TMI)	500/1200° C (Air/LP/NS) 2400° C(H_2/LP) 2800° C(St/LP) 2800° C(St/HP)

*1 Japanese ficical year (FY) starts on April 1 and ends on March 30 in the following year.

*2 Preliminary setup in cold area before installation in the hot cell.

St: steam atmosphere, H_2:hydrogen atmoshere, He:helium atmosphere, Air: air atmosphere

LP: low pressure about 0.1MPa, HP: high pressure condition about 1.0MPa

NS: with re-irradiation in the NSRR for short life fission product generation

TMI: TMI core recovery debris for sample

Table 4 Specification of induction furnace for VEGA tests

Item		Specification	Remarks
Sample	length, mm	≦60	
	diameter, mm	≦14.3	
	mass, g	≦100	
Temperature	maximum, ° C	≦3000	
	duration, min	≦60	
	heat up rate, ° C/s	1 -10	programmed control
	induction power	≦40kW	
Atmosphere	flow gas	H_2O+(H_2)+He H_2+He He Air+He	ThO_2, ZrO_2 W, ThO_2, ZrO_2 W, ZrO_2 ThO_2, ZrO_2
	flow rate, mol/min	0.1 -10.0	
	system pressure, MPa	0.1 -1.0	±0.01

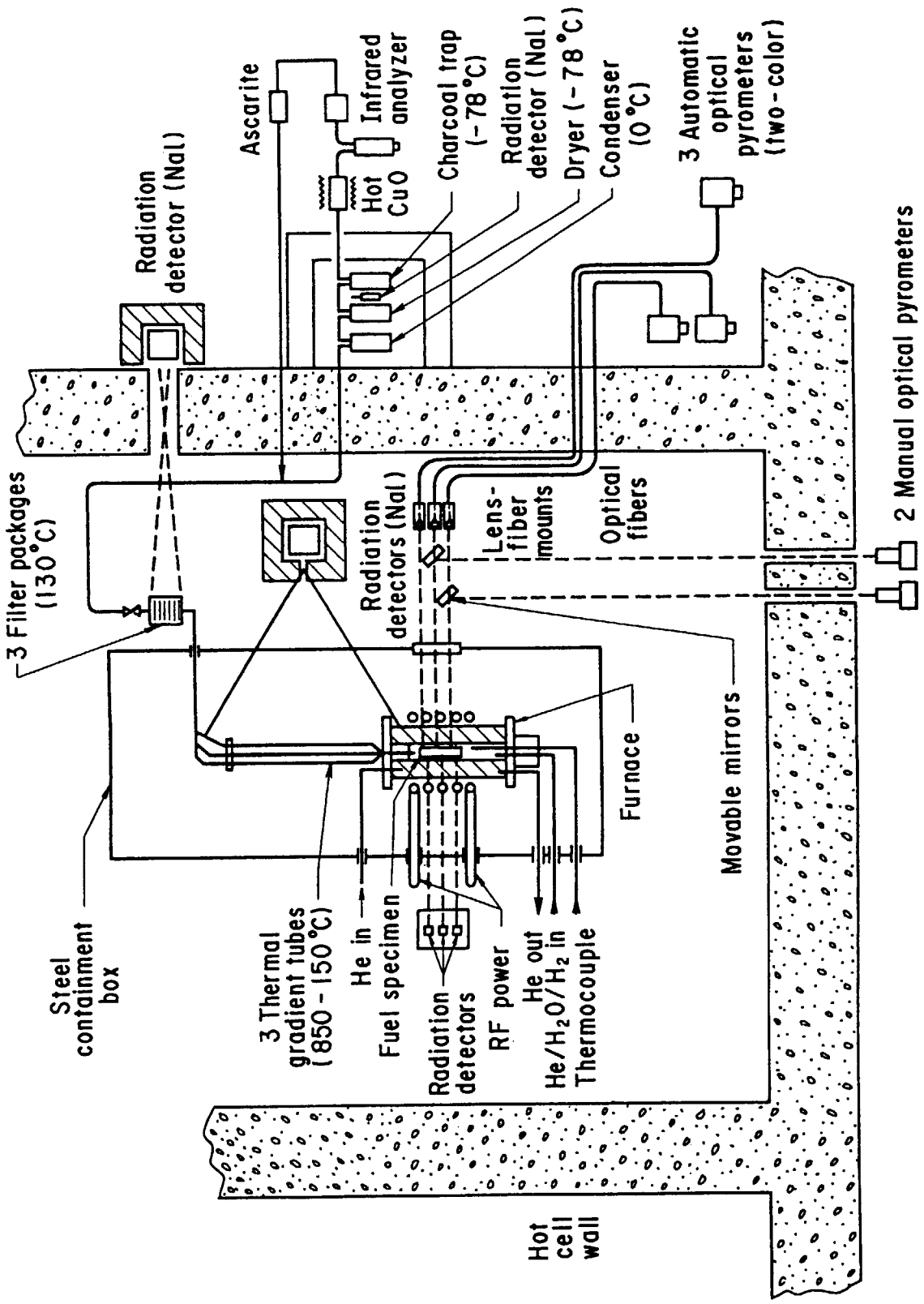

Fig. 1 Schematic configuration of ORNL VI fission product release tests [2].

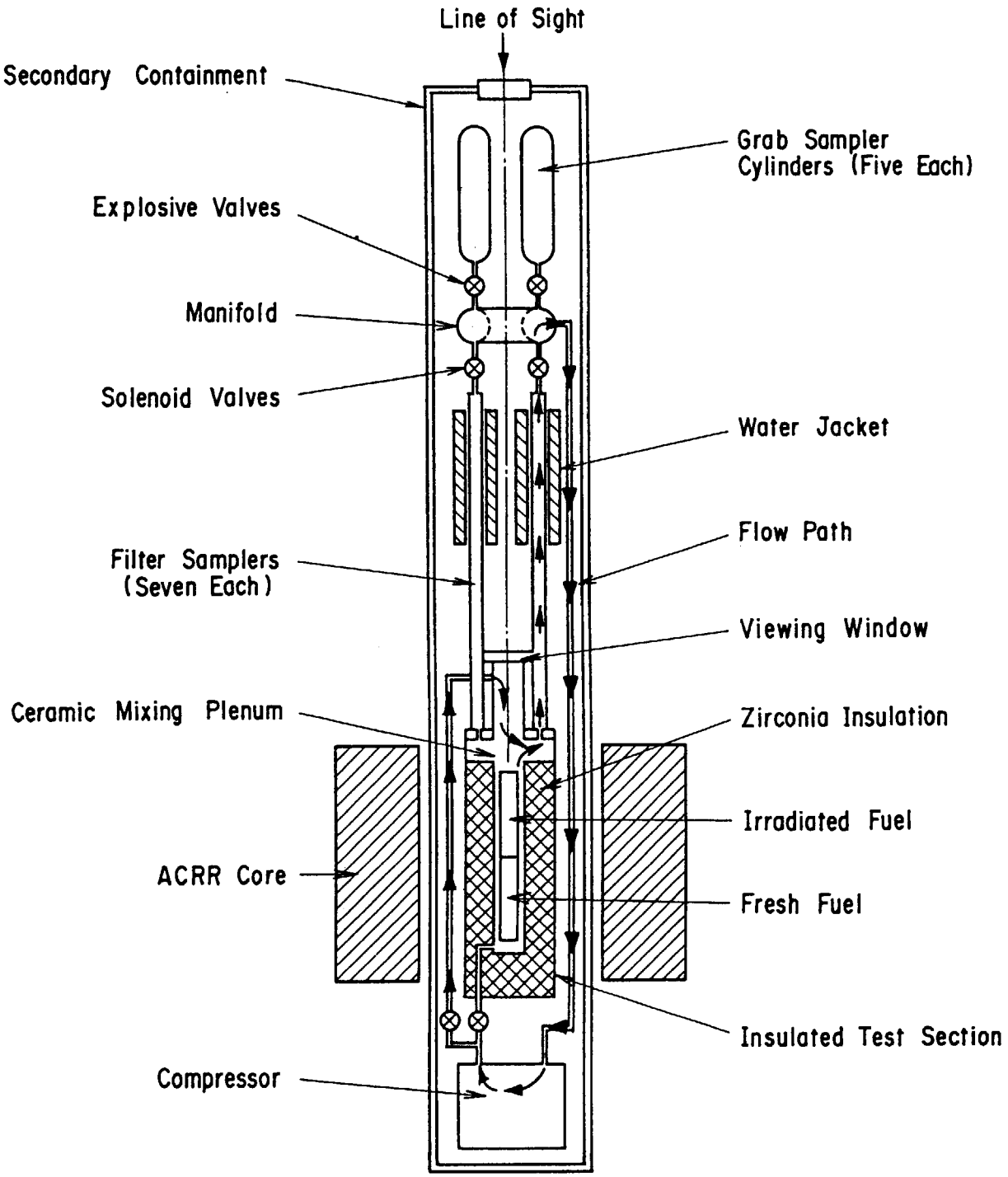

Fig. 2 Schematic configuration of SNL ST fission product release tests [8].

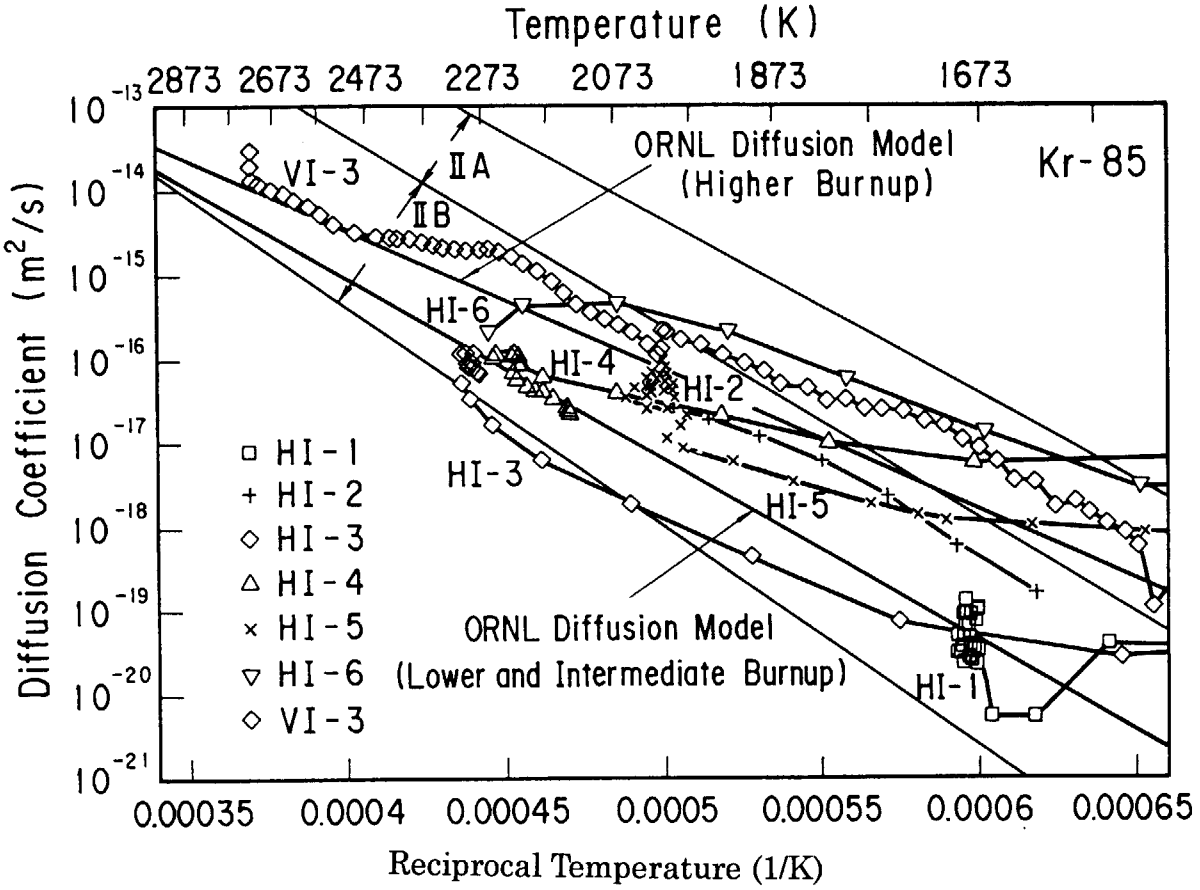

Fig. 3 Diffusion coefficient of Kr directly calculated from ORNL HI/VI test results.

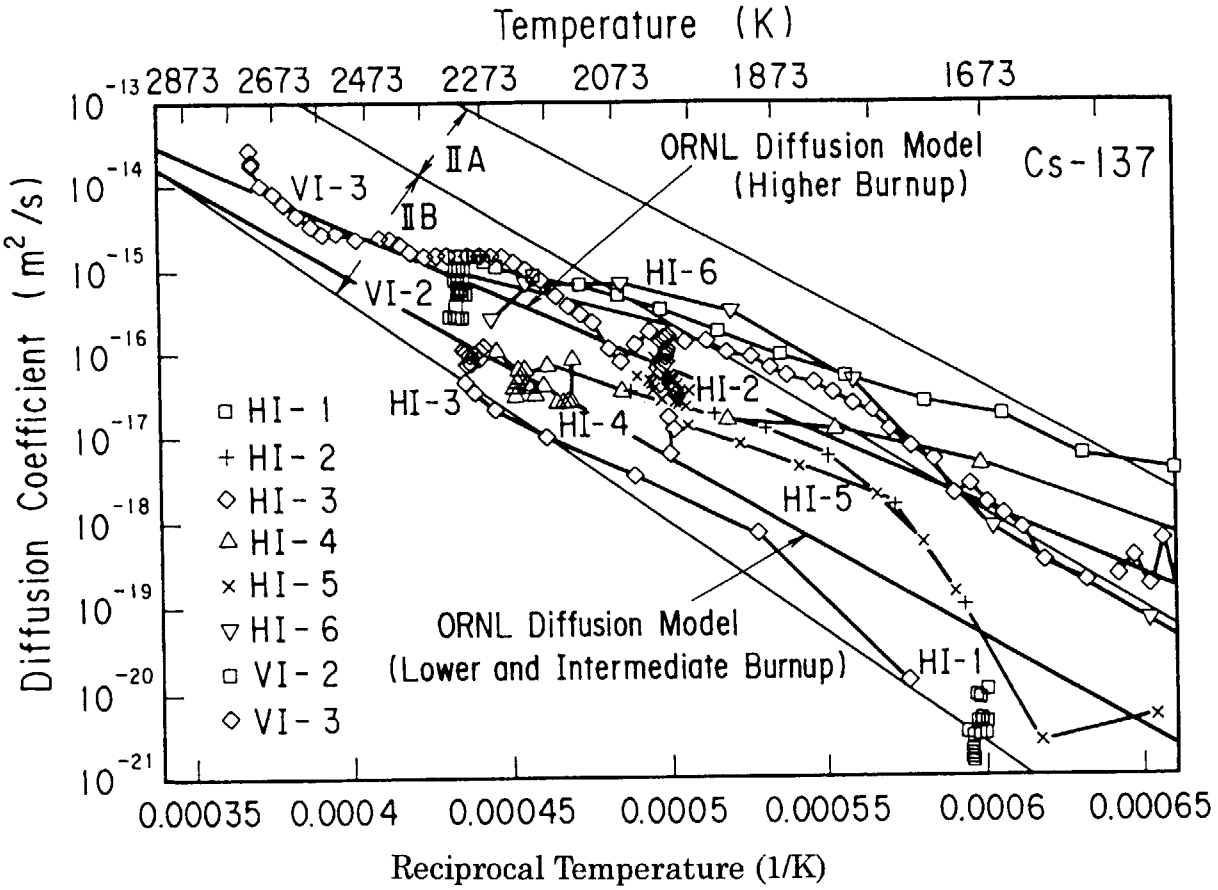

Fig. 4 Diffusion coefficient of Cs directly calculated from ORNL HI/VI test results, assuming Booth type diffusion controlling the release. Best fit diffusion coefficients for two burnup groups have been obtained as a function of temperature. The scatters of the test results indicate effects of test conditions other than temperature.

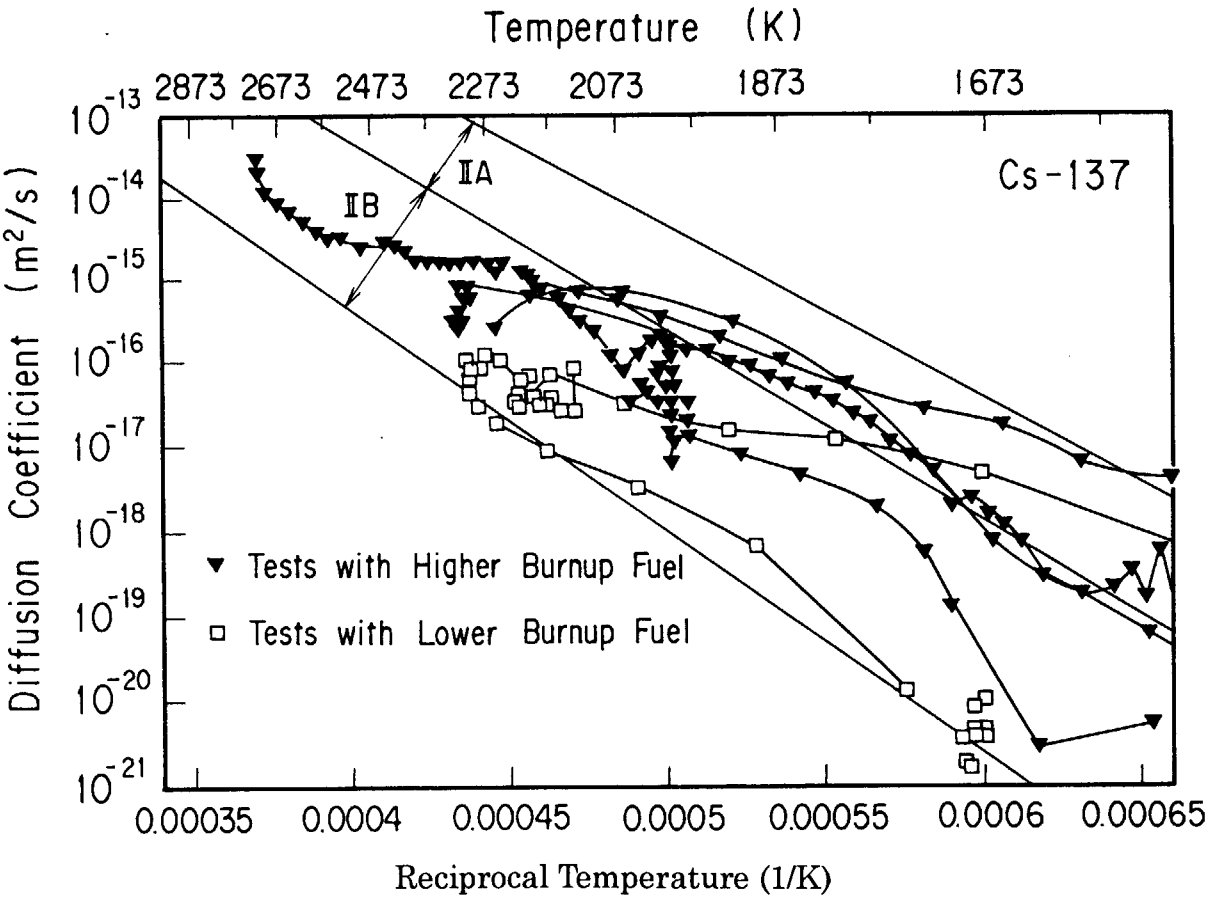

Fig. 5 Burnup dependence of diffusion coefficients of cesium.

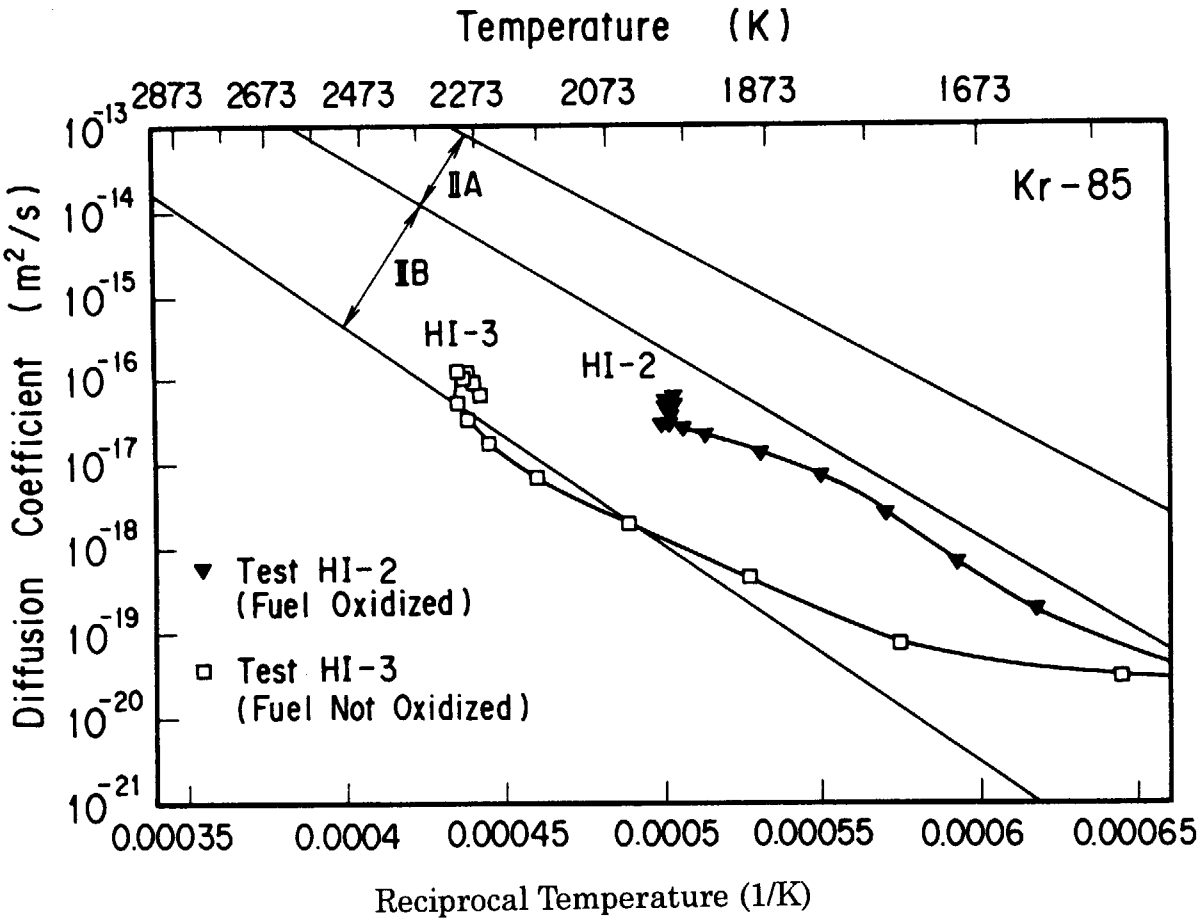

Fig. 6 Krypton release from fuel oxidized by steam in Test HI-2 and the release from un-oxidized fuel in Test HI-3.

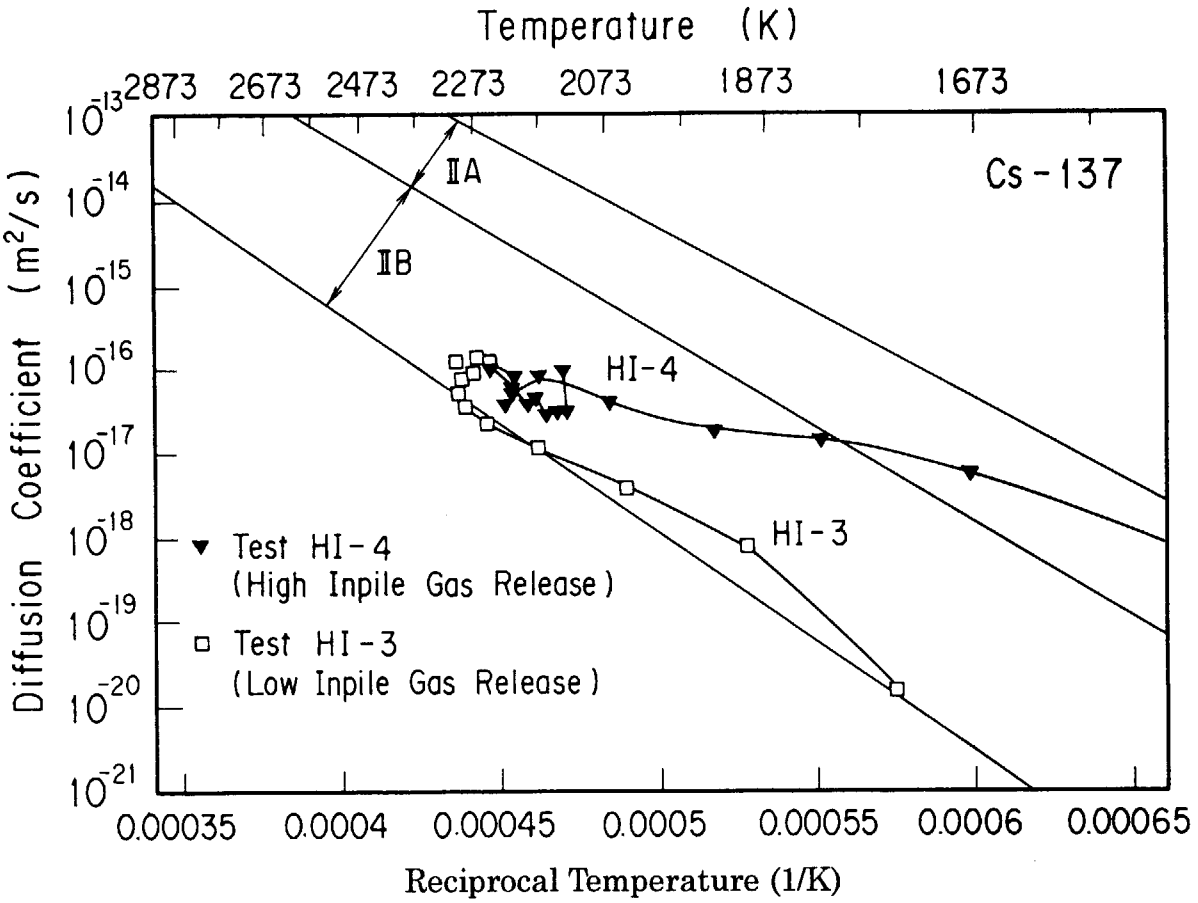

Fig. 7 Cesium releases from fuels with low and high inpile gas releases

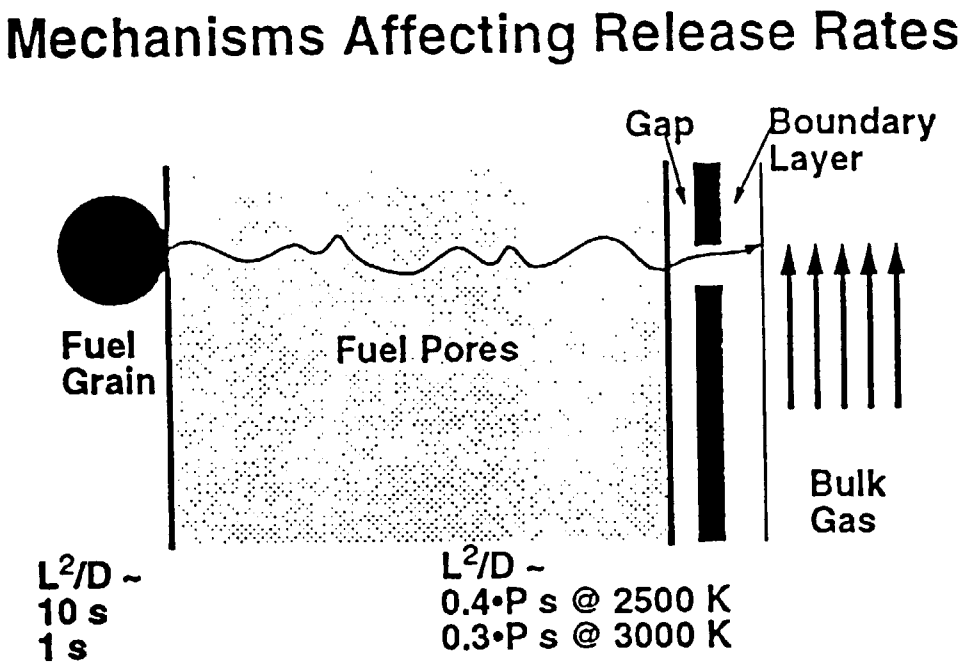

Fig. 8 Schematic of VICTORIA model for FP release process [25]

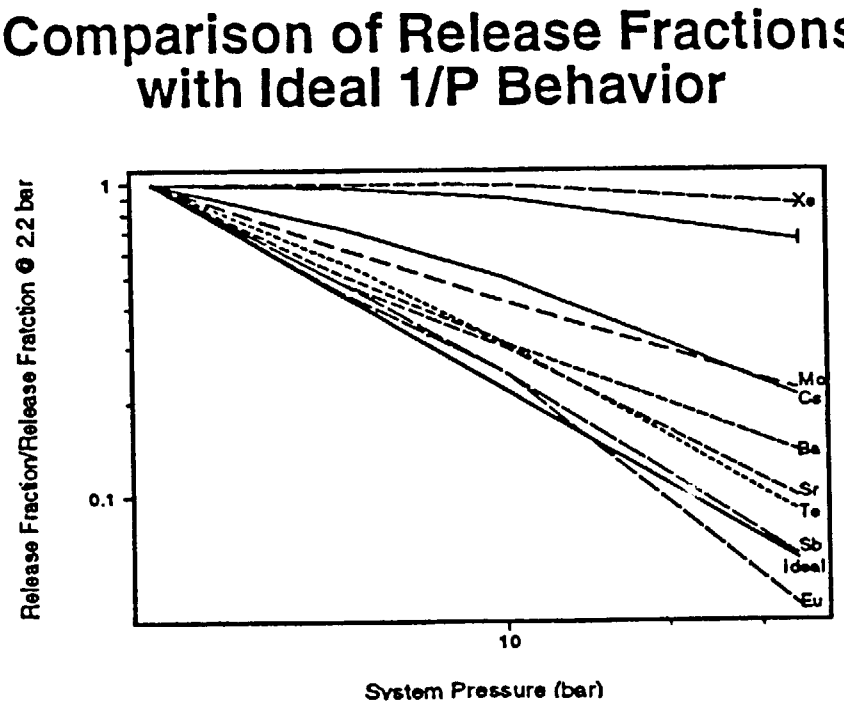

Fig. 9 Effect of pressure on FP release rate calculated by VICTORIA [27]

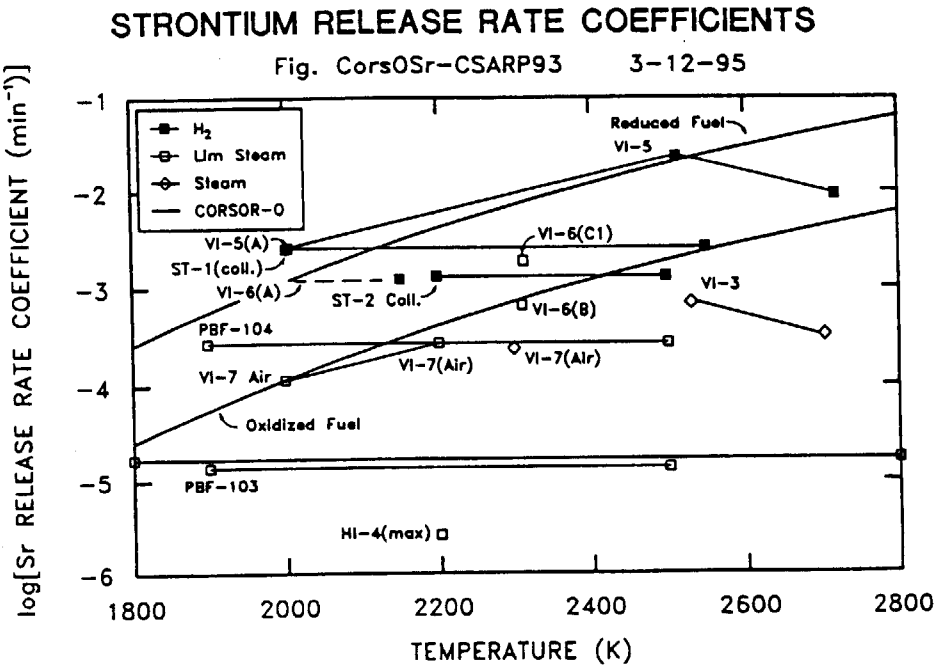

Fig. 10 Release rate coefficients for strontium [28]

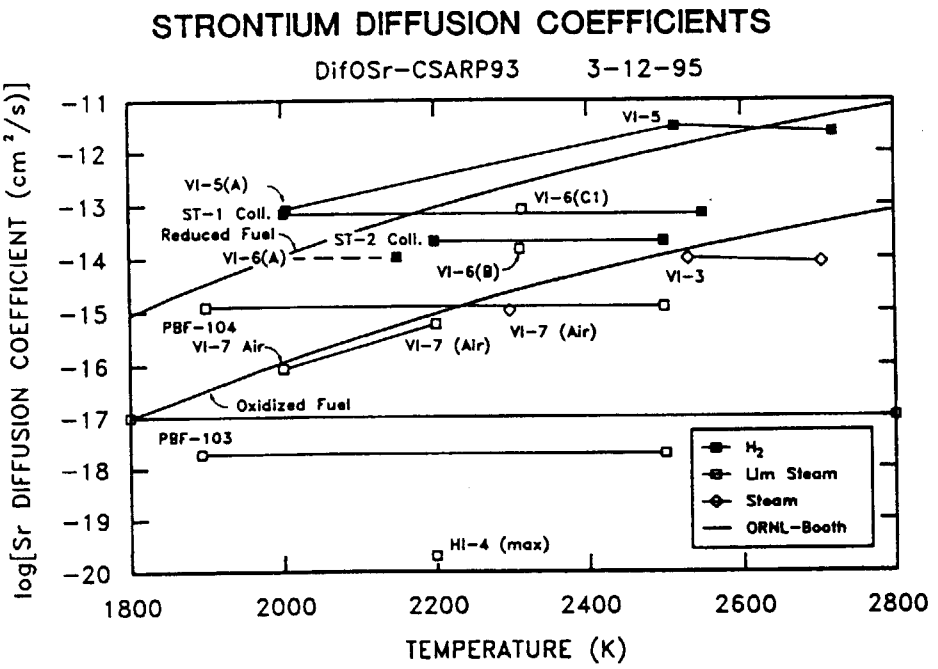

Fig. 11 Diffusion coefficients for strontium [28]

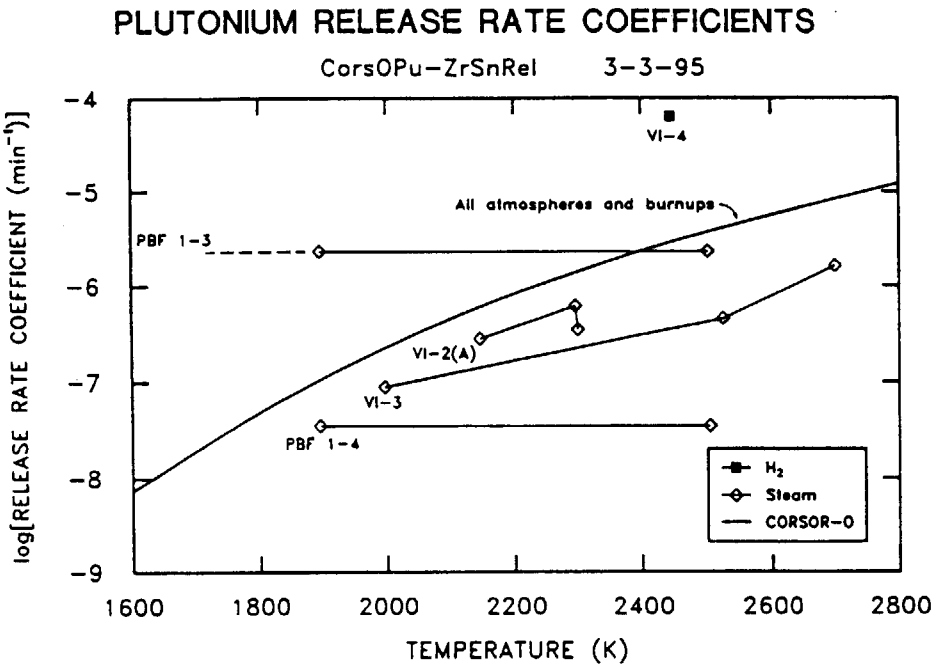

Fig. 12 Release rate coefficients for plutonium [28]

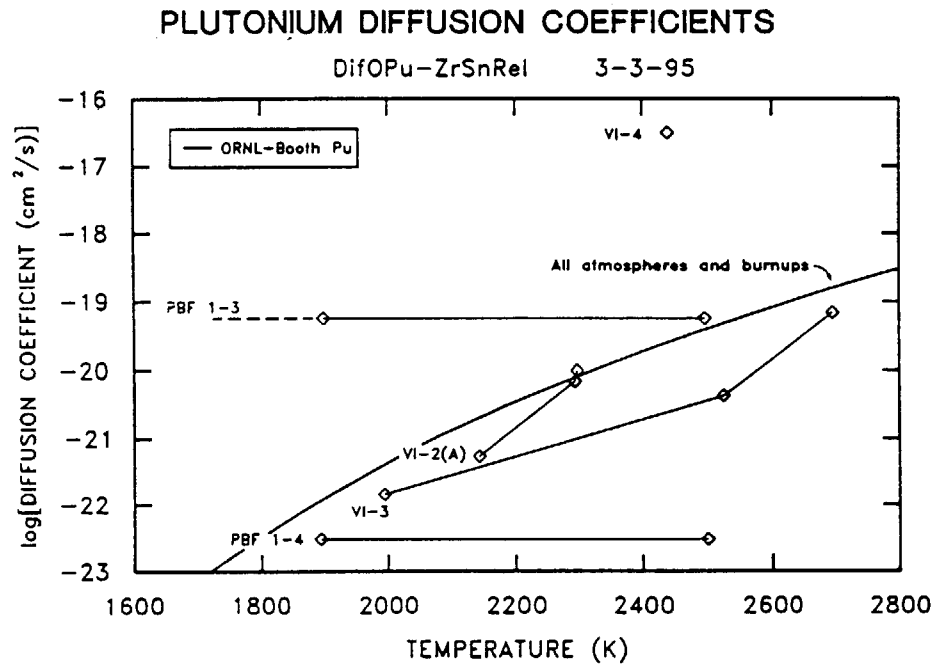

Fig. 13 Diffusion coefficients for plutonium [28]

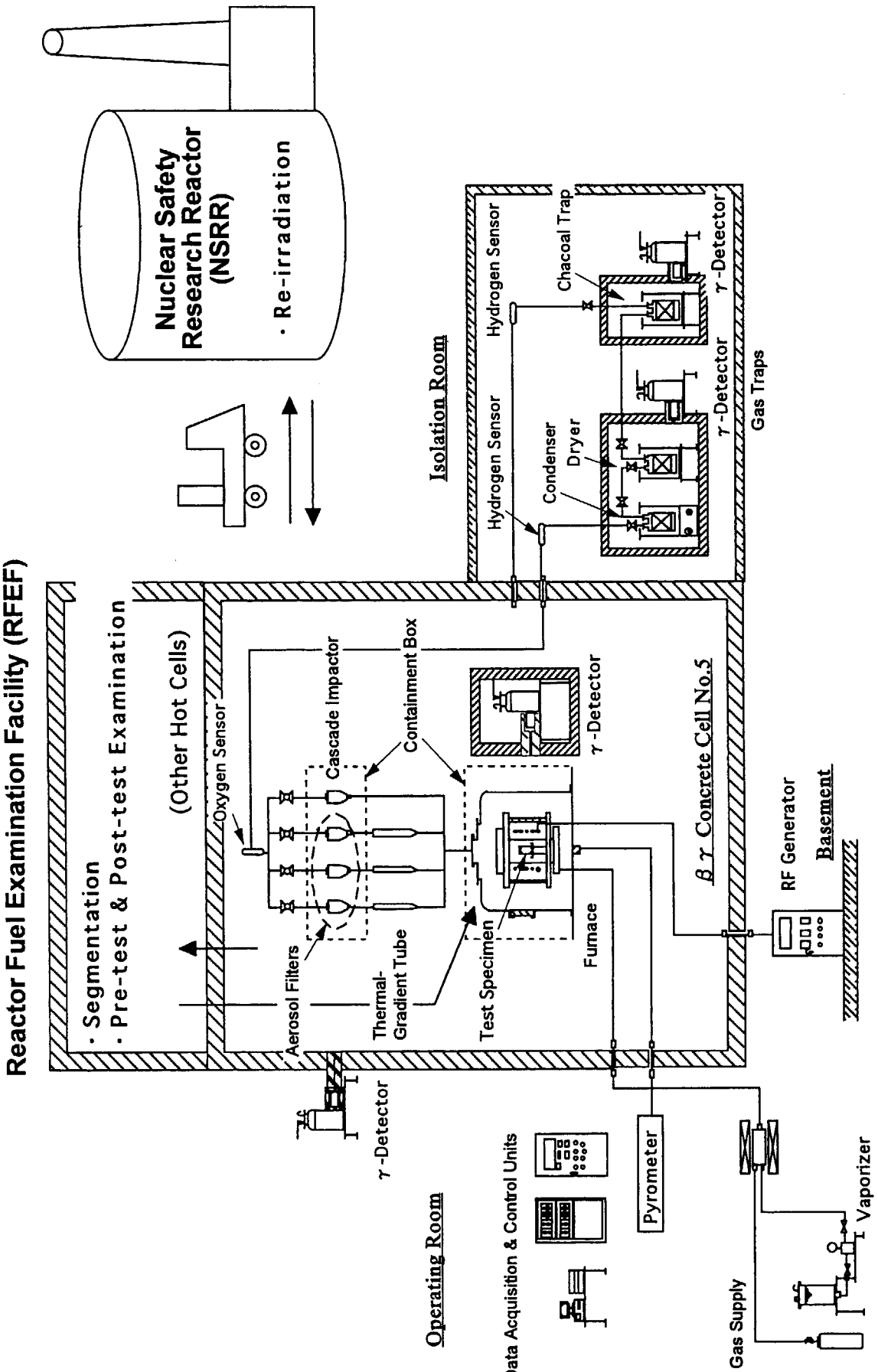

Fig. 14 Fission product release and transport experiment (VEGA)

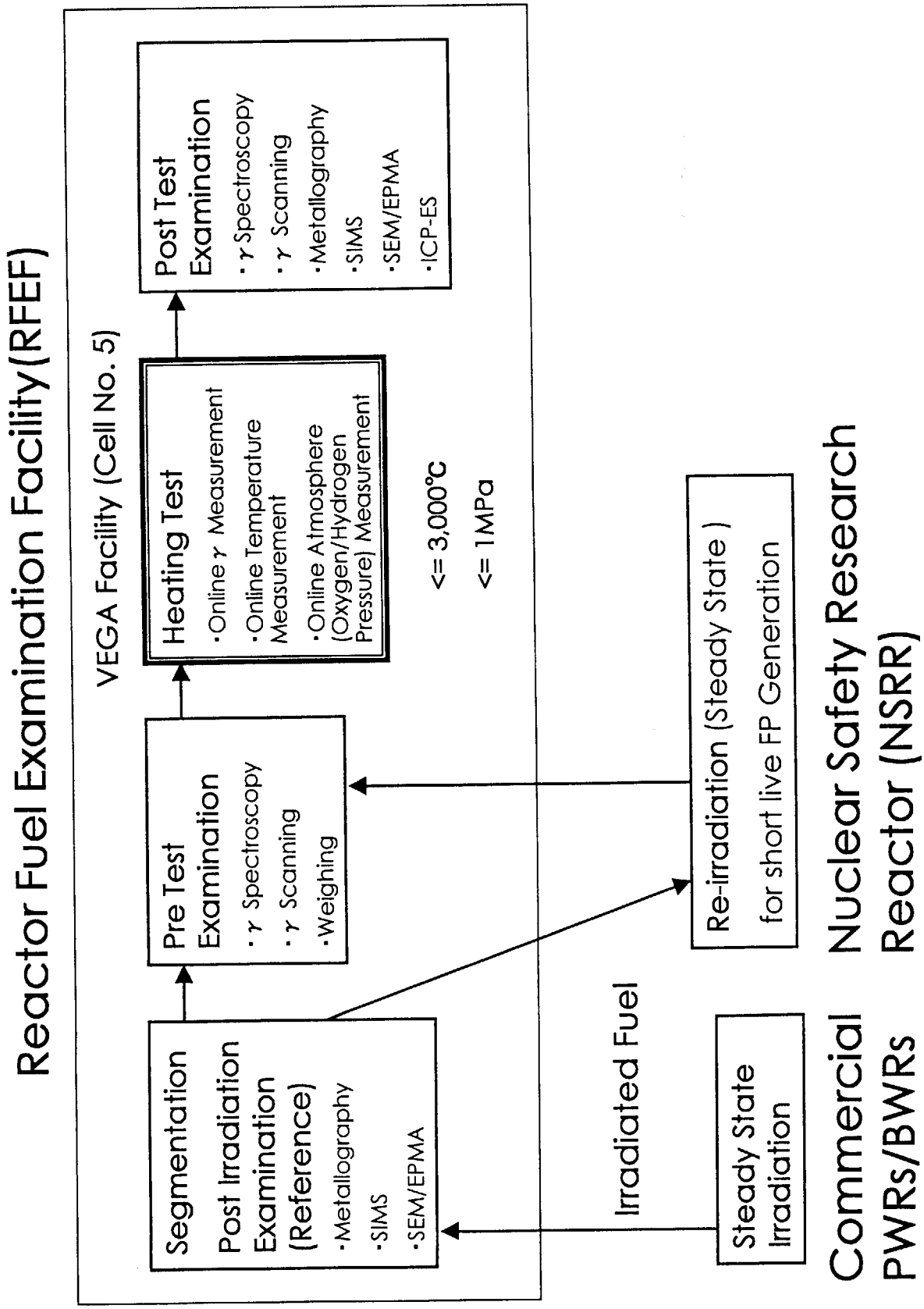

Fig. 15 Outline of test procedure of VEGA

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
長さ	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 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 eV = 1.60218 × 10^{-19} J

1 u = 1.66054 × 10^{-27} kg

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

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	$m \cdot kg/s^2$
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	ジーメンス	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	℃	
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 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
レントゲン	R
ラド	rad
レム	rem

1 Å = 0.1 nm = 10^{-10} m

1 b = 100 fm^2 = 10^{-28} m^2

1 bar = 0.1 MPa = 10^5 Pa

1 Gal = 1 cm/s^2 = 10^{-2} m/s^2

1 Ci = 3.7 × 10^{10} Bq

1 R = 2.58 × 10^{-4} C/kg

1 rad = 1 cGy = 10^{-2} Gy

1 rem = 1 cSv = 10^{-2} Sv

（注）

1. 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。
2. 表4には海里，ノット，アール，ヘクタ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。
3. 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。
4. 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

粘度　1 Pa·s(N·s/m^2) = 10 P(ポアズ)(g/(cm·s))

動粘度　1 m^2/s = 10^4 St(ストークス)(cm^2/s)

圧力

MPa(=10 bar)	kgf/cm^2	atm	mmHg(Torr)	lbf/in^2(psi)
1	10.1972	9.86923	7.50062 × 10^3	145.038
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 cal = 4.18605 J（計量法）

= 4.184 J （熱化学）

= 4.1855 J （15 °C）

= 4.1868 J（国際蒸気表）

仕事率　1 PS（仏馬力）

= 75 kgf·m/s

= 735.499 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

（86年12月26日現在）

RESEARCH PROGRAM (VEGA) ON THE FISSION PRODUCT RELEASE FROM IRRADIATED FUEL