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SECOND INTERNATIONAL COMPARISON ON MEASURING TECHNIQUES OF TRITIUM PRODUCTION RATE FOR FUSION NEUTRONICS EXPERIMENTS (ICMT-2)

Fabruary 1993

(Written and Editted by)
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Second International Comparison on Measuring Techniques of Tritium Production Rate for Fusion Neutronics Experiments (ICMT-2)

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An second international comparison on measuring techniques of tritium production rates for fusion neutronics experiments (ICMT-2) has been performed. The purpose is to evaluate the measurement accuracy of tritium production rates in the current measurement techniques. Two 14 MeV neutron source facilities, FNS at JAERI-Japan and LOTUS at EPFL-Switzerland, were used for this purpose. Nine groups out of seven

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countries participated in this program. A fusion simulated blanket assembly of simple-geometry was served as the test bed at each facility, in which Li-containing samples from the participants were irradiated in an uniform neutron field. The tritium production rates were determined by the participants using their own ways by using the liquid scintillation counting method. Tritiated water sample with unknown but the same concentration was also distributed and its concentration was measured to make a common reference. The standard deviation of measured tritium production rates among participants was about 10% for both FNS and LOTUS irradiation levels: 4×10^{-13} T-atoms/Li-atom and 1.6×10^{-12} T-atoms/Li-atom at a sample, respectively. This standard deviation exceeds the expected deviation of 5% in this program. It is presumed that the deviation of 10% is caused mainly by the systematic and unknown errors in a process of tritium extraction from the irradiated samples depending on each organization.

Keywords: NEA-NSC, International Comparison, Tritium Production Rate, Fusion Neutronics, FNS, LOTUS, Lithium, Tritium, Tritiated Water, Liquid Scintillation Counting

核融合中性子工学実験のためのトリチウム生成率測定法の国際比較 (ICMT－2)

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第2回核融合中性子工学実験のためのトリチウム生成率測定法の国際比較 (ICMT－2) が実施された。この目的は，現在の測定技術によるトリチウム生成率の測定精度を評価することである。この目的のため，原研FNSとローザンヌ工科大学 (スイス) のLOTUSの2つの14MeV中性子源施設が使われた。7ヶ国から9グループがこのプログラムに参加した。それぞれの施設では単純幾何形状からなる核融合模擬ブランケット体系が供試体として用意され，その体系中の均一中性子場で，各参加機関が持ち寄ったリチウムを含んだ試料が照射された。液体シンチレーション計数法を使った独自の方法で，各機関がトリチウム生成率を決定した。未知ではあるが同濃度のトリチウム水試料が配布され，その濃度を各機関が測定して共通の基準とされた。測定されたトリチウム生成率の機関内の標準偏差は，FNS照射レベル，LOTUS照射レベル (それぞれ試料位置で$4x10^{-13}$，$1.6x10^{-12}$トリチウム／リチウム原子) 共に約10％であった。この標準偏差の値はこのプログラムで期待されていた値である5％を越えている。10％のバラつきは，照射試料からのトリチウム抽出過程における各機関ごとの系統的で未知な誤差に原因があると推察された。

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1. Introduction

Tritium self-sufficiency is a key issue, from the view point of fuel cycle, in the development of fusion reactors based on D-T burning. Accordingly, tritium breeding capability has been estimated for various blanket design concepts in many countries. It turned out that it is not easy to keep high tritium breeding ratio (TBR) as designs direct to more realistic shape. When the TBR is marginal, the uncertainties in the data and methods used in the nuclear design calculation becomes of great concern. For example, an accuracy less than 5 % for TBR prediction is requested from designs to assure a good confidence in breeding performance.

Hence, experimental verifications are needed to confirm the accuracy of the nuclear data and calculation methods used in design studies through integral neutronic experiments for fusion blankets and the measurement methods adopted therein should be accurate enough. Up to now, a good number of blanket integral experiments have been reported from different research institutes, though still not many enough. A few to several percentage of accuracy has been typically claimed in tritium production rate determination. However, recovery of very small amount of tritium, from a fraction of Bq to hundreds Bq, out of the probe and normalizing it to an absolute value are not an easy matter. There are many factors affecting the final result and error assignment, and there might be unrecognized ones in some cases. For this reason, it is advisable to take an action, in an international cooperation, to validate the current status of the accuracy in determining tritium production rate objectively. This is especially important when one wants to use experimental results submitted by different experimentalists as a set of bench mark data to check the data and methods from different origins in a unified base.

The idea of the program, "International Comparison on Measuring Techniques of Tritium Production Rate for Fusion Neutronics Experiments" by using the 14 MeV neutron source, FNS, was first proposed from the JAERI representative at the 30th NEACRP meeting. The representative of Switzerland offered to use in addition the LOTUS facility to strengthen the program. It was advised at the meeting that a work plan be prepared by a cooperation of the experimental groups at JAERI, Japan and EPFL, Switzerland. The plan thus prepared through the exchange of opinions between FNS and LOTUS staffs was endorsed at the 31st NEACRP meeting, and participation in the program was solicited via the representatives of the committee for the research institutes in the member countries. Eight institutes from six countries have taken part in the comparison program. They are AECL/CRNL-Canada, ANL-U.S.A., CEN/Cadarache-France, ENEA/Casaccia-Italy, EPFL-Switzerland, Osaka Univ., the Univ. of Tokyo and JAERI-Japan, and are listed in **Table 1.1**.

The first irradiation experiment was carried out in 1989 using both FNS and LOTUS

facilities. Li-containing pellets prepared by each participants were irradiated in the simulated fusion blanket environment at both facilities. These pellets were sent back to the participants and tritium production rates were measured in the respective laboratories by their established procedure. At the same time, tritiated water of unknown but the same concentration, named as "blind sample", was distributed to the participants by JAERI and the concentration was also measured. Most of the results were summarized into a interim report and were presented at the 32nd NEACRP meeting.[a] Measured tritium concentration of the blind sample was well agreed within a few percentage among all the participants, while measured tritium production rate in a pellet was scattered about 10 % and a few tens of percentage for LOTUS and FNS irradiation, respectively. This result was out of the range of the expected accuracy, that is, 5 %.

At the 32nd meeting, the following two actions were endorsed.

Action 1: JAERI asks the items relevant to tritium counting techniques to each participant by questionnaire, and try to extract the reason of discrepancy.

Action 2: Each participant measures the concentration of diluted tritium water samples distributed by Argonne National Laboratory. The distributed samples had three types of different concentration, and this action would reveal the relation between the concentration of tritiated water and the accuracy of measurement.

The summary of the **Action 1** and **2** were presented at the 33rd NEACRP meeting.[b] Though the answers of questionnaire for the **Action 1** were examined in detail, no reasons of discrepancy for the measured tritium production rate were derived. From the results of the **Action 2**, good agreement was observed among the measured tritium concentration by each participants. In response to the results, the second irradiation experiment was proposed and endorsed at the meeting. In the spring of 1991 the second irradiation experiment was performed at the FNS and the LOTUS facilities with almost the same conditions as the first irradiation experiments. Nine organizations from seven countries were participated in the experiment; two organizations (ECN, MEPI) were newly joined in this program, while one of the members (ANL) had taken

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a) MAEKAWA H., NAKAMURA T. and HALDY P. A.: "International Comparison on Measuring Techniques of Tritium Production Rate for Fusion Neutronics Experiments (Interim Report)", NEACRP-A-1021, Session B.3.6 (Oct. 1989) ; JAERI-memo 01-442 (Dec. 1989).

b) H. MAEKAWA, "International Comparison on Measuring Techniques of Tritium Production Rate for Fusion Neutronics Experiments --- Summary of Additional Questionnaire and Result for ANL Samples ---", NEACRP-A-1080, Session B.3.6 (Oct. 1989); JAERI-memo 03-069 (March 1991).

part in this program so far was retired. The participants for the second experiment are listed in **Table 1.2**.

A brief review of the previous progress, the first experiment (**ICMT-1**) and results of ANL samples are presented in **Chapter 2**. Detailed explanations for the irradiation experiments at FNS/JAERI and LOTUS/EPFL are described in **Chapter 3** and **4**, respectively. **Chapter 4** is based on the manuscript prepared by Dr. D.-A.Haldy for **ICMT-1** Interim Report (See footnote b) in the previous page) and Dr. S. Azam for **ICMT-2**. Experimental procedure of tritium production-rate measurement by each participant is summarized in **Chapter 5** are based on documents prepared by the participants. Chapter 6 presents the results of second irradiation experiment (**ICMT-2**) and discussions. Concluding remarks are given in **Chapter 7**.

2. Review of the Previous Progress

The first irradiation experiments were carried out in 1989. Each participating group, listed in **Table 1.1**, prepared its own Li-containing samples. They were gathered at the neutron source facilities, FNS and LOTUS. The samples from the eight participants were irradiated so as to assure just identical neutron spectrum and fluence in a simulated fusion blanket assembly, at each facility respectively. The samples irradiated at both facilities were then sent back to the participants: sample processing and determination of tritium amount were carried out by their own ways based on the liquid scintillation counting method. At the same time, tritiated water of equal but blind concentration was distributed to each participant to provide a common reference in the comparison. Then the results have been collected at the coordinators, FNS and LOTUS. A comparative study on the results of eight different groups at two different neutron fluence and environments is able to represent the accuracy status of the techniques adopted.

It was intended that participants keep their own procedures, except for irradiations. Minimum conditions have been imposed from the coordinators on: selection in sample material, sample processing, counting procedure, data processing, corrections, error assignment and so on. This is to make the comparison as blind as possible at the first stage. After the first comparison of the results in overall performance was over, more detailed comparative study in each step was recommended so as to inter-compare the used technique of each group and assess possible causes of differences due to characteristic biases.

Six out of eight groups submitted the full set of results to JAERI by the middle of September, 1989. JAERI examined and summarized the results with the assistance of LOTUS group. At the 32nd NEACRP meeting, the summarized result was presented as an interim report (See footnote a) in the previous chapter). Some data were added and revised after the meeting. **Tables 2.1, 2.2** and **2.3** show the update of the measured tritium production rates, and **Table 2.4** shows that of the measured tritium concentration in the blind sample.

From the inter-comparison described in the summarized report, the following facts can be derived:

(1) For the irradiation level of LOTUS, typically several tens Bq of tritium per samples, agreement of tritium production rates from different methods turned out to be around 10 % for one standard deviation without any adjustment. The agreement for several Bq of tritium production in the FNS irradiation went worse to several tens %. This shows that the degree of agreement depends on the tritium amounts. The error assigned by the participants are not consistent with the observation.

(2) The interim result is against the requirement and expectation for a benchmark experiment, i.e., the measured data are deviated among the participants over the expected accuracy of 5 %. The observation suggests that there exist unidentified and organization-dependent systematic errors not directly related to counting statistics nor difference in tritium standards and it should reflect the dependence upon the procedures adopted at the organization..

(3) It is most important to identify the causes that bring forth the discrepancy by comparing and examining the whole procedures of the participants before mentioning on the true accuracy of the current measuring techniques. The examination includes the selection of Li-containing probe, physical and chemical processing of irradiated samples, preparation of liquid scintillation samples, counting of foreground and background, efficiency determination and calibration, tritium standard used, and error assignments on each factor.

According to the "Next Step" proposed to the 32nd NEACRP meeting, the following two "Actions" have been done.

Action 1

JAERI asks the items relevant to tritium counting techniques to each participant by a questionnaire, and examines and summarizes them.

Action 2

(1) Three types of diluted tritium water samples, i.e., around 10,000 Bq/g, several 10 Bq/g and several Bq/g, are made from the certified HTO standard with high accuracy, and are distributed to each participant.

(2) The participants measure the tritium concentration of the three samples and report it to JAERI. JAERI examines and summarizes the results reported.

After the 32nd NEACRP meeting, the **Actions 1** and **2** were started. For the Action 1, the following items were asked to each participant.

- How to process the irradiated samples chemically / physically ?
- How to make the samples for liquid scintillation counting ?
- Method of efficiency determination
- Specification of tritiated water standard
- Procedure of tritium counting

- Background samples for tritium counting
- How to determine the background level ?
- What type of correction had been done ?
- How to estimate the error ?

For the Action 2, three certified HTO standards of different concentration were distributed to the participants by ANL and concentrations of them were measured. The results of **Actions 1** and **2** were summarized into a report by JAERI and was presented at the 33rd NEACRP meeting, the summarized result was presented (See footnote b) in the previous chapter).

From the **Action 1**, no concrete reasons why tritium production rate reported for the first irradiation experiment was deviated so large were derived, though the answers from each participant were examined in detail. The measured tritium concentration of the blind sample for the **Action 2** is summarized in **Table 2.5**. The following conclusion was derived from the result.

(1) Using an appropriate tritium water standard with high accuracy for the calibration of liquid scintillation counting system, we can expect a good agreement for a blind sample among the participants. This is also supported by the result of JAERI's blind sample distributed at the first irradiation experiment (**ICMT-1**).

(2) All participants have the ability of measurement for the tritium concentration level of several Bq/g. Namely, we can measure the tritium production rate in a simulated fusion blanket under the irradiation level of 7×10^{15}. This total neutron yield can be obtained by the D-T neutron yield rate of 2×10^{11} n/s and 10 hours irradiation. This irradiation level is corresponding to the last experiment at FNS.

(3) It is important to obtain a liquid scintillation sample keeping the loss of tritium produced as low as possible without any contamination. Namely, the method and technique are essential for the chemical treatment of irradiated sample with lithium. If all participants adopt an appropriate method to extract the tritium produced from the irradiated sample, and make a good liquid scintillation sample, a good agreement is expected among the results of tritium production rate measured by the participants.

The results of the first irradiation experiment and the measurement of tritium concentration of the certified HTO standard suggest that the chemical form of Li-containing pellets and the method of chemical handling for the irradiated samples are essential. At the

33rd NEACRP meeting, the second International Comparison on Measuring Techniques of Tritium Production Rate for Fusion Neutronics Experiments (**ICMT-2**) was proposed from the JAERI representative and endorsed. The irradiation condition for the second irradiation experiment was almost the same as the first experiment conducted at both FNS/JAERI and LOTUS/EPFL, but the technics of tritium production rate measurement were reviewed and modified by each participant, if necessary.

3. Irradiation Experiments at FNS

3.1 Experimental Assembly

The experimental assembly for the sample irradiation was just the same as that used in **ICMT-1** at Fusion Neutronics Source (FNS) facility[1]. The experimental arrangement and configuration similar to the previous blanket benchmark experiment[2] are shown in **Figs. 3.1** and **3.2.** Area-equivalent diameter and thickness of assembly were 630 and 602.4 mm, respectively. Two types of lithium-oxide blocks were stacked in a framework made of thin-wall aluminum tubes to form a cylindrical assembly. The outside dimensions of the blocks had an area of 50.6 mm x 50.6 mm and lengths of 101.2 and 202.4 mm, respectively. Bricks, which were almost cubic, were made from natural Li_2O powder by cold pressing. Density of Li_2O bricks was 75.5 % of theoretical density. Two and four Li_2O bricks, respectively, were encapsulated to make up a block in a 0.2 mm thick Type 304 stainless steel box, edge-welded with a 2.5 mm deep rim to prevent the moisture from atmosphere.

A cavity of 20 mm x 253 mm x 253 mm was made in the assembly to set a rotating disk. Distance between the surface and cavity was 98.8 mm. To keep this cavity a stainless steel spacer of 20 mm-wide and 1 mm-thick was set in it. An experimental channel, i.e., a set of sheath and drawer made of 0.2 mm-thick stainless steel, was placed at the backside of the cavity along the central axis of the assembly. Special-sized Li_2O blocks with 6 mm x 6 mm hole, which were a little smaller than normal ones, were loaded in this drawer in order to allow inserting a driving shaft for the rotating disk.

3.2 Neutron Source

The 80° beam line in the first target room of Fusion Neutronics Source (FNS)[1] was used for this sample irradiation experiment. The neutron yield rate of 80° beam line is one order lower than that of 0° beam line in the second target room but the 80° beam line was selected. Because the effect of room-returned neutrons in the first target room was remarkably smaller than that in the second target room. A high speed water cooled target assembly was set at the end of the beam line. A 370 GBq (10 Ci) Ti-T target was mounted on the target assembly. Neutrons were generated at the distance of 100 mm from the Li_2O assembly surface on its central axis. The setting accuracy was estimated to be within ± 1 mm. The layout of the first target room (15 m x 15 m) is shown in **Fig. 3.3.** The distances from the target to west and south walls are 5.5 m, and those to the ceiling, the grating floor and the basement floor

are 7.9, 1.8 and 4.8 m, respectively. Neutron yield was determined by means of the associated α-particle detection method.[3] A small silicon surface-barrier detector was mounted in the beam duct to detect the α-particle of $^{3}T(d,n)^{4}He$ reaction.

In order to obtain a uniform irradiation condition for all samples supplied from each participant, a rotating disk was adopted in this experiment. **Figure 3.4** shows the rotating disk made of 1.6 mm-thick aluminum. It has 45 holes for placing the samples. The centers of 20 and 25 holes were on the same concentric circles of 140 and 210 mm in diameter, respectively. The disk was fixed on one end of the driving shaft. The shaft was inserted in the small hole of Li_2O blocks, and the rotating disk was located into the cavity in the Li_2O assembly mentioned above. A synchronous motor of 5 rpm was mounted on the other end of the shaft. The layout of arrangement is shown in **Fig. 3.2**.

3.3 Sample Specification

The sample specifications used in the second irradiation experiment are summarized in **Table 3.1**. The chemical forms of samples were lithium-zirconate for ECN, lithium-oxide for JAERI and lithium-carbonate for the other participants. Most of samples were covered with aluminum foil or case for sealing. The sample size including sealing material was from 10 mm to 26 mm in diameter and from 2 mm to 6 mm in thickness.

The mass analysis for the pellets sent by the participants was performed to confirm that the atomic ratios of lithium-6 and lithium-7 were equal to the natural abundance. The mass analysis was made by Mr. and Mrs. Tamura of Analytical Chemistry Laboratory, JAERI. The results are shown also in **Table 3.1**. Unfortunately the lithium-6 ratio to all lithium atoms of ENEA (5.48 %) was not the natural abundance, so simple comparisons of tritium production rates between ENEA and the other organization could not be made. The lithium-6 ratio for all organizations except ENEA are looked on as the natural abundance, though a few scatter of the ratio, from 7.44 to 7.66, is observed. It was found from the calculated result that the effect of this scatter is less than 0.5 %, and the effect could be neglected against the expected accuracy of 5 % for this program.

3.4 Irradiation Procedure

All samples for the FNS irradiation had been kept in a desiccator since they arrived at JAERI from the participants. One day before the irradiation (Apr. 25, 1991), the samples to

be irradiated were set in the holes of rotating disk by Scotch tape shown in **Fig. 3.4**. Since special attention was paid for positioning, each sample was located at the center of each hole within at least 1 mm for radial direction and 0.5 mm for the perpendicular direction. The rotating disk with samples was sealed in a vinyl bag and kept in the desiccator until just before the irradiation.

ECN sent us ten irradiation samples (five Li_8ZrO_6 and five $Li_6Zr_2O_7$ samples) while number of holes assigned for ECN samples were five. Three Li_8ZrO_6 samples and two $Li_6Zr_2O_7$ samples were located at the outer and inner band holes of rotating disk, respectively. The rest of samples (two Li_8ZrO_6 samples and three $Li_6Zr_2O_7$ samples) were fixed by scotch tape on the space of rotating disk between the inner and outer band holes, i.e., on the circle of 178 mm in diameter. Since the later five samples were irradiated just the same condition, it was possible to investigate the effect caused by different chemical forms of samples.

On Apr. 26, 1991 the disk with samples was mounted on the end of driving shaft and the distance between the disk and the backside of the cavity was measured. After that, twenty-five Li_2O blocks were loaded in front of the cavity. The average distance between the front surface of cavity and the center of disk was estimated to be 8.4 mm.

The irradiation was performed from 10:01 to 21:01 except for an interruption for five minutes by a trouble of the accelerator. The beam conditions and total neutron yield at the target during the irradiation are as follows:

Energy of d^+ beam	: 350 keV
Beam current	: 1.7 ~ 2.0 mA
Beam spot size	: within 15 mm diam.
Total neutron yield	: 9.45×10^{15} neutrons (1.9 %)

The disk with the samples was removed from the experimental assembly and sealed in new double vinyl bags. On May 28, 1991 all samples were dismounted from the disk and sealed in a can. On May 30, 1991 the samples were sent back to the participants together with a blind sample.

3.5 Blind Sample

A sample of common tritium concentration is very useful for normalizing the values measured by tritium counting systems used in different organizations. For this purpose JAERI provided a blind sample and distributed to all participants. About 22 ml of water

containing tritium was sealed in a polyethylene vial. The vial was sealed in some sheets of nylon bags. It was sealed in a can with irradiated samples and sent to the participants.

3.6 Analysis

An experimental analysis was performed after the second irradiation experiment by the two-dimensional transport code DOT3.5[4]. The calculation model is shown in **Fig. 3.5**. **Table 3.2** shows the homogenized nuclide density used in the calculation for each region. The GRTUNCL code was used to calculate the first collision source in order to eliminate the "ray effect". The number of Legendre polynomials was 5; and the number of angular quadratures was 16. The 125-group cross section set FUSION-J3[5] was used in the calculation. This set based on the JENDL-3 nuclear data file.

Calculated tritium production rate for natural lithium (^{6}Li : 7.56 atom %) are as follows:

outer position : 4.292×10^{-29} [T-atoms/ Li-atom/ source neutrons]
inner position : 4.862×10^{-29} [T-atoms/ Li-atom/ source neutrons]

Calculated tritium production rate distributions are shown in **Figs. 3.6** and **3.7**.

4. Irradiation Experiment at LOTUS

4.1 Experimental Assembly

The LOTUS[6] facility is an experimental facility situated in one of the two experimental bays of the IGA (Institut de Génie Atomique) at Ecole Polytechnique Federale de Lausanne (EPFL). Its experimental cavity is surrounded with a massive concrete shielding having a thickness of roughly 2 meters. The inner dimensions of the cavity are 2.4 m x 3.6 m and 3.0 m high. Blankets and experimental equipments was loaded in the cavity by opening the top cover or through a small lateral door. **Figure 4.1** shows how components were placed in the cavity for the tritium breeding benchmark experiment. The neutron generator was positioned in such a way that the neutron producing target would be placed at the center of the cavity. This was easily done because the neutron tube is mounted on a trolley which could be moved back and forth along the axis of the cavity.

The lithium blanket module was mainly composed of two different zones. The first one is a natural lithium zone for tritium breeding. It was followed by the second zone made of graphite, used as a reflector. Lithium pellets and graphite pencils were contained in stainless steel tubes. The lithium pellets occupied the first 60 cm and graphite the last 20 cm. In the irradiation experiment, around 700 tubes were arranged in such a way to realize an almost cylindrical geometry. The lithium module was facing the neutron tube, on the same axis. The distance between the center of the target and the lithium module front face was fixed at 20 cm. The samples were arranged on a disk, rotating at a constant speed (1 rpm) by means of a synchronous motor, situated 10 cm inside the blanket module. The gap needed to place the disk was 1 cm-wide. Zirconium foils on the x axis were used to monitor the 14 MeV neutron fluence, for normalization with previous works done on the same module.

4.2 Neutron Generator

The Haefely Neutron Generator (HNG), which produces a high intensity flux of 14 MeV neutron, has been developed at the Kernforschungszentrum Karlsruhe (Germany), and was manufactured and commercialized by the firm Haefely located in Basel (Switzerland). The outstanding feature of this generator is the high neutron source strength available in a very compact design. The device in which the neutrons are produced is a sealed tube, thus avoiding tritium contamination, based on the penning ion source type. The whole device has a cylindrical geometry, the target being placed on the x axis shown **Fig. 4.2**. The target is

filled with 18.5 TBq (500 Ci) of tritium and an equivalent amount of deuterium absorbed in a thin scandium film deposited on a copper support. A 400 mA mixed beam of deuterium and tritium ions is accelerated toward the target, with an energy approaching up to 180 keV. Thus neutrons are produced from DT and DD reactions. Taking into account the respective cross-sections, the 14 MeV neutron production represents more than 99 % of the total neutron output. The intensity can be levelled up to 5 x 10^{12} n/s and is directly controlled through the acceleration voltage, the ion beam current, and the ring electrode voltage. The tube is water cooled. The HNG works only in continuous mode and is able to deliver neutrons at a given intensity for many hours even at the highest intensity. More than 70 % of the total neutron output is directed towards the front face of the tube and is available for blanket irradiation. The tube is located in the LOTUS cavity, while the power supply, cooling system, control circuits and other equipments are placed outside the shielding. Running the generator is fully automatized.

4.3 Irradiation Procedure

The essential aim of the benchmark experiment was to irradiate each sample in identical flux conditions during the whole irradiation time. Nine different organizations participated in the experiment, most of them having supplied their own sets of samples (The Netherlands Energy Research Foundation supplied two sets). Each set having 5 samples for irradiation and a few for background measurements. There were 9 different types of samples (as far as dimensions, chemical composition and sealing material are concerned). In addition, the irradiation had to be independent on any eventual asymmetry in neutron flux: due to the neutron generator, the blanket, or the experimental cavity. Taking this into account and referring to the previous JAERI set-up conditions for the first experiment, we placed the samples on a sample-holder. The sample-holder, similar to that of FNS, is a disk of 250 mm diameter and 2 mm thick, made of aluminum. It was installed inside the lithium module at 10 cm from the front face and on the axis. The disk was given a slow rotating motion (1 rpm) by a synchronous motor connected to a grid. The current supplied to the motor was measured during the whole irradiation to ensure a proper run.

The irradiation experiment started on the 24th of May 1991 at 13 h 35 (1.35 p.m.), as scheduled. Once the ion beams had been switched on, it took 5 minutes to reach the nominal neutron output. The nominal intensity was fixed at 3.5 x 10^{12} n/s. The generator then run at a steady state mode for 360 minutes. Only three high voltage breakdowns were recorded during the whole irradiation time, given a total breakdown time smaller than 60 seconds. This

implies that the generator worked remarkably well during all the experiment. One Zirconium foil was fixed on the center of the rotating disk. The fluence for the irradiation measured at this point has given a value of 2.5 $x10^{12}$ n/cm^2.

5. Measurement of Tritium Production Rate

Measurement methods of tritium production rate adopted by each participants are summarized in **Table 5.1**. This table is based on the questionnaire answered by each participant. Because answers of questionnaire are available only for AECL/CRL, CEA/Cadarache, ENEA, MEPI, Osaka Univ. and JAERI, and not for the other organizations, this Table is partly incomplete.

All the organizations used liquid scintillation counting method. To extract tritium from irradiated pellets, so called Dierckx's method[7] and cryo-distillation method are generally used. The AECL/CRL and ECN groups applied simple dissolution method. More detailed explanations about measurement methods for each participants are as follows:

5.1 AECL/CRL

Li_2CO_3 pellets were removed from aluminum cladding and then dissolved in 10 ml of dilute HCl. 2 ml of solution was pipetted into a 20 ml glass vial and 10 ml of scintillation cocktail, "Beckman Ready-Safe", was added. The errors of tritium production rate were estimated as sum of counting errors plus weighing errors from sample weight, aliquots for counting and the weight of HTO standard spike. Counting errors have been calculated at the 2σ -level.

5.2 CEA/Cadarache

Tow different methods were adopted. One was hydrolysis method for all the FNS samples and one LOTUS sample which is one of three outer samples. The another was the Dierckx's method for four LOTUS samples except for the one outer sample. Both methods are described in **Sections 5.2.1** and **5.2.2**, respectively.

5.2.1 Hydrolysis Method

a) Chemical treatment

a-1) Basic principle

A flow of the procedure is shown in **Fig. 5.1**. In the course of the hydrolysis of a

tritiated sample, the tritium contained in the sample is distributed between the aqueous phase and the gas phase including hydrogen and tritium. A specially designed tight circuit and constant argon sweeping flow allows us to drag the hydrogen and tritium forward. This mixture enters a catalytic oxidation furnace in order to transform it into water vapor. The tritiated water vapor is recovered in a cold trap at the outlet of the furnace. A bubbler is placed just after the cold trap with purpose of capturing any tritiated water that might possibly escape from the cold trap. Tritium dosage is carried out on each phase (aqueous phase, cold trap and bubbler phases) through liquid scintillation.

a-2) Equipment

The material used for the hydrolysis is standard laboratory equipment made of glass. It is decontaminated and checked after each use. The water used is permuted water.

b) Counting through Scintillation

b-1) Preparation of the Counting Solutions

The counting is carried out on 20 ml solutions made up as follows : 5 ml of solution to be dosed, and 15 ml of cocktail liquid scintillation. Three counting solutions were prepared for the distillate, three for the cold trap and three for the bubbler.

c) Calculations

c-1) Calculation of the Specific Activity

The maximum counting duration allows by the apparatus being 100 minutes, a series of 4 x 3 countings is performed. The results were then expressed in counts per minute (cpm) for each distillate, trap, bubble and blank (See **Table 5.2**).

(i) Calculation of the Efficiency (E_j)

A standard source is used for the measurement of the efficiency. The count rates are about 100 times greater than those obtained for the distillates sub-samples (100000 compared to 100 c/min). E is given in percent = 35.0 %.

(ii) Activity of the Blank (B_j)

The average value over the 3 x (4 countings over 100 minutes) is taken. A new measurement is performed for each sample. The values are around 30 c/min. The blank did not differ sufficiently from the background level; therefore it was used as a back ground itself.

(iii) Activity of the Distillate (d_j)

The average value of the 4 x 3 countings over 100 minutes is taken, then the average activity is calculated. The blank is then subtracted and the obtained value was corrected for the ratio of volumes and the efficiency.

(iv) Activity of the Trap (t_j)

The average value of the 4 x 3 countings over 100 minutes is taken, then the average activity is calculated. The blank is then subtracted and the obtained value is corrected for the ratio of volumes, the efficiency, and the furnace yield (0.85).

(v) Activity of the Bubbler (b_j)

The average value of the 4 x 3 countings over 100 minutes is taken, then the average activity is calculated. The blank is then subtracted and the obtained value is corrected for the ratio of volumes, the efficiency, and the furnace yield (0.85).

(vi) Activity of the Sample (a_j)

It is calculated as: $a_j = d_j + t_j + b_j$

(vii) Specific Activity of the Sample (A_j).

It is calculated as: $A_j = a_j / m_j$, where m_j is the mass of the lithium measured by flame atomic absorption spectrophotometry.

c-2) Estimation of the Errors on the Specific Activity

(i) Error due to the Countings ($\Delta C/C$)

The fact that the counting of the same solution is repeated 3 x 4 times during 100 min allows us to estimate a counting standard deviation (σ_c). The error due to the countings within a confidence limit of about 90%, is calculated to be $\Delta C/C$.

(ii) Error due to the Efficiency ($\Delta E/E$) and Furnace Yield Measurements ($\Delta Y/Y$)

The error due to the efficiency is calculated $\Delta E/E = 2.4$ %.

The relative error on the furnace yield was 6%. Its influence on the total activity is then:

1.5 % $< \Delta y/y <$ 2.3% according to samples.

(iii) Error due to the Volume ($\Delta V/V$) and Mass Measurements ($\Delta M/M$)

The relative errors due to the volumes (2 by sample) and mass measurements are calculated:

$$4\frac{\Delta V}{V} = 4\left(\frac{5}{1000}\right)$$

(iv) Total Error ($\Delta A/A$):

$$(\Delta A/A)^2 = (\Delta C/C)^2 + (\Delta E/E)^2 + (\Delta Y/Y)^2 + 4(\Delta V/V)^2$$

($\Delta A/A$ with a confidence limit of about 90%).

5.2.2 Dierckx's method

a) Chemical Treatment

The irradiated Li_2CO_3 pellet was placed in a glass vial; 1 cm^3 of permuted water and 2 cm^3 of CH_3COOH (100%) were added in the vial. One day after, 0.9 cm^3 of hydrofluoric acid (40%), then 10 cm^3 of liquid scintillation cocktail (Instagel) were added in it. The vial was closed and centrifuged; the LiF was precipitated at the bottom of the vial. Tritium count in each sample was measured through liquid scintillation. The counting time for each sample was 40 minutes. Two counting cycles were carried out; Each counting was repeated four times. The results were expressed in counts per minute (cpm).

b) Calculation to the Efficiency

To determine the efficiency E, a sample was made using unirradiated Li_2CO_3 (0.7 g); 1 cm^3 of tritiated water standard and 2 cm^3 of CH_3COOH were added; then, the same procedure mentioned before. Two counting cycles were carried out; each counting was repeated four times. The efficiency was determined and given in percent. The background level was determined using the same procedure as that for the irradiated samples except using unirradiated Li_2CO_3.

c) Estimation of the Errors on the Specific Activity

(i) Error due to the countings $\Delta C/C$

The counting of the same sample was repeated 8 times during 40 minutes. A counting standard deviation within a confidence limit of 90% was estimated as follows:

$\Delta C/C = 0.3 \sim 0.7$ %

(ii) Error due to the efficiency ($\Delta E/E$):

$\Delta E/E = 2.0$ %

(iii) Error due to the mass and volume measurements:

$\Delta M/M = 2$ %

(iv) Total Error $\Delta A/A$:

$$(\Delta A/A)^2 = (\Delta C/C)^2 + (\Delta E/E)^2 + (\Delta M/M)^2$$

($\Delta A/A$ with a confidence limit of 90 %).

5.3 ECN

After having received the samples, irradiated in both the FNS and LOTUS facility, determination of the amount of tritium generated in the samples and the tritium concentration in the blind sample water was started. The tritium concentration in the latter was found to be:

0.803 ± 0.009 nCi/g or 29.711 Bq/g $\pm$ 1.1 %.

The measurements were carried out at June 20 and 21, 1991. The decay corrected tritium concentration is therefore:

$29.711 \times 1.0085 = 29.964$ Bq/g.

The measurements were continued with the irradiated samples. Each pellet was divided into four parts, which were subsequently dissolved in 32 % HCl except one part which was kept in reserve. Considerable scattering was observed in the results of the measurements of the three part of each pellet. This is most likely caused by "foreign" beta activity. However, so far, this foreign beta activity was not identified and the contribution of this activity to the tritium activity could not be determined.

After dissolution the samples and measuring the tritium concentration by liquid scintillation, part of the solution was cryo-distilled. This distillate was again measured by liquid scintillation. The residue was solved and also measured. In most cases a significant difference was found between the tritium concentration in the distillate and in the residue, with in some cases a much higher concentration in the residue and in some cases almost equal concentrations. In only a few cases the tritium concentration in the distillate was higher compared to that in the residue.

In **Table 5.3**, an example is given of the range of measured tritium concentrations in nCi/g (not corrected for decay) for the $Li_6Zr_2O_7$ samples irradiated in LOTUS.

5.4 IGA/EPFL

Dierckx's method[7] was used in its original form for chemical processing the samples. Each irradiated sample of lithium-carbonate, having a mass of 0.7 g and a density of 1.34 g/cm^3, was dissolved in 1 ml distilled water using a 20 ml vial. Acetic acid (96 %) of 2.1 ml was added to the it in a later stage. After about one day, 0.7 ml hydrofluoric acid (40 %) was added to the cocktail. As a result of the appropriate chemical reactions, LiF salt was formed in the vial. Liquid scintillation cocktail of 16 ml was added to the vial before it was centrifuged, to precipitate the LiF salt.

A Packard TRICARB β-counter was used for measurement of tritium. Two background

samples were prepared for each series of measurements. Although a background sample was not irradiated, it was chemically prepared under the same conditions as the irradiated ones. Each sample was counted first for 20 minutes. The average values (N_1, in cpm) were the results of three counting cycles. The internal standard method was used in order to know the overall counting efficiency of each sample: A known quantity of tritiated water, with known activity (A_0, in dpm), was added to each sample, and they were counted immediately for a second time. The second average values (N_2, in cpm) were also the results of three counting cycles of 20 minutes each. The efficiency of the samples was defined as follows:

$$\varepsilon = (N_2 - N_1) / A_0 \quad \text{(in cpm/dpm)}.$$

After the efficiency of each sample was determined, only the first counting cycles were considered for the determination of tritium production rate. The average background was subtracted from N_1 counts. The systematic error was estimated to be about 6 %.

The following errors were taken into account. Quadrature addition performed to arrive at the net error.

- Counting statistics
- Efficiency determination
- Pellet mass error
- HTO volume error

5.5 ENEA

Dierckx' method was adopted for chemical processing of the irradiated Li_2CO_3 pellets. The irradiated pellets were divided into three samples, each having a mass about 0.7 ~ 0.8 g. Each sample was put in a polyethylene vial, adding to it 1.0 ml of distilled water and 2.0 ml of acetic acid. One day after, one milliliter of hydrofluoric acid (40 %) was added in the vial. To this solution were also added 15 ml of liquid scintillation cocktail (Hionic-Fluor by Packard). This cocktail, with floating LiF salt, formed by chemical reaction, was centrifuged, to precipitate LiF salt.

Each sample was counted using a Packard Tri-Carb 2250 CA, calibrated with a set of HTO standards, at various quenching. Background samples, i.e. Li_2CO_3 not irradiated, were prepared under the same conditions as the irradiated samples. An external standard was used to determine the efficiency for each sample.

5.6 MEPI

Tritium production rate values were measured by the liquid scintillation counting method, using samples with Li_2CO_3 pellets. The sectional view of the sample pack is shown in **Fig. 5.2**. We used this pack in order to estimate tritium escape during the irradiation.

The irradiation Li_2CO_3 pellet was placed in a reaction flask to which 0.9 ml of 35% hydrochloric and 1.8 ml of 100% acetic acids were added. Complete solution required about 5 hours. The solution was vacuum distilled with an simple apparatus. The solution was frozen in a liquid nitrogen dewar. The apparatus was then evacuated to the pressure of about 120 Pa and the sealed. Then the measuring vial was cooled down with a liquid nitrogen dewar, and the reaction flask was warmed up in a water bath. Under such conditions tritium containing phase was removed from reaction flask and was condensed in the measuring vial. Next, 17 ml of liquid scintillator, 70 ml 4 - izo - pronildiphenil + 80 g 1 - metilnaphthalene + 5 g PPO + 850 ml dioxane, were added to the vial, and the sample was ready for counting in a Mark-III (model 6881) spectrometer.

For control the chemical treatment of samples special Li_2CO_3 pellets irradiated in the cavity of a thermal column F1 reactor were used.

The tritium is incorporated into hydrogen molecules converted into tritiated water using fresh cupric oxide and collected. The escaped tritium rate was (0.2 ± 0.1) % for Li_2CO_3 pellets.

5.7 Osaka University

Dierckx's method[7] was adopted for chemical processing of irradiated samples. The irradiated pellets were divided into four pieces. Each piece was put in a polyethylene vial. Distilled water of 1.2 ml and acetic acid of 2.0 ml were added in the vial. One day after, hydrofluoric acid (46 %) of 1.0 ml was added in the vial. Then liquid scintillation cocktail (Aquasol-II) of 15 ml was added in it. Floating LiF crystals were precipitated at the bottom perfectly by a centrifugal separator. Six liquid scintillation samples for calibration were made using unirradiated samples by the same procedure mentioned above.

Tritium count in each sample was measured by liquid scintillation counting system, "Aloka LSC-703". One measuring time for each sample was 20 minutes, and ten cycles of measurement were carried out.

The external standard source ratio method was adopted to determine the efficiency for each sample. The efficiency curve was calibrated by a commercial standard tritiated water.

The errors were estimated as

$$(\text{error}\ [\%])^2 = (\text{counting statistics error}\ [\%])^2 + (\text{standard HTO error}\ [\%])^2 + (Li_2CO_3\ \text{purity error}\ [\%])^2.$$

The standard HTO error was 4.7 % and the error due to purity of Li_2CO_3 was 0.2 %.

The perturbation effect due to difference of atomic density of lithium between pellets and experimental assemblies were corrected. The correction factors were 0.99894, 0.99874, 1.00891 and 1.00895 for FNS-outer, -inner, LOTUS-outer and inner samples, respectively.

5.8 The University of Tokyo

Each irradiated samples was put in a Teflon vial. Distilled water of 0.5 cc and acetic anhydride of 1.0 cc were added into the vial. After waiting for about one day to complete the chemical reaction; $Li_2CO_3(T) + 2CH_3COOH\ 2nH_2O \rightarrow 2(CH_3COOLi)nH_2O + CO_2 + H_2O$ in each vial, hydrogen fluoride acid (46 %) of 0.3 cc was added with the vial being shook to proceed the reaction; $2(CH_3COOLi)nH_2O + HF \rightarrow LiF + CH_3COOH + nH_2O(T)$. Confirming the formation of white precipitates of LiF, liquid scintillator (Scintisol EX-H, WAKO Chemicals) of 10cc was added and stirred sufficiently by shaking the vial. These vials were left undisturbed overnight in a dark place to deposit the LiF completely at the bottom and to decay the chemical luminescence of the scintillator.

The absolute amount of tritium in each sample was quantified by using the commercial liquid scintillation counting system; "Aloka LSC-903". The quenching correction and efficiency determination in this system were automatically made with the external standard source ratio method. The efficiency curve was obtained from six samples for calibration, which were prepared with standard tritiated water and unirradiated samples in almost the same procedure as above, controlling the degree of quenching. Contents of uncertainty included in the present measurement are estimated as follows;

Counting statistics	: 0.2 ~ 3.0 %
Sample weight	: < 0.5 %
Efficiency determination (including quenching correction)	: 1.5 %
Accuracy of standard tritiated water	: 2.0 %

5.9 JAERI

Irradiated Li_2O pellets were dissolved in 8 cm^3 of distilled water; complete solution required at least two days. The concentration of tritium in the distilled water had been checked and it was found to be background level. The solution was distilled with an apparatus in vacuum to obtain pure water with tritium produced. As the first step of distillation, the solution was poured into a "boiling flask" and frozen in a liquid nitrogen dewar. The apparatus was then evacuated by a rotary vacuum pump; upon complete evacuation, the flask was warmed up in a water bath of 80 ' C while a "condensing flask" was, in turn, cooled down the other water bath of 5 ' C. This resulted in the transfer of almost all of the water to the condensing flask except the water in LiOH. The collected water of 6 cm^3 was pipetted into a 20 ml Teflon vial; then, 14 cm^3 of liquid scintillation cocktail, "Clear Sol", was added. The "Clear Sol" is a new type scintillation cocktail and makes not a two-phase state but clear solution of single phase under wide water fraction (up to 50 %).

Tritium activity in the sample was measured by a low background scintillation counting system (Oken LSC-7100). The external standard ratio method was adapted to obtain the efficiency of each sample. The time chart of measurement for each sample in one cycle was as follows; five repeats of 60 seconds for the pre-ratio measurement, five repeats of 300 seconds for tritium counting and five repeats of 60 seconds for after-ratio measurement. Fourteen cycles were performed for thirty samples; ten irradiated samples, three blind tritiated water samples, three NIST standard tritiated water samples, seven calibration samples and seven background samples. The calibration samples were prepared for determination of a calibration curve. Four of them had several drops of carbon-tetrachloride as quencher. The calibration curve, that was relation between external standard ratio and relative efficiency, was determined with linear fitting by the least square method. The tritium concentration was normalized to that of the NIST standard. Three of background samples were made from distilled water, while four of background samples were made with just the same procedure as that for the irradiated pellets except using unirradiated pellets. No difference were found about counting rate between two kinds of background samples.

The tritium concentration of each sample was deduced by averaging over the data of fourteen cycles and statistic errors were estimated to calculate standard deviations among the obtained tritium concentration for all the cycles. The total tritium escape during the irradiation and chemical treatment was estimated separately and found to be (6.9 ± 0.9) % of total tritium produced. Finally, the tritium production rates were obtained after the correction of the tritium escape and the extrapolation of decay to the irradiation dates. The estimated errors in the measured tritium production rate except those in the statistics errors are as follows:

Sample weight	0.005	%
Transfer efficiency of water during the distillation	0.009	%
NIST tritiated water standard	0.86	%
Half-life of tritium (12.33 ± 0.05 y)	0.02	%
Efficiency for scintillation counting	0.78	%
Escape of tritium	0.97	%
Total except for statistics error	1.5	%

6. Results and Discussions

6.1 General View

Measured tritium production rates (TPRs) reported by the participants are summarized in **Tables 6.1** and **6.2** for the second irradiation experiments (**ICMT-2**) performed at FNS/JAERI and LOTUS/IGA, respectively. As described in chapters 3 and 4, five Li-containing pellets for each participant were irradiated at each neutron source. Three of them (#1 ~ #3) were on the outer position and two of them (#4 and #5) were on the inner position of the rotating disk. These pellets were irradiated under the uniform neutron field by using the rotating disk among three outer or two inner pellets. The uniformity was ascertained by the $^{93}Nb(n,2n)^{92m}Nb$ reaction rate distribution around the rotating disk when the first irradiation experiment was conducted at FNS. The measured values shown in **Tables 6.1** and **6.2** were averaged separately among the outer pellets and among the inner pellets as summarized in **Table 6.3**. **Figure 6.1** shows averaged TPRs without a normalization by the neutron yield. Measured tritium concentrations in the blind HTO samples distributed by JAERI are summarized in **Table 6.4.**

As recognized from **Tables 6.1**, **6.2** and **6.4**, all the participants gave the measured values for tritium concentration of blind sample, while some of them failed to measure the TPRs in the irradiated pellets. The reasons of failure are tritium pollution of vial glasses (CEA), usage of depleted lithium carbonate pellets (ENEA) and unknown factors (ECN). These failures are seen in the results of the first irradiation experiments more frequently. This fact of failure implies that the chemical/physical processing of irradiated pellet to make a tritiated sample for liquid scintillation counting is more difficult technically than the measurement of tritium concentration of the sample. Therefore more attentions should be payed in the processing of irradiated pellets to surely obtain TPRs experimentally.

The TPRs of ENEA for the FNS irradiation are converted to those for natural abundances by factors of 1.037 and 1.040 for inner and outer positions, respectively. The factors are based on the calculated TPRs of lithium-6 and lithium-7. The other failed results are excluded from the **Table 6.3** because it is impossible to compare the failed results with the others in the same condition.

In the **Chapter 5**, the methods of error assignment were described by several participants in detail. Experimental errors were assigned by each participant on the different basis. It is not meaningful to compare the absolute values of the errors assigned on different basis, and the assigned errors only gave the outline of experimental accuracy. However the following fact could be found from the assigned errors. All the errors for tritium concentration of the blind samples are less than 3.2 %, while there are some errors larger than 5 % and up to 10 %

for measured TPRs for both FNS and LOTUS. The required accuracy of 5 % for TPR measurement was not satisfied as long as the assigned errors are considered. To obtain the accuracy of 5 %, some organizations have to examine the sources of errors and to take some counter-measures to decrease them.

6.2 Result of Blind HTO Sample

Whole procedure of TPR measurement is divided into three steps:

Step-1 irradiation of Li-containing pellets,

Step-2 chemical/physical processing of the irradiated pellets into HTO-containing samples,

Step-3 counting of the samples combined with the calibration by tritium standard.

As mentioned in the above section, Li-containing pellets were irradiated under the uniform neutron field. The uniformity of the irradiation is guessed better than 1 %, so that there are little possibility in the **Step-1** to distort the measured TPRs among the organizations originated. The causes of unevenness of measured TPRs are mostly due to in the processes of the **Step-2** and **-3**. It is not possible to distinguish the causes of unevenness between the **Step-2** and the **Step-3**. While the measurement process of tritium concentration in the blind HTO sample concerns only the **Step-3** and enable to estimate the status about measurement accuracy of tritium concentration.

Measurements of tritium concentration in tritiated water were carried out three times during this program, that is, measurements of the blind sample for the **ICMT-1**, those of the ANL sample and those of the blind sample for the **ICMT-2**. Their results are shown in **Tables 2.4, 2.5** and **6.4,** respectively. **Figure 6.2** shows all the results. The standard deviation of **ICMT-2** (8.5 %) is larger than that of **ICMT-1** (3.0 %) and the ANL sample (2.8 ~ 7.5 %). It is found from the **Fig. 6.2** that the standard deviations above 50 Bq/g are almost constant around 3 %, while they become worse with decrement of tritium concentration below 50 Bq/g. This fact implies that the standard deviation depends on signal to background (noise) ratio (S/N ratio) below about 50 Bq/g. The tritium concentrations of extracted tritiated water for all the participants are roughly estimated as 1 ~ 5 Bq/g for FNS irradiation and 2.5 ~ 20 Bq/g for LOTUS irradiation, and these values are indicated at the right side of **Fig. 6.2**. Below the tritium concentration of 20 Bq/g, which is the maximum tritium concentration obtained from irradiated pellets, it is expected that the standard deviation of measured tritium concentration among organizations exceeds the 5 %, the expected accuracy on this program.

From the results of blind sample, it is concluded that to satisfy the expected accuracy of 5 % is difficult even for the measurement of tritium concentration of less than 20 Bq/g, and

much more for the measurement of TPRs. The tritium concentration of 20 Bq/g is not small values when we do this kind of irradiation experiment of pellets in a simulated fusion blanket using the existing D-T neutron sources. Hence it is necessary for the accuracy of 5 % to increase the S/N ratio in the process of liquid scintillation counting, that is, to increase the tritium concentration of extracted tritiated water from pellets and to reduce the background level.

6.3 Consistency of TPRs within an Organization

Measured TPRs by an organization completely at the identical neutron field, such as three TPRs of outer band of rotating disk for the second irradiation at FNS, must be consistent within the assigned errors in themselves if all the errors are accounted for. From **Tables 6.1** and **6.2**, most of the organizations give consistent results for the second irradiation. But the measured TPRs by CEA scatter more than three times of their errors which are assigned with confidence limit of 90 %. So there would be unknown error sources in the CEA data.

The TPR ratios of inner band to outer band and those of FNS to LOTUS are shown in **Table 6.3**. Standard deviations for these ratios among participants, 2.9 %, 5.6 % and 6.7 %, are much smaller than those for absolute TPRs, around 10 %. Considering the assigned errors, these ratios agree among the organizations. Hence consistency for relative measurements by each organization is confirmed.

From discussions in this section, it is concluded that most of the organizations give relatively consistent results as long as we discuss the results of one organization.

6.4 Comparison of Absolute TPRs among Organizations

Measured TPRs averaged over some pellets are given in **Table 6.3**. The values scatter ± 15 ~ 20 % to the average among the organizations. Standard deviations of TPRs are 10 ~ 14 % for four irradiation field, that is, FNS-outer, -inner, LOTUS-outer and -inner fields. No differences of standard deviations are recognized between FNS and LOTUS irradiations which are different neutron fluence levels. In contrast, in the **ICMT-1**, standard deviations for LOTUS, of which the neutron fluence are six times larger than FNS, are 10 % and better than the deviations of 30 ~ 50 % for FNS. The measurement accuracy of TPRs for the **ICMT-2** at FNS, comparing with the first irradiation, is largely improved to almost the same accuracy (10 %) as LOTUS irradiation. But the standard deviations for the **ICMT-2** at both

FNS and LOTUS, 10 ~ 14 %, are not different so much from those for the **ICMT-1** at LOTUS, 10 %. The measurement accuracy in relatively poor S/N ratios as the FNS irradiation level is improved largely by second trial, while no progress is seen in high S/N ratios as the LOTUS irradiation level.

In **Table 6.3** all the assigned errors are less than 10 % while the standard deviations for TPRs among the organizations are more than 10 %. So it is not possible to conclude that the measured TPRs agree each other, and there may be unknown error sources for some or all the organizations.

6.5 Normalization by Measured Concentration of Blind HTO

Standard deviations of measured tritium concentration for blind HTO are as follows.

- all nine organizations 8.5 %
- seven organizations which gives TPRs for LOTUS irradiation 5.4 %
- seven organizations which gives TPRs for FNS irradiation 8.9 %

The standard deviations of 5.4 % and 8.9 % of blind HTO can not be neglected against the about 10 % of standard deviations of TPRs. If we assume that the deviation of blind HTO is due to a difference of calibrated HTO standard used by individual organizations to make tritium concentration absolute, deviations of the same degree as blind HTO are included in the measured TPRs. When we normalize the measured TPRs of each organization as the measured concentration of blind HTO become the same values for all the organizations, we can exclude the influence of the difference of HTO standard used. In other words, we can extract only **Step-3** mentioned in the **Section 6.2** from the mixture of **Step-2** and **-3** by the normalization.

Figures 6.3 and **6.4** show ratios of measured absolute TPRs to averaged TPRs among organizations for the second irradiation before and after the normalization, respectively. From these figures, it is seen that the deviation of TPRs is improved in some degree by the normalization. This improvement is mainly due to an approach of Osaka Univ. to the other organizations. Changes of standard deviation are summarized in **Table 6.5**. In **ICMT-1**, an accuracy of blind HTO is high and no improvements of standard deviation by the normalization are seen. Whereas in **ICMT-2**, standard deviations are decreased about 1 % by the normalization. In this case, the following relation is almost held.

$$(\sigma_{before})^2 = (\sigma_{after})^2 + (\sigma_{blind})^2$$

where σ_{before}, σ_{after} and σ_{blind} are standard deviations of TPRs before and after the normalization and that of blind HTO, respectively. These three standard deviations are regarded as those for **Step-2** + **Step-3**, **Step-2** only and **Step-3** only, respectively. So it is concluded that the

measurement accuracy of TPR for present experiment can be separated into two components; about 10 % for **Step-2** and about 7 % for **Step-3**. The accuracy in **Step-2** is much worse than that in **Step-3**.

6.6 Systematic Error and Random Error

As mentioned in the **Section 6.3**, standard deviations for absolute TPRs are 10 ~ 14 % and those for three relative ratios of TPR, inner/outer for FNS and LOTUS and FNS/LOTUS, are 2.9 %, 5.6 % and 6.7 %, respectively. Standard deviations of these relative ratios are not made by systematic errors but random errors because any systematic errors are canceled each other in a derivation process of TPRs. So the standard deviations of relative ratios are due to only random errors and those of absolute TPRs are due to both random and systematic errors. Assuming that a typical standard deviation due to random errors (σ_{ran}) as 5 %, that due to both errors (σ_{tot}) as 11 % and the following equation,

$$(\sigma_{tot})^2 = (\sigma_{ran})^2 + (\sigma_{sys})^2$$

where σ_{sys} is a standard deviation due to systematic errors, σ_{sys} is about 10 %. The σ_{sys} of 10 % is much larger than the σ_{ran} of 5 %. It can be pointed out that the deviations of absolute TPRs are not caused by random errors but systematic errors.

6.7 Comparisons between Organizations

Standard deviations among all the organization have been discussed as an indicator of the agreement in this chapter so far. In this section, we focus on comparisons of TPRs between organizations. The following facts are found from the **Figs. 6.3** and **6.4**.

- MEPI and JAERI agree within 5 % for all the four positions before and after the normalization. But their values are larger than those of the other organizations.
- Any other couples of organizations do not agree within 10 % for all the four positions both before and after the normalization.
- CEA, IGA and the Univ. of Tokyo agree within 10 % for all the four positions before the normalization. But this agreement become worse by the normalization.
- IGA and Osaka Univ. agree within 5 % except for the LOTUS-inner position after the normalization.
- CRL gives 10 % smaller results for FNS irradiation relative to LOTUS irradiation comparing with the other organizations.

- The Univ. of Tokyo gives 7 % larger results for the FNS irradiation relative to LOTUS irradiation comparing with the other organizations.

An agreement of TPRs within 10 % is achieved by few couples of organizations. Because we do not know the true TPRs which only the god know, we can not point out which organizations is reliable or not.

6.8 Comparison with Calculation

Calculated TPRs for FNS irradiation are shown in **Tables 2.3** and **6.3**. It is noticed that an agreement between measured and calculated TPR gives only an outline of goodness of the measured value roughly. A measured TPR which is the closest to the calculation is not always the most accurate value. Because the calculated TPRs are not true values due to accuracy of used cross sections, transport code and modeling, and the measured TPRs may include some errors which commonly included in the results for all the organizations such as errors of neutron yield and location of assembly.

For **ICMT-2** most of the measured TPRs are smaller than the calculated ones and ratios of calculated to measured values range from 0.97 to 1.34. MEPI and JAERI gives the closest TPRs to the calculation within ± 3 %. ENEA agrees with the calculation within 10 %. Calculated to measured values (C/Es) of IGA and Univ. of Tokyo are about 1.13.

The results of the Univ. of Tokyo and JAERI agree with the calculation within ± 12 % for both **ICMT-1** and **ICMT-2**. The ratio of inner/outer for **ICMT-2** agrees between the calculation and the average of measurement.

7. Concluding Remarks

Following the first irradiation experiment (**ICMT-1**) and the measurement of ANL samples, the second irradiation experiment (**ICMT-2**) was successfully conducted for both FNS and LOTUS. The FNS irradiation is roughly characterized as 4×10^{-13} [T-atoms/Li-atom] or 1 ~ 5 [Bq/g] in concentration of extracted HTO, and the LOTUS irradiation is as 1.6×10^{-12} [T-atoms/Li-atom] or 2.5 ~ 20 [Bq/g] in concentration of extracted HTO. Comparing with **ICMT-1**, standard deviation of tritium production rate (TPR) measurement among the organizations are largely improved to about 10 % for the FNS irradiation; relatively low fluence. While for the LOTUS irradiation, relatively high fluence, the standard deviations of **ICMT-1**, i.e., 10 %, do not progressed in the **ICMT-2**. As a result, it is found that the present accuracy of TPR measurement is about 10 % in one standard deviation and measured TPRs are scattered within ± 20 % among organizations. This present status is far from the expected accuracy of this international comparison program, 5 %.

Through examinations about measured data, some problems on TPR measurement are clarified. Only in a process of tritium concentration measurement for less than a few tens Bq/g, a standard deviation of measured concentration is expected to be larger than 5 %. In fact in present blind HTO measurement, 30 Bq/g in concentration, the actual standard deviations are larger than 5 %. But this concentration is higher than any concentrations obtained from irradiated pellets for the present experiments, and they are not low but highest class of concentrations as those extracted from pellet irradiated by existing D-T neutron sources. To achieve the accuracy of 5 % for TPR measurement, it is necessary to make measurement accuracy of tritium concentration below a few tens of Bq/g higher, or to extract tritiated water of which concentration is higher than a few tens of Bq/g.

It is found that the most of deviations among organizations (1σ = 10 %) are systematic and due to extraction processes of HTO from irradiated pellets. These systematic errors are inherent in each organization, and experimenters are not conscious of all the error sources.

Consequently, the accuracy of the "relative" TPR measurement by most of organizations were less than 5 %. But the standard deviation among organizations exceeds 10 % and it is larger than the required accuracy of 5 %. These must be unknown error sources depending on the organizations.

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The authors would like to thank Drs. Y. Kaneko and T. Asaoka of JAERI for their promotion of this program. They are also grateful to the members of NEACRP/NEA-NSC for their support of this program, and to the stuffs of FNS and LOTUS facilities for their operation and support of the irradiation experiments.

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Table 1.1 List of participants for the first irradiation experiment (ICMT-1).

Organization	Abbreviation	Country	Personnel(s)
Argonne National Laboratory	ANL	U.S.A.	K. Porges
Atomic Energy of Canada Limited Chalk River Nuclear Laboratories	AECL/CRNL	Canada	I. J. Hastings J. M. Miller J. D. Sullivan
Commissariat a l'Energie Atomique Cadarache	CEA/Cadarache	France	P. Michaille
Institut de Genie Atomique École Polytechnique Fédérale de Lausanne	IGA/EPFL	Switzerland	P. -A. Haldy S. Azam
delle'Energia Nucleare e delle Energie Alternaive	ENEA	Italy	A. Moauro
Osaka University	Osaka Univ.	Japan	Takahashi, A. Yoshida, S.
The University of Toktyo	Univ. of Tokyo	Japan	Iguchi, T. Nakazawa, M.
Japan Atomic Energy Research Institute	JAERI	Japan	Maekawa, H.

Table 1.2 List of participants for the second irradiation experiment (ICMT-2).

Organization	Abbreviation	Country	Personnel(s)
Atomic Energy of Canada Limited Chalk River Laboratories	AECL/CRL	Canada	I. J. Hastings J. M. Miller J. D. Sullivan
Commissariat a l'Energie Atomique Cadarache	CEA/Cadarache	France	C. Latge S. Desreumaux
Energieonderzoek Centrum Nederland	ECN	Netherlands	H. Kwast
Institut de Genie Atomique École Polytechnique Fédérale de Lausanne	IGA/EPFL	Switzerland	P. -A. Haldy S. Azam O. P. Joneja
delle'Energia Nucleare e delle Energie Alternaive	ENEA	Italy	A. Moauro
Moscow Engineering Physics Institute	MEPI	Russian Federation	V. L. Romodanov Y. M. Verzilov
Osaka University	Osaka Univ.	Japan	Takahashi, A. Yoshida, S.
The University of Toktyo	Univ. of Tokyo	Japan	Iguchi, T. Nakazawa, M.
Japan Atomic Energy Research Institute	JAERI	Japan	Maekawa, F. Maekawa, H.

Table 2.1 Measured tritium production rates for ICMT-1 at FNS/JAERI,

Organization	Measured tritium production rate [x10^{-29} T atoms/Li atom/Source Neutron], (error %)									
	outer position						inner position			
	#1		#2		#3		#4		#5	
ANL	4.394	-	-	-	4.432	-	5.233	-	5.189	-
AECL/CRNL	-	-	1570	(5.7)	3730	(4.1)	675.6	(7.0)	517.8	(4.2)
IGA/EPFL*	1.612	-	1.804	-	1.808	-	2.174	-	2.095	-
ENEA	-	-	-	-	-	-	-	-	-	-
CEA/Cadarache	9.32	(4.4)	5.56	(4.4)	7.93	(4.4)	7.39	(4.4)	5.36	(4.4)
Univ. of Tokyo	-	-	4.00	(3.3)	3.94	(3.3)	4.51	(3.3)	4.57	(3.3)
Osaka Univ.	2.95	(5.1)	2.96	(5.1)	2.85	(5.1)	3.30	(5.1)	3.44	(5.1)
JAERI	3.983	(3.3)	3.89	(3.0)	3.97	(2.5)	4.40	(3.6)	4.42	(2.9)

* Statistical errors are bigger than 15 %.

Table 2.2 Measured tritium production rates for ICMT-1 at LOTUS/IGA.

Organization	Measured tritium production rate [x10^{-29} T atoms/Li atom/Source Neutron], (error %)									
	outer position						inner position			
	#1		#2		#3		#4		#5	
ANL	2.378	-	2.415	-	2.420	-	2.651	-	2.608	-
AECL/CRNL	172.1	(3.2)	199.3	(2.9)	365	(2.6)	311	(2.7)	328	(2.2)
IGA/EPFL	1.823	(2)	2.061	(2)	1.817	(1)	2.009	(1)	2.235	(1)
ENEA	-	-	-	-	-	-	-	-	-	-
CEA/Cadarache	2.747	(5.3)	2.507	(3.4)	2.439	(3.4)	3.618	(3.4)	2.986	(5.1)
Univ. of Tokyo	2.20	(2.6)	2.25	(2.6)	2.21	(2.6)	2.36	(2.6)	2.42	(2.6)
Osaka Univ.	2.11	(4.7)	2.09	(4.7)	2.11	(4.7)	2.30	(4.7)	2.24	(4.7)
JAERI	2.215	(1.3)	2.250	(1.4)	2.251	(1.4)	2.417	(1.4)	2.414	(1.8)

Table 2.3 Averaged tritium production rates for ICMT-1 experiments at FNS/JAERI and LOTUS/IGA.

Organization	Averaged TPR [x10^{-29} T atoms/Li atom/Source Neutron], (error %[1)]) and ratio											
	FNS irradiation					LOTUS irradiation					ratio FNS(in+out)/LOTUS(in+out)
	outer TPR	outer error[1)]	inner TPR	inner error	ratio inner/outer	outer TPR	outer error	inner TPR	inner error	ratio inner/outer	
ANL	4.41	(-)	5.21	(-)	1.18	2.40	(-)	2.63	(-)	1.09	1.91
AECL/CRNL	2650	(4.9)	2927	(5.6)	1.10	246	(2.9)	320	(2.5)	1.30	9.85
IGA/EPFL	1.74	(-)	2.14	(-)	1.23	1.90	(1.7)	2.12	(1)	1.12	0.97
ENEA	-	(-)	-	(-)	-	-	(-)	-	(-)	-	-
CEA/Cadarache[2)]	7.60	(4.4)	6.38	(4.4)	0.84	2.56	(4.0)	2.80	(4.3)	1.09	2.61
Univ. of Tokyo	3.97	(3.3)	4.54	(3.3)	1.14	2.22	(2.6)	2.39	(2.6)	1.08	1.83
Osaka Univ.	2.92	(5.1)	3.37	(5.1)	1.15	2.10	(4.7)	2.27	(4.7)	1.08	1.44
JAERI	3.90	(2.9)	4.41	(3.3)	1.13	2.24	(1.4)	2.42	(1.6)	1.08	1.78
Average[2)] [1σ]	4.09 [48.1 %]		4.33 [33.8 %]		1.11 [12.4 %]	2.24 [10.3 %]		2.44 [10.0 %]		1.09 [1.4 %]	1.76 [30.9 %]
Calculation	4.292		4.862		1.13						

1) The error was calculated simply as the average of original errors.

2) The average and the standard deviation (1σ) were calculated for six organizations except AECL/CRNL and ENEA.

Table 2.4 Measured tritium concentration in the blind sample distributed at the ICMT-1 experiment by JAERI.

Organization	Tritium Concentration [Bq/g]	error [%]	Measured / Average
ANL	65.82	(1.5)	0.977
AECL/CRNL	66.6	(6)	0.988
IGA/EPFL	69	(6)	1.024
ENEA	66.14	(1.13)[1)]	0.981
CEA/Cadarache	66.64	(2.5)	0.989
Univ. of Tokyo	71.0	(2.5)	1.054
Osaka Univ.	65.63	(1.70)[2)]	0.974
JAERI	68.25	(1.20)	1.013
Avarage [1σ]	67.39	[±1.88, 2.79 %]	1.000

1) Statistic error only (1σ)
2) Statistic error only (3σ)

Table 2.5 Tritium concentration in the samples distributed by ANL.

Organization	Tritium Concentration, (error %)						Measured / Assigned		
	ANL-A		ANL-B		ANL-C		ANL-A	-B	-C
ANL	257.3	(1.06)	48.42	(0.83)	3.893	(0.54)	1.001	0.998	1.006
AECL/CRNL	261.8	(1.75)	49.0	(1.83)	3.88	(2.4)	1.019	1.009	1.003
IGA/EPFL	258.8	(0.4)	51.5	(1.0)	4.1	(3.2)	1.007	1.061	1.059
ENEA	270.2	(2.0)	50.4	(1.0)	4.69	(8.1)	1.051	1.038	1.212
CEA/Cadarache	253.6	(0.17)	48.10	(0.52)	3.798	(0.61)	0.987	0.991	0.981
Univ. of Tokyo	247.3	(0.77)	46.88	(1.2)	3.78	(4.0)	0.962	0.966	0.977
Osaka Univ.	270.9	(0.28)	52.02	(0.64)	4.172	(2.2)	1.054	1.072	1.078
JAERI	260.0	(1.28)	49.12	(1.23)	3.898	(1.46)	1.012	1.012	1.007
Assigned	257.0	(0.86)	48.54	(0.86)	3.87	(0.86)	1.000	1.000	1.000
Average [1σ]	260.0 [3.04 %]		49.43 [3.55 %]		4.026 [7.48 %]		1.012	1.018	1.040

Table 3.1 Specification of pellets for ICMT-2.

Organization	Chemical form	Size [mm]*	^{6}Li ratio [atom %]**
AECL/CRL	Li_2CO_3	23 ϕ x 2.3	7.44 ± 0.01
CEA/Cadarache	Li_2CO_3	15 ϕ x 6	7.60 ± 0.01
ECN	Li_8ZrO_6	17.8 ϕ x 2	7.52 ± 0.02
	$Li_6Zr_2O_7$	17.8 ϕ x 2	7.54 ± 0.01
IGA/EPFL	Li_2CO_3	15 ϕ x 6	7.60 ± 0.01
ENEA	Li_2CO_3	26 ϕ x 4	5.48 ± 0.01
MEPI	Li_2CO_3	26 ϕ x 6	7.66 ± 0.01
Osaka Univ.	Li_2CO_3	24 ϕ x 2.3	7.57 ± 0.01
Univ. of Tokyo	Li_2CO_3	10 ϕ x 5	7.56 ± 0.02
JAERI	Li_2O	18 ϕ x 2.4	7.53 ± 0.01

* Including sealing material.
** The mass analysis was done by Mr. and Mrs. tamura of Analytical Chemistry Laboratory, JAERI.

Table 3.2 Homogenized nuclide density for each region.

Region*	Li_2O^{*1}	Li_2O^{*2}	SS^{*3}	SS^{*4}	Al	Air
^{6}Li	4.1915-3**	4.2211-3	----	----	----	----
^{7}Li	5.2404-2	5.2774-2	----	----	----	----
^{16}O	2.8298-2	2.8498-2	----	----	----	1.040-5
^{14}N	----	----	----	----	----	3.881-5
Cr	3.0675-4	1.4333-4	6.3975-3	1.5302-2	----	----
Fe	1.1307-3	5.2773-4	2.3574-2	6.1471-2	----	----
Ni	1.3516-4	6.2963-5	2.8121-3	8.2258-3	----	----
^{27}Al	----	----	----	----	6.0039-2	----

* See **Fig. 3.5**.
** Read as 4.1915×10^{-3} [10^{24} atoms/cm^3].

Table 5.1 Summary of measurement method of tritium production rate for ICMT-2.

Organization	Chemical form	Sample size [mm]	Sample weight [g]	Processing Method	Material of vial	(a)	(b)	(c)	Counting efficiency	Measurment time per vial [min]	B.G. level [cps/vial]	S/N ratio (FNS)	S/N ratio (LOTUS)
AECL/CRN	Li_2CO_3	23 ϕ x 2.3	1.0	dissolution	glass	int.	20 % x ?	0.33	25.1 %	120 (FNS) 60 (LOTUS)	0.61	0.8	2.0
CEA/Cadarache	Li_2CO_3	17.8 ϕ x 2	0.7	Dierckx' (cryo-distillation)	glass	ext.	100 %	1.1	26.6 %	1200 (FNS) 320 (LOTUS)	0.292	-	25
ECN	Li_8ZrO_6 $Li_6Zr_2O_7$	15 ϕ x 6	- -	dissolution and cryo-distillution	- -	- -	- -	- -	- -	- -	- -	- -	- -
IGA/EPFL	Li_2CO_3	17.8 ϕ x 2	0.7	Dierckx'	glass	int.	100 %	1.1	20.58	120 -	0..233	7.7	28
ENEA	Li_2CO_3	26 ϕ x 4	2.1~2.4	Dierckx'	P.E.	ext.	33 % x 3	1.2	22.7 % (FNS) 23.5 % (LOTUS)	180	0.231	10	46
MEPI	Li_2CO_3	26 ϕ x 6	3	cryo-distillation	glass	int.	25 % x 3	1.2	21 %	300	0.47	4.6	17
Osaka Univ.	Li_2CO_3	24 ϕ x 2.3	2	Dierckx'	P.E.	ext.	25 % x 4	0.8	18.59 %	200	0.245 (FNS) 0.215 (LOTUS)	5.5	24
Univ. of Tokyo	Li_2CO_3	10 ϕ x 5	0.64	Dierckx'	Teflon	ext.	100 %	1.0	29.8 %	900	0.23	9.8	33.0
JAERI	Li_2O	18 ϕ x 2.4	0.38	cryo-distillation	Teflon	ext.	75 %	1.1	25.7 %	350	0.083	27	101

(a) Method of efficiency determination: ext.; external standard method int.; internal standard method

(b) Transfer efficiency; (tritium amount transfer into a vial) / (tritium amount produced in a pellet)
(x 3) and (x 4) means that three and four vials were produced from a pellet, respectively.

(c) Amount of tritium in a vial (in arbitrary unit)

Table 5.2 Measured results for FNS irradiation by CEA/Cadarache.

Countings					Efficiency	Activities							
Sample	$\bar{d}$	$\bar{t}$	$\bar{b}$	$\overline{blank}$	%	d	t	b	Total Activity	m Li	Activity	data(1991)	Activity on 20/04/91
(i)	cpm	cpm	cpm	cpm		Bq	Bq	Bq	Bq	g	Bg/g	dd/mm	Bq/g
1	61.94	43.60	42.35	22.00	35.0	38.03	6.04	10.58	54.65	0.1380	396.0	13/08	402.3
2	141.40	41.60	52.76	26.40	35.0	54.58	4.18	13.11	71.86	0.1396	514.8	10/09	525.7
3	48.90	37.60	36.85	31.10	35.0	8.48	1.82	3.28	13.58	0.1366	99.4	07/10	101.8
4	50.41	39.50	38.44	32.40	35.0	8.57	1.99	3.53	14.09	0.1340	105.2	21/10	108.0
5	31.74	26.92	26.52	24.30	35.0	7.09	0.74	1.58	9.41	0.1320	71.17	04/11	72.8

Table 5.3 An example of the range of measured tritium concentrations in nCi/g (not corrected for decay) for the $Li_6Zr_2O_7$ samples irradiated in LOTUS.

Sample	outer band		inner band	
	distillate	residue	distillate	residue
6	8-12	5-93		
7	5-26	8-144		
8	15-77	26-258		
9			5-33	6-13
10			10-79	10-300

Table 6.1 Measured tritium production rates for ICMT-2 irradiated at FNS/JAERI.

Organization	Measured tritium production rate [x10^{-29} T atoms/Li atom/Source Neutron], (error %)									
	outer position						inner position			
	#1		#2		#3		#4		#5	
AECL/CRL[1]	3.79	(10.6)	3.87	(10.0)	3.88	(9.2)	4.29	(8.8)	3.92	(9.2)
CEA/Cadarache[2]	27.55[4]	(7.5)	36.00[4]	(8.25)	6.976	(5.4)	7.416	(4.8)	5.015	(3.1)
ECN	-	-	-	-	-	-	-	-	-	-
IGA/EPFL	3.851	(6)	3.753	(6)	3.731	(6)	4.176	(6)	4.254	(6)
ENEA[3]	4.062	(4.0)	4.120	(6.4)	3.985	(7.1)	4.576	(6.3)	4.182	(3.1)
MEPI	4.17	(3.5)	4.21	(3.5)	4.16	(3.5)	4.88	(3.5)	4.81	(3.5)
Osaka Univ.	3.232	(8.02)	3.157	(7.81)	3.217	(7.88)	3.732	(8.08)	3.631	(7.86)
Univ. of Tokyo	3.837	(3.91)	3.767	(3.11)	3.829	(3.08)	4.393	(4.12)	4.276	(3.86)
JAERI	4.38	(2.4)	4.35	(3.0)	4.41	(2.3)	4.95	(2.5)	5.05	(2.3)

1) The error was estimated as (2σ counting error) + (weighting error).
2) The error was estimated with a confidence limit of 90 %.
3) Results of ENEA are not comparable with the others because the isotope ratio of samples is not natural abundance.
4) Glass polution because of previous analysis on high tritiated wastes

Table 6.2 Measured tritium production rates for ICMT-2 irradiated at LOTUS/IGA.

Organization	Measured tritium production rate [x10^{-29} T atoms/Li atom/Source Neutron], (error %)									
	outer position						inner position			
	#1		#2		#3		#4		#5	
AECL/CRL[1]	1.98	(6.8)	2.32	(6.0)	2.03	(6.3)	2.39	(6.3)	2.35	(5.7)
CEA/Cadarache[2]	2.199	(4)	1.715	(3)	2.132	(3)	1.963	(3)	2.097	(3)
ECN	-	-	-	-	-	-	-	-	-	-
IGA/EPFL	2.014	(6)	1.995	(6)	-	-	2.140	(6)	2.150	(6)
ENEA[3]	2.048	(0.45)	2.029	(1.30)	2.052	(0.56)	2.206	(1.16)	2.200	(0.64)
MEPI	2.20	(3.5)	2.23	(3.5)	2.22	(3.5)	2.42	(3.5)	2.44	(3.5)
Osaka Univ.	1.704	(8.12)	1.609	(7.84)	1.622	(8.23)	1.609	(8.71)	1.502	(9.99)
Univ. of Tokyo	1.839	(2.00)	1.858	(1.85)	1.867	(2.15)	1.999	(1.84)	2.017	(1.88)
JAERI	2.21	(1.8)	2.19	(1.8)	2.21	(1.9)	2.47	(1.7)	2.37	(1.7)

1) The error was estimated as (2σ counting error) + (weighting error).
2) The error was estimated with a confidence limit of 90 %.
3) Results of ENEA are not comparable with the others because the isotope ratio of samples is not natural abundance.

Table 6.3 Averaged tritium production rates for ICMT-2 experiments at FNS/JAERI and LOTUS/IGA.

Organization	Averaged TPR [x10^{-29} T atoms/Li atom/Source Neutron], (error %[4]) and ratio											
	FNS irradiation					LOTUS irradiation					ratio FNS(in+out) / LOTUS(in+out)
	outer		inner		ratio	outer		inner		ratio	
	TPR	error[3]	TPR	error	inner/outer	TPR	error	TPR	error	inner/outer	
AECL/CRL[1]	3.85	(9.9)	4.11	(9.0)	1.07	2.11	(6.4)	2.37	(6.0)	1.12	1.78
CEA/Cadarache[2]	-	(-)	-	(-)	-	2.02	(3.3)	2.03	(3.0)	1.01	-
ECN	-	(-)	-	(-)	-	-	(-)	-	(-)	-	-
IGA/EPFL	3.78	(6.0)	4.22	(6.0)	1.12	2.00	(6.0)	2.15	(6.0)	1.07	1.93
ENEA[3]	4.22	(5.8)	4.54	(4.7)	1.08	-	(-)	-	(-)	-	-
MEPI	4.18	(3.5)	4.85	(3.5)	1.16	2.22	(3.5)	2.43	(3.5)	1.10	1.94
Osaka Univ.	3.20	(7.9)	3.68	(8.0)	1.15	1.65	(8.1)	1.56	(9.4)	0.95	2.14
Univ. of Tokyo	3.81	(3.4)	4.33	(4.0)	1.14	1.85	(2.0)	2.01	(1.9)	1.08	2.11
JAERI	4.38	(2.6)	5.00	(2.4)	1.14	2.20	(1.8)	2.42	(1.7)	1.10	2.03
Average [1σ]	3.92 [10.0 %]		4.39 [10.3 %]		1.12 [3.1 %]	2.01 [10.1 %]		2.14 [14.1 %]		1.07 [5.6 %]	1.99 [6.7 %]
Calculation	4.292		4.862		1.13						

1) The error was estimated as (2σ counting error) + (weighting error).

2) The error was estimated with a confidence limit of 90 %.

3) TPRs for FNS were corrected for the isotope ratio of natural lithium on the basis of the calculation.

4) The error was calculated simply as the average of original errors.

Table 6.4 Measured tritium concentration in the blind sample distributed at the second irradiation experiment by JAERI.

Organization	Tritium Concentration [Bq/g]	error [%]	Measured / Average
AECL/CRNL	31.3	0.9	0.983
CEA/Cadarache	30.16	2.6*	0.947
ECN	29.964	1.1	0.941
IGA/EPFL	34.4	1	1.079
ENEA	37.77	0.7	1.186
MEPI	31.1	3.2	0.977
Osaka Univ.	28.908	1.07	0.908
Univ. of Tokyo	31.12	2.08	0.977
JAERI	31.84	1.5	1.000
Avarage [1σ]	31.84	[±2.69, 8.45 %]	1.000

* The error was estimated with a confidence of 90 %.

Table 6.5 Changes of standard deviations by normalization with the blind HTO water as the standard.

Irradiation		Standard deviation		
		before normalization	after normalization	blind HTO water
ICMT-1	FNS-outer	48.1 %	49.2 %	2.8 %
	FNS-inner	33.8 %	34.7 %	
	LOTUS-outer	10.3 %	11.7 %	
	LOTUS-inner	10.0 %	11.6 %	
ICMT-2	FNS-outer	10.0 %	9.3 %	8.9 %
	FNS-inner	10.3 %	11.1 %	
	LOTUS-outer	10.1 %	9.0 %	5.4 %
	LOTUS-inner	14.1 %	12.7 %	

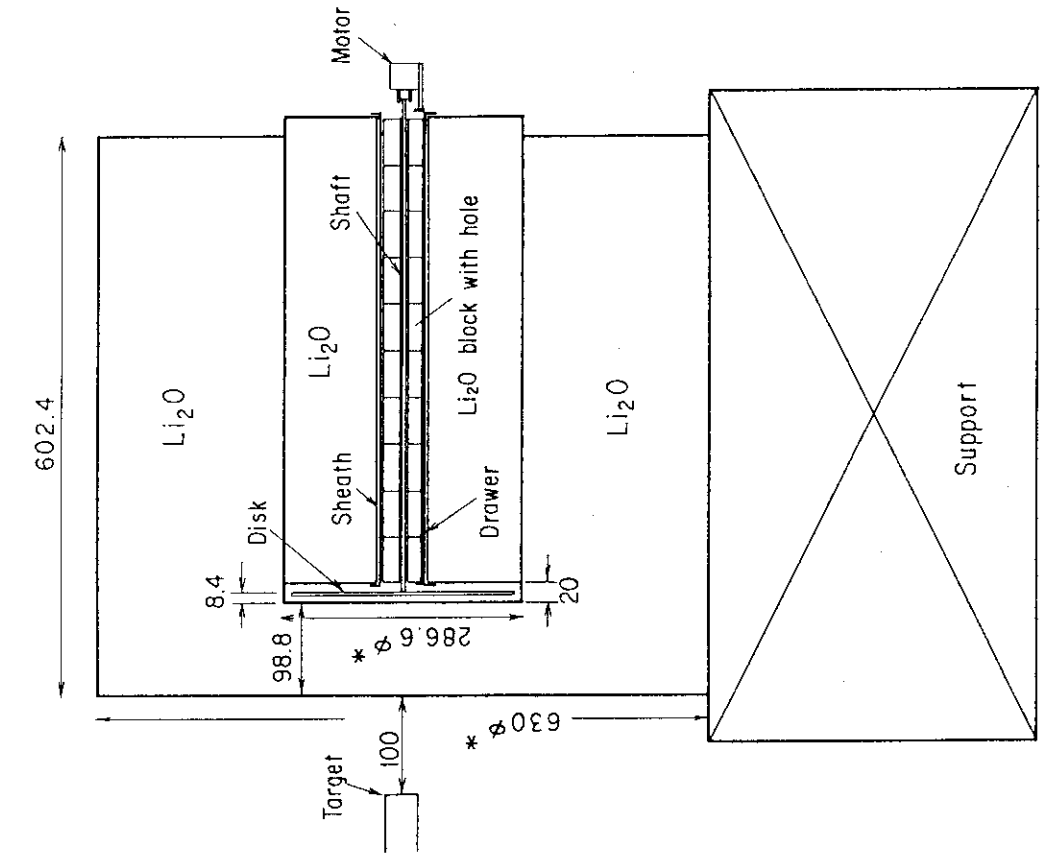


Fig. 3.2 Experimental arrangement at FNS.

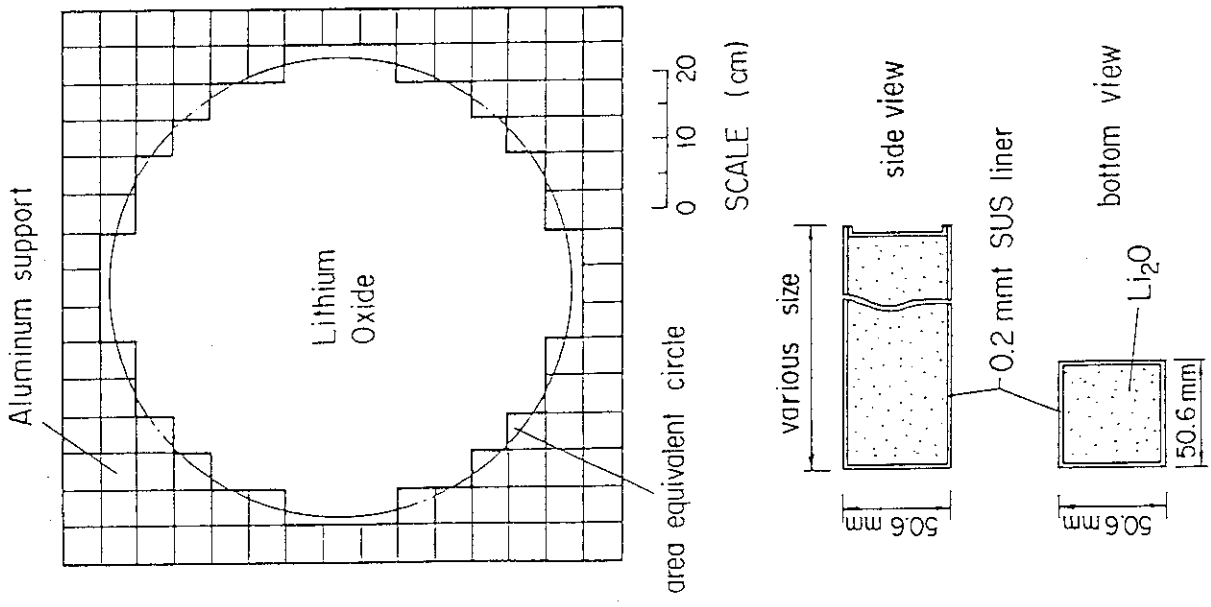


Fig. 3.1 Sectional views of Li_2O assembly and blocks.

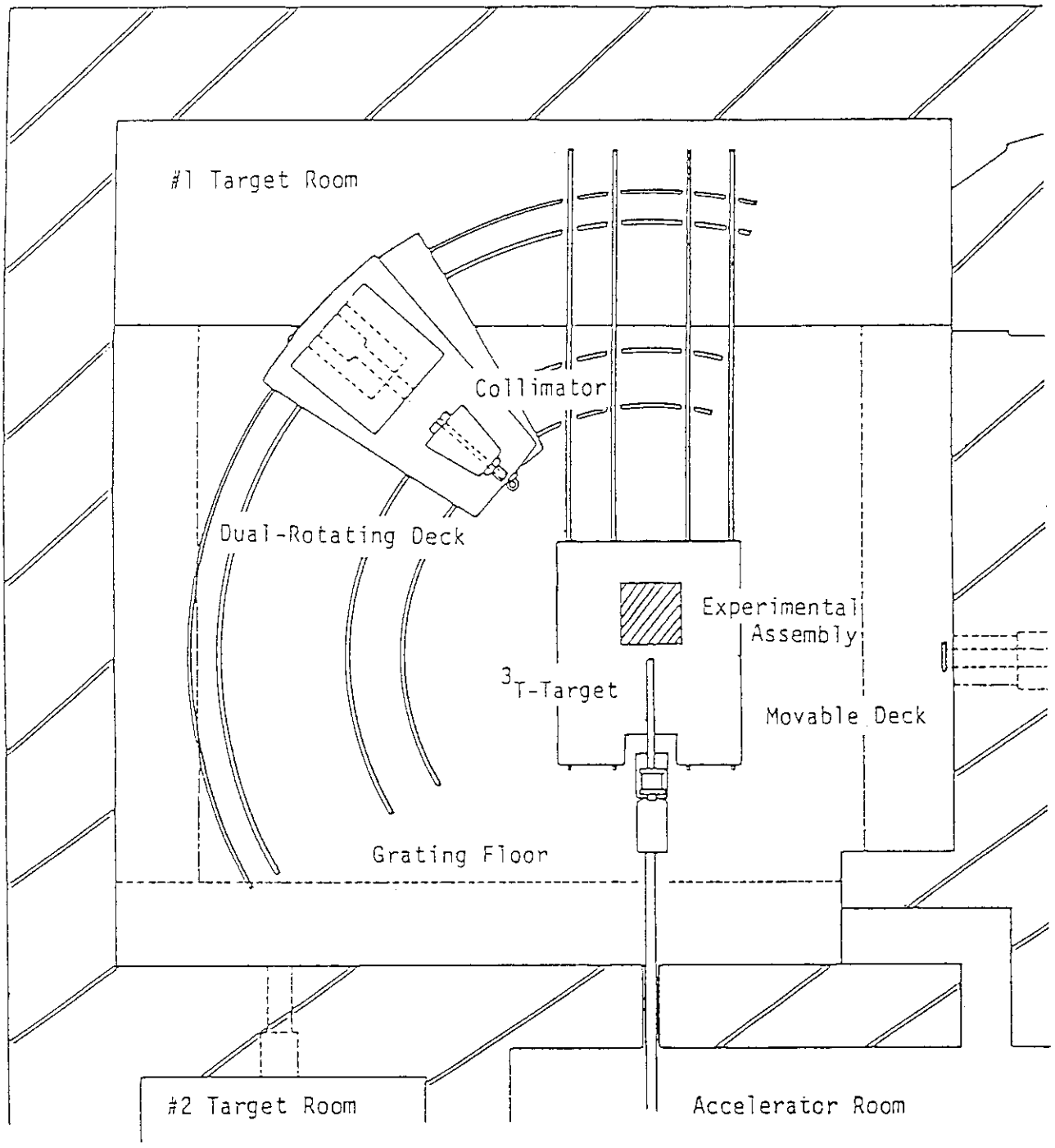


Fig. 3.3 Layout of FNS first target room.

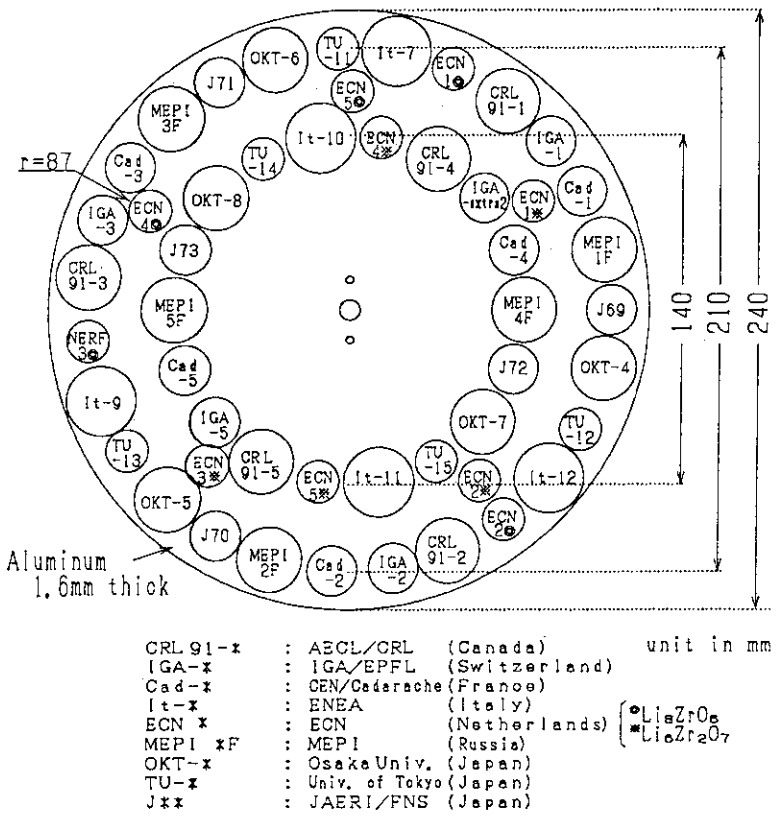


Fig. 3.4 Rotating disk and sample arrangement for FNS irradiation experiment.

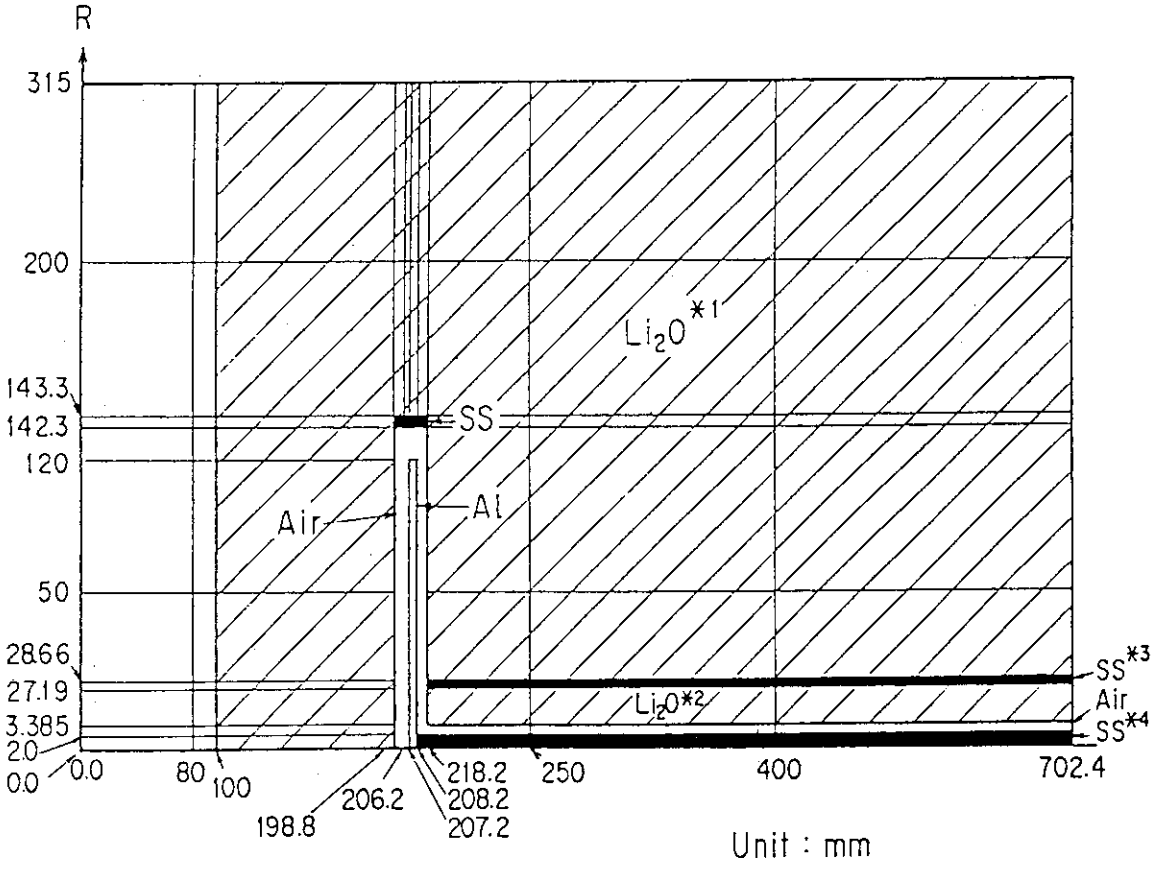


Fig. 3.5 Calculational model of pre-analysis for FNS irradiation experiment.

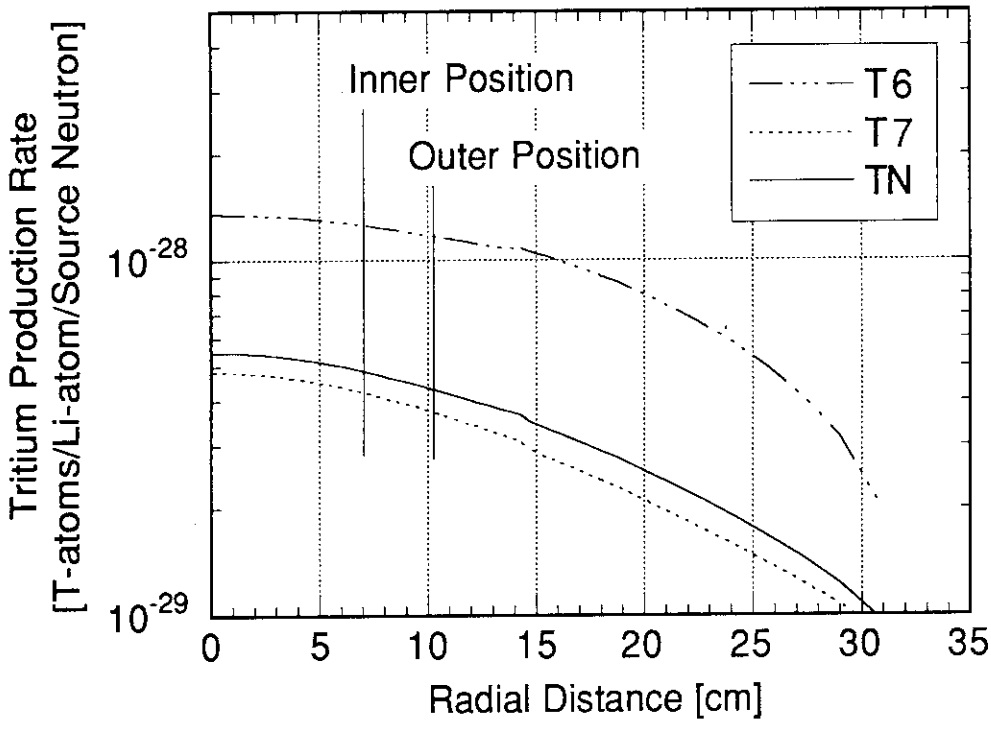


Fig. 3.6 Calculated tritium production rate distribution for radial direction.

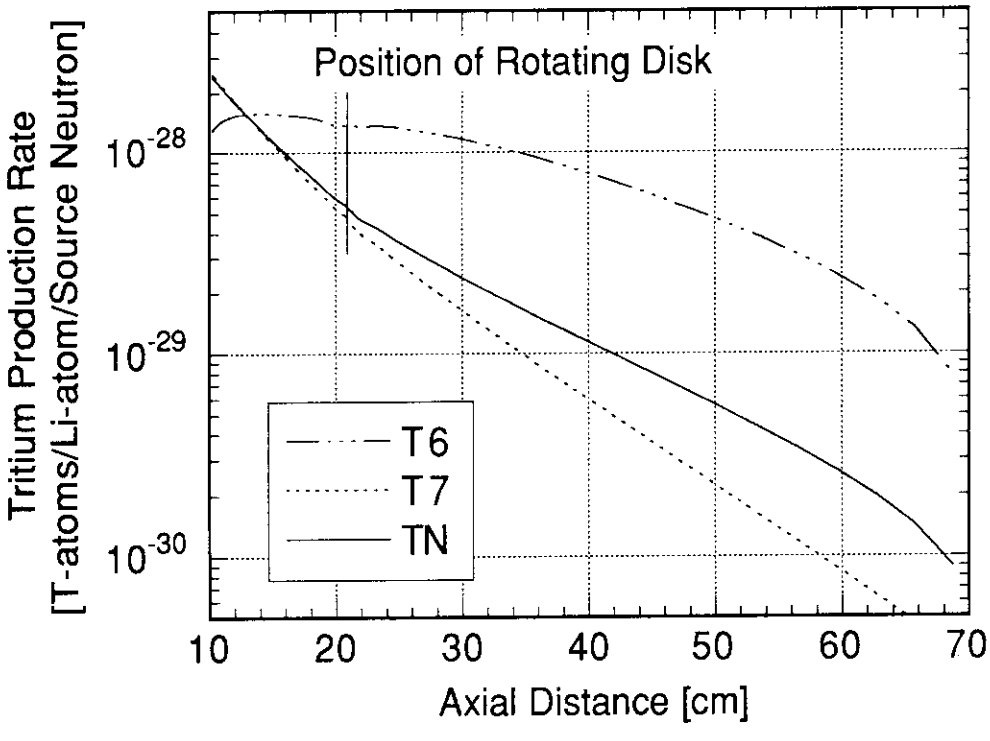
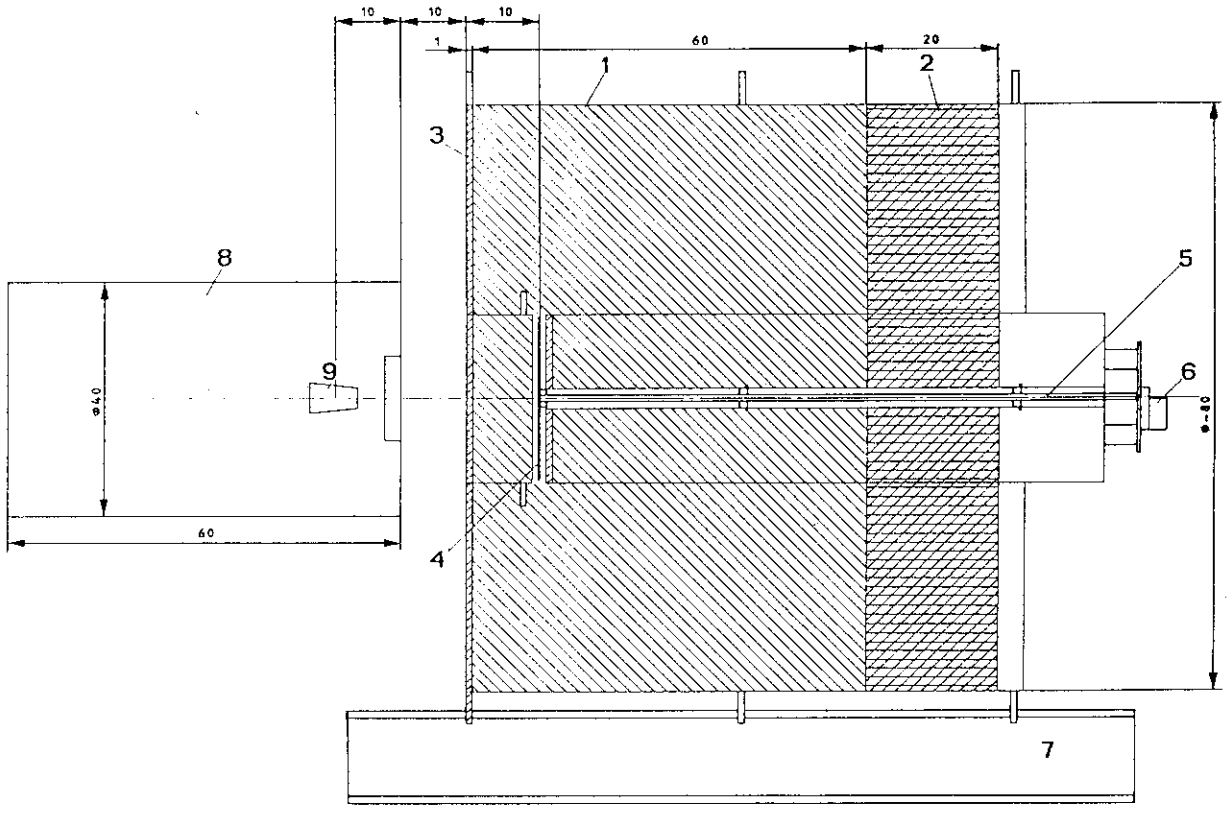


Fig. 3.7 Calculated tritium production for axial direction



1 Natural lithium zone (tritium breeding zone)
2 Graphite zone (reflector)
3 Stainless steel zone (first wall)
4 Samples holder (rotating disk)
5 Transmission rod
6 Synchrous motor and reducer (1rpm)
7 Blanket support
8 Neutron generator (sealed tube)
9 Target (neutrons output device)

Fig. 4.1 Experimental arrangement at LOTUS (Neutron tube and lithium module).

1: high voltage connector
2: target
3: mineral oil insulation
4: cooling water inlet
5: concentric ions beams
6: magnetic coils
7: glas insulator
8: void chamber

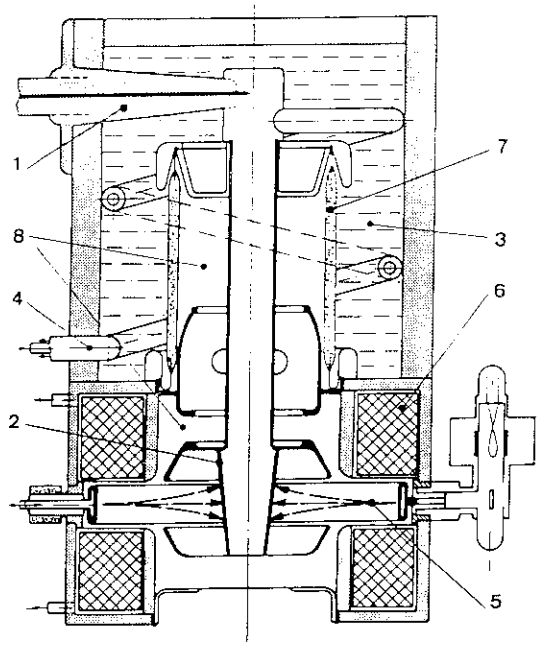


Fig. 4.2 Sectional view of neutron tube.

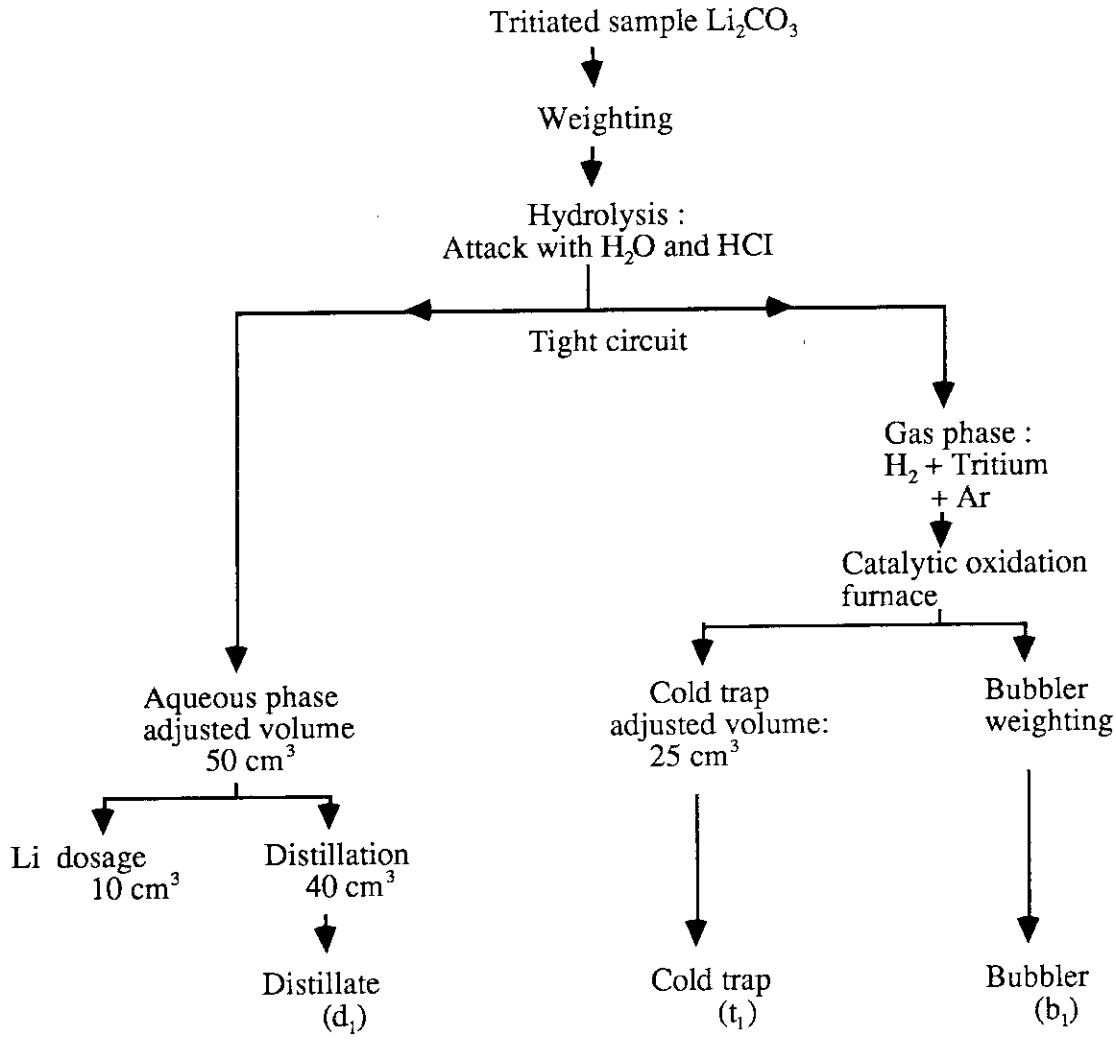


Fig. 5.1 Flow of chemical treatment by CEA.

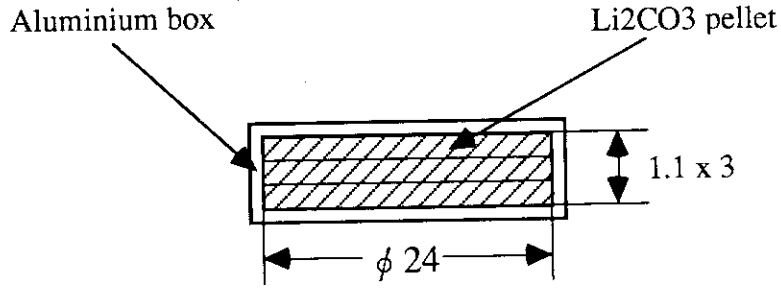


Fig. 5.2 Sectional view of sample pack of MEPI (unit in mm).

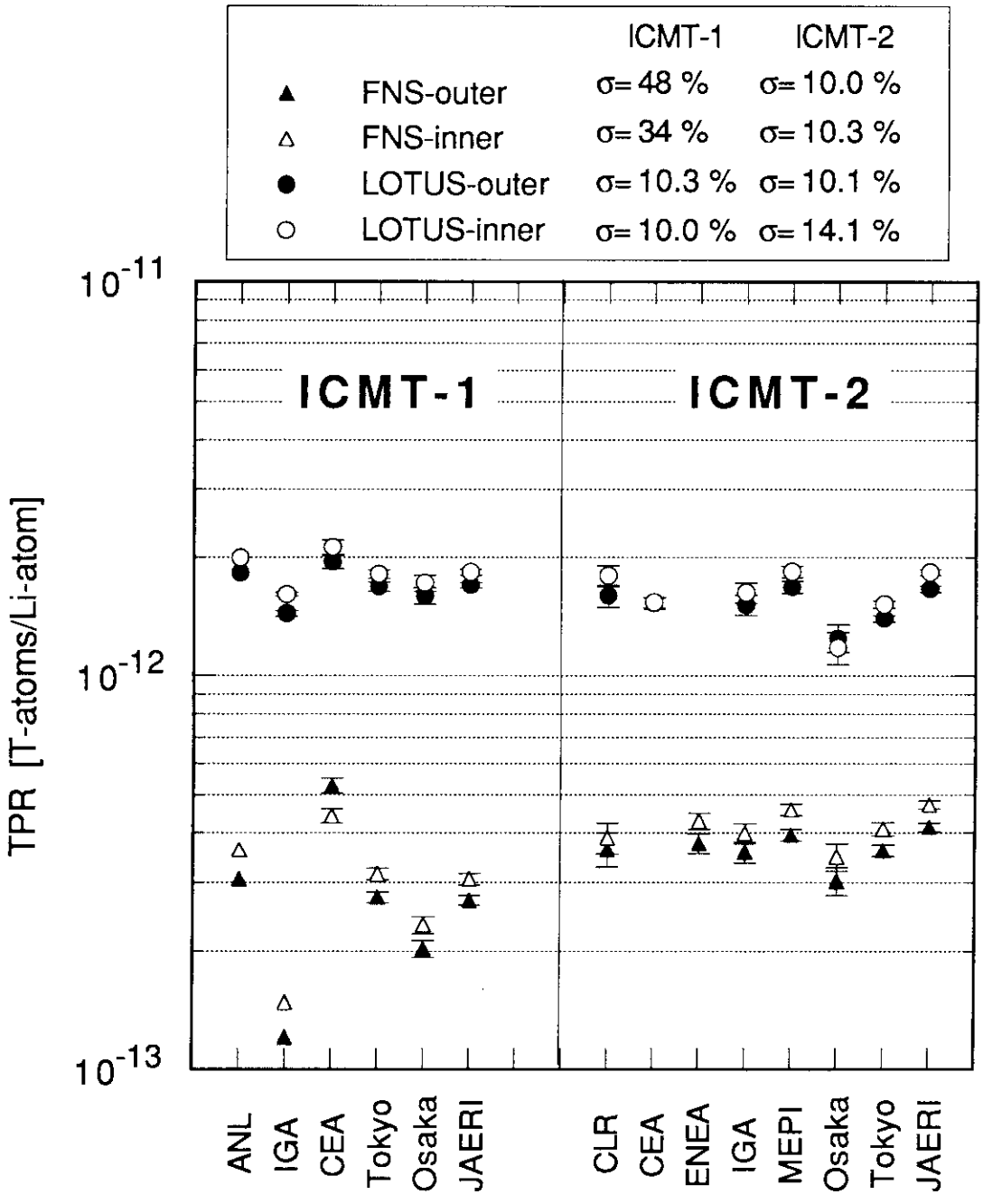


Fig. 6.1 Measured TPRs for the ICMT-1 and ICMT-2 programs.

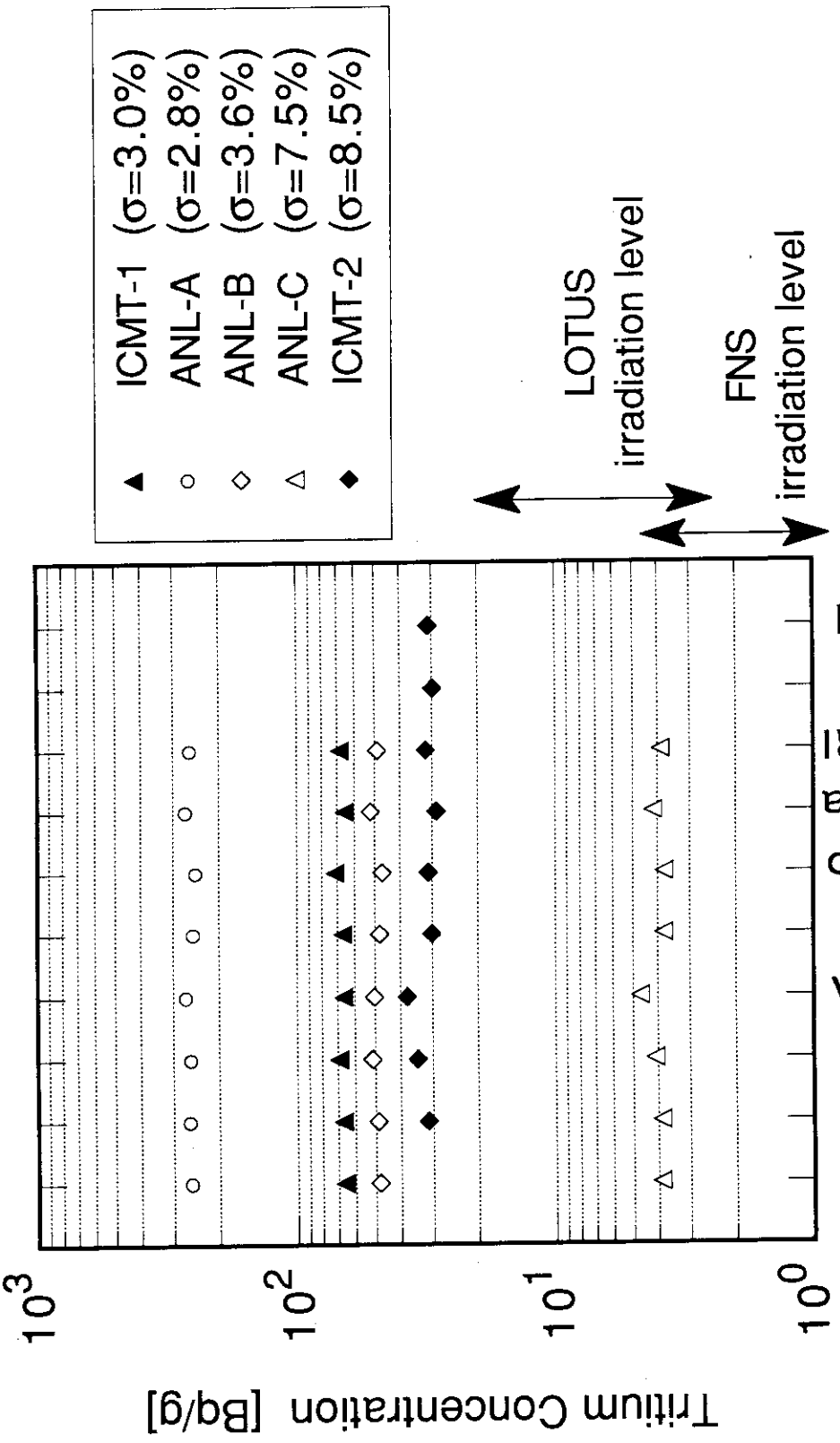


Fig. 6.2 Measured tritium concentration in blind samples for the ICMT-1 and ICMT-2 irradiations, and ANL samples.

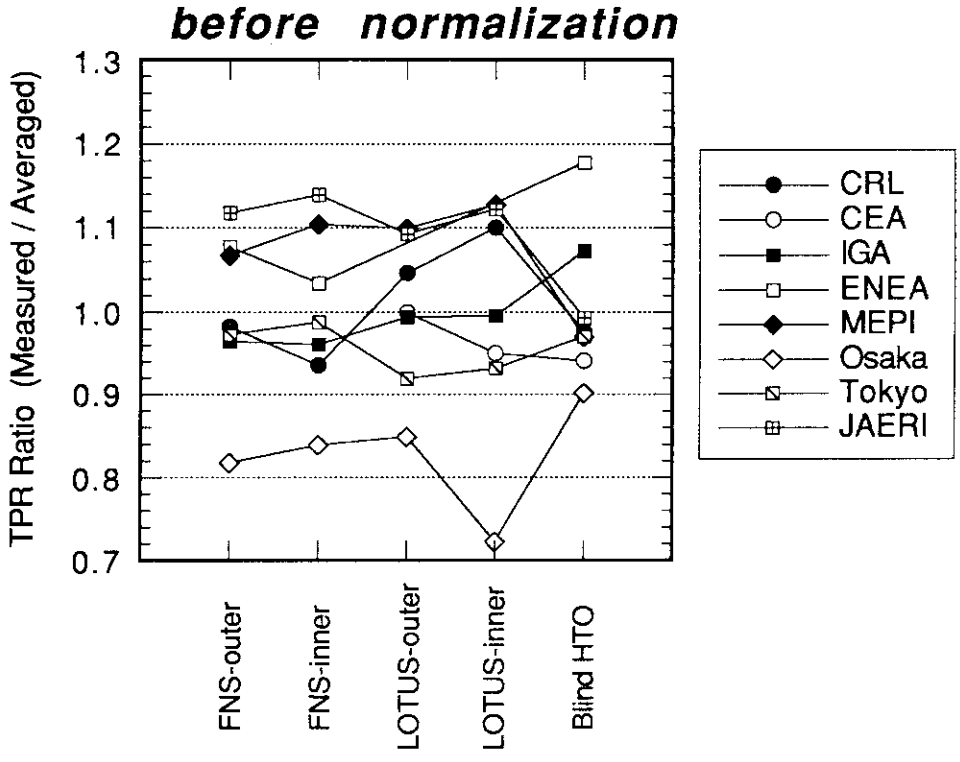


Fig. 6.3 Ratios of measured to averaged TPRs before normalization by the results of blind HOT.

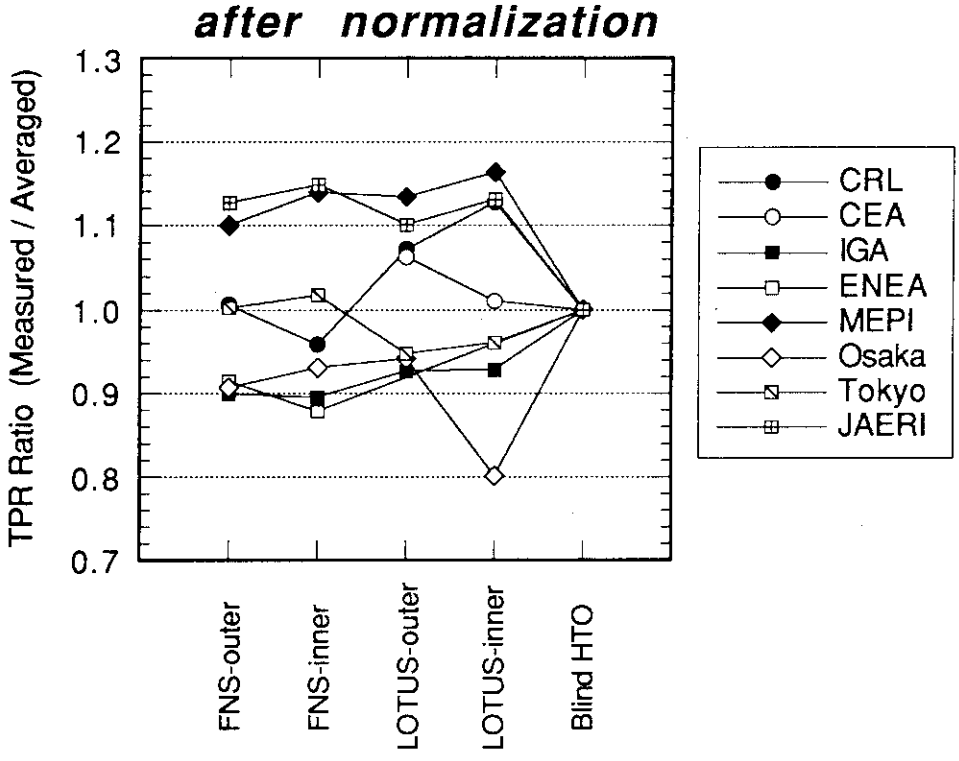


Fig. 6.4 Ratios of measured to averaged TPRs after normalization by the results of blind HTO.

Appendix Comment from Participants

A.1 Comment from Dr. J. M. Miller (AECL/CRL)

Firstly, you have carried out a very detailed analysis of the measurement data provided by the contributors to the measurement program and I support your general conclusions on the results of the international comparisons studies. However, I believe there is scope for obtaining the accuracy of 5% for the TPR measurement and this could be best achieved by having one organization focus on method development and understanding their systematic errors with periodic checks with other organizations with at least the same caliber of technique and with international tritium standards.

As you correctly point out in the text, errors were calculated in various ways by the contributors. To really analyze the absolute values of the errors, it is necessary to review the raw data and determine the errors in a consistent manner. The largest single error in the tritium measurement is that due to the counting statistics. Even in the 1 ~ 25 Bq/g range, the counting error (tritium measurement error) can be reduced to a few percent (2 ~ 3 %) by ensuring that sufficient counts are obtained on the sample. This can be done by increasing the amount of sample, the counting time or reducing the counts of the blank, or as in the case of the TPR measurement, of increasing the tritium concentration in the extraction sample. I am not sure that this requirement (i.e., as low a measurement error as possible) was appreciated by the various contributors. If I examine our data, the error (2 sigma) was reduced from ~10% in FNS to ~ 6% in LOTUS simply because of the larger cpm's obtained from the LOTUS samples. The contribution of systematic errors introduced in the processing of the irradiated pellet must also be considered in the accuracy of the TPR measurement, as you point out. Again, this may be best achieved by having one or two groups focus on the techniques and their contribution to errors. Your target accuracy of 5 % may still be achievable with this type of more focused approach.

A.2 Comment from Dr. O. P. Joneja (IGA)

- It would be important to confirm the details of the procedures adopted for blind sample counting, because the agreement between the different laboratories is extremely poor. This might through some light on the reasons for large differences.

- It should be noted that out of the four participants who have employed bigger size probes, three have finally submitted the results as shown in **Fig. 6.3** (Osaka Univ., MEPI and CRL). The results of these participants exhibit large discrepancies in comparison to the others who have used small probes (with the exception of JAERI). This need to be highlighted and if possible supported by some calculations that due to such difference in sizes/volumes, the tritium production could be different or otherwise.

- I am not quite sure because the blind activity level is quite different from the activity level of the standard probes employed at different laboratories, therefore by simply normalizing the counting rate of the blind sample would not exclude the influence of the HTO standard used.

- It should be stated that one requires higher accuracies for tritium concentrations below a few tens of Bq/g to achieve an accuracy of 5 % on the measured TPR values.