MEASUREMENT AND ANALYSIS OF NEUTRON AND GAMMA-RAY DOSES ON CRITICALITY ACCIDENTS OF LOW-ENRICHED URANYL NITRATE SOLUTION USING TISSUE-EQUIVALENT DOSIMETERS AT THE TRACY FACILITY

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ABSTRACT

Dosimetry data on hypothetical solution criticality accidents are necessary for safety design and assessment of nuclear fuel reprocessing plants. To obtain the data on low-enriched uranyl nitrate solution which is treated in the commercial reprocessing plants adopting the PUREX method, a series of dosimetry experiments was initiated using the Transient Experiment Critical Facility (TRACY) at the Japan Atomic Energy Research Institute in 1998. In the experiments, two kinds of tissue-equivalent dosimeters were used: a polymer-alanine dosimeter and a thermo luminescence detector made of depressed lithium tetra borate. The sensitivity of these dosimeters to neutron and/or gamma-ray is equivalent to that of muscle tissue.

In the experiments, neutron and/or gamma-ray absorbed doses in human bodies were measured under various transient conditions. The dosimeters showed excellent linearity between dose level and released energy, and were consequently applicable to dose measurements on the criticality accidents. In addition, the spatial distribution of dose per fission is insensitive to the transient conditions such as inserted excess reactivity and reactivity insertion rate.

For the analyses of the experiments, two continuous-energy Monte Carlo codes were used: the MCNP version 4B and the MVP. In comparison with the measurements, each code could calculate both neutron and gamma-ray absorbed doses within 30 % in spite of a simple and static model.

1. INTRODUCTION

For safety design and assessment of nuclear fuel reprocessing plants, dosimetry data are required on hypothetical solution criticality accidents. Such data have been provided using solution-fueled supercritical facilities, e.g., the SHEBA at the Los Alamos National Laboratory (LANL) in the USA,[1] the CRAC and the SILENE at the Institut de Protection et de Sûreté Nucléaire (IPSN) in France.[2] However, no experimental dosimetry data have been reported on the criticality accidents of low-enriched uranyl nitrate solution which is treated in the commercial reprocessing plants adopting the PUREX method. To obtain the data, a series of dosimetry experiments was initiated using the Transient Experiment Critical Facility (TRACY)[3] at the Japan Atomic Energy Research Institute (JAERI) in 1998.

Activation foils or thermo-luminescence detectors (TLDs) have been frequently used in such dosimetry experiments. The sensitivity correction of the dosimeters is, however, required to evaluate absorbed doses in human bodies, since most of their materials are not equivalent to the body. Hence two kinds of tissue-equivalent dosimeters, a polymer-alanine dosimeter (alanine dosimeter)[4] and a TLD made of depressed lithium tetra borate (DLTB dosimeter),[5] were used in the experiments at TRACY.

This report describes measurement of neutron and/or gamma-ray absorbed doses on the solution criticality accidents, applicability of the tissue-equivalent dosimeters, and computational analyses of the experiments.

2. EXPERIMENT

2.1 FACILITY AND CONDITIONS

TRACY is a solution-fueled supercritical facilities. The fuel is 9.97-wt%-enriched uranyl nitrate aqueous solution. The shape of the core tank is annular and made of austenitic stainless steel. The dimensions are 200 cm in height, 50 cm in outer diameter and 7.6 cm in inner diameter, respectively. A transient rod is vertically driven in the central guide tube to insert excess reactivity up to 3 \$. This rod consists of natural B_4C pellets.

The dosimetry experiments included five transient operations initiated by withdrawal of the transient rod. The transient conditions were varied in inserted excess reactivity, reactivity insertion rate and total released energy. The conditions of the fuel and the transient operations are summarized in Tables I and II, respectively. Figures 1 and 2 show power profiles during the transient operations.

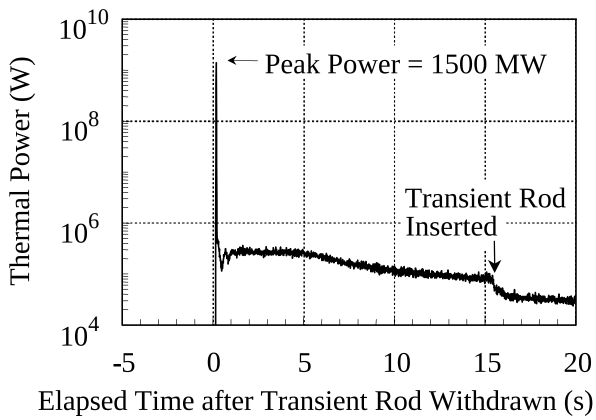
Table I. Conditions of Fuel

Operation No.	Uranium concentration at 25 ^{o}C	Free nitric acid molarity at 25 ^{o}C	Solution density at 25 ^{o}C
R104	395.3 gU/ℓ	0.70 mol/ℓ	1.5502 g/cm^3
R105	399.0 gU/ℓ	0.71 mol/ℓ	1.5557 g/cm^3
R106	399.4 gU/ℓ	0.69 mol/ℓ	1.5562 g/cm^3
R107	388.8 gU/ℓ	0.70 mol/ℓ	1.5425 g/cm^3
R108	389.4 gU/ℓ	0.69 mol/ℓ	1.5431 g/cm^3

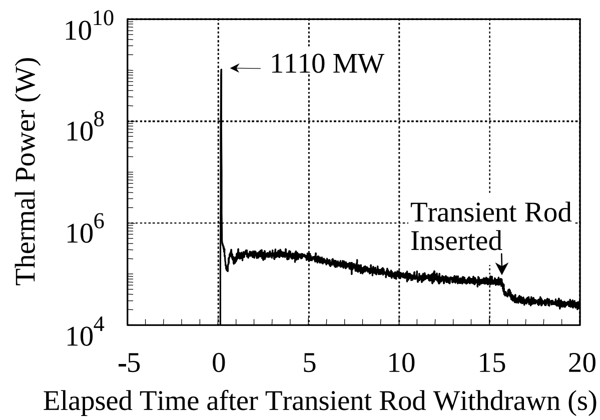
Table II. Conditions of Transient Operations

Operation No.	Withdrawal of transient rod	Inserted reactivity worth	Reactivity insertion rate	Total released energy	Solution level**	Solution temperature**
R104	Rapid withdrawal	2.91 $	Step	20.72 MJ	59.48 cm	25.4 °C
R105		2.60 $	Step	17.15 MJ	57.76 cm	25.2 °C
R106		1.80 $	Step	9.73 MJ	54.72 cm	25.4 °C
R107	Ramp withdrawal	2.96 $	23.8 cent/s*	21.42 MJ	62.27 cm	25.6 °C
R108		2.95 $	69.3 cent/s*	8.68 MJ	62.09 cm	25.5 °C

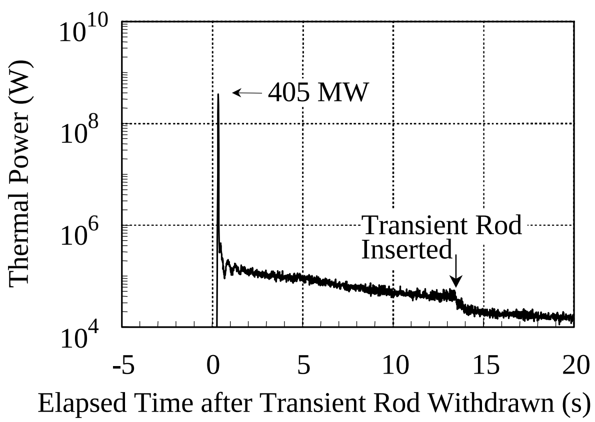
* average values
** values at the beginning of transient operations



(a) R104

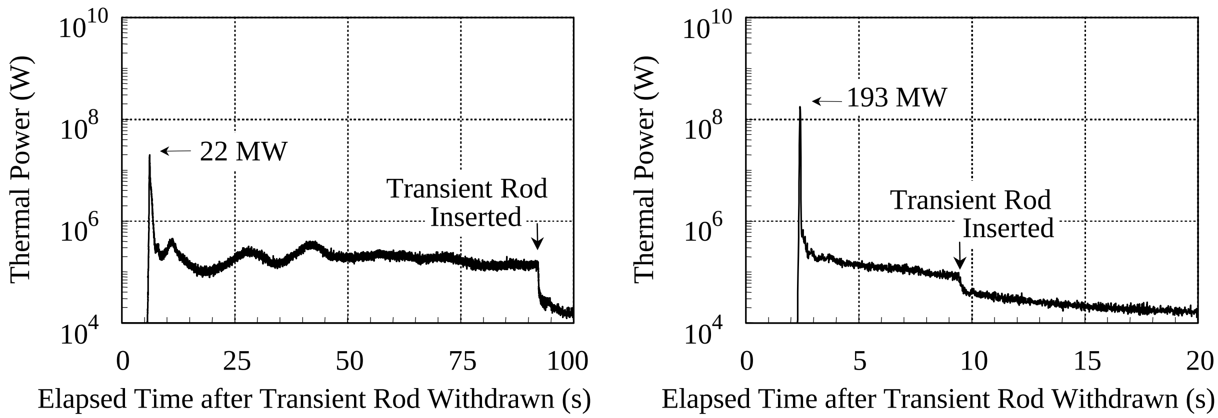


(b) R105



(c) R106

Fig. 1. Power Profiles during Transient Operations Initiated by Rapid Withdrawal of Transient Rod



(a) R107 (b) R108

Fig. 2. Power Profiles during Transient Operations Initiated by Ramp Withdrawal of Transient Rod

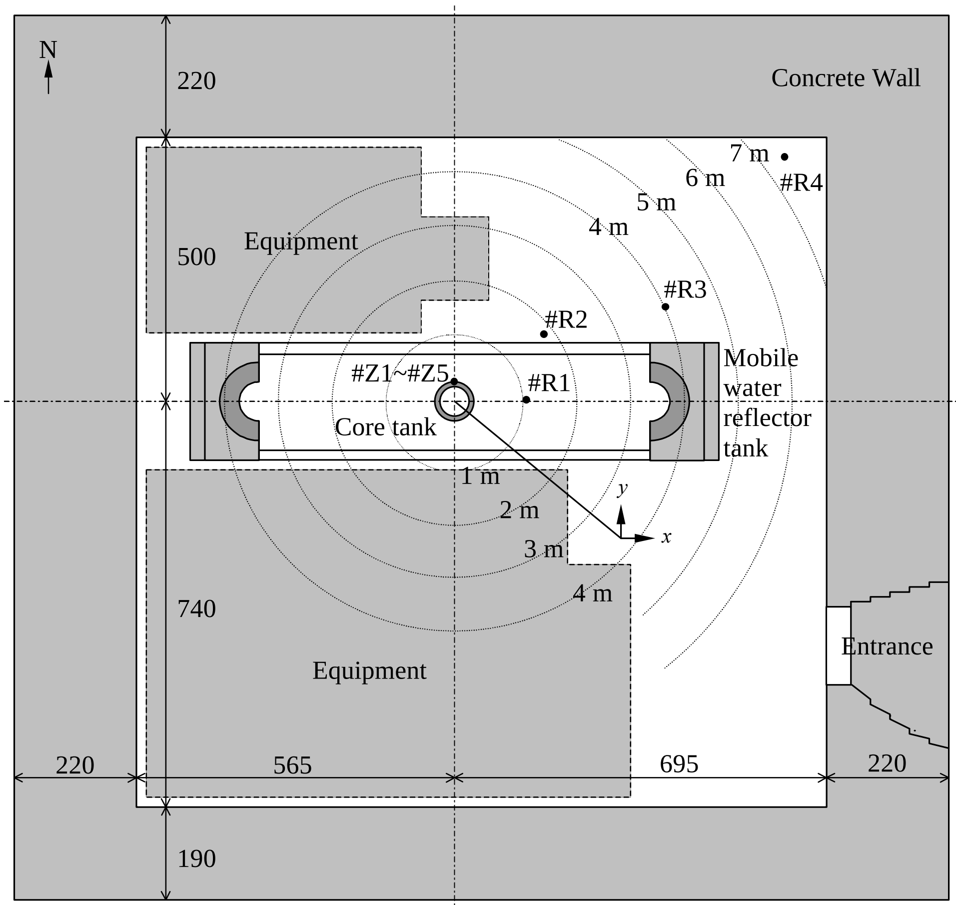
2.2 DOSIMETERS

A set of two tissue-equivalent dosimeters containing alanine or DLTB was used to measure neutron and/or gamma-ray absorbed doses in human bodies. These dosimeters are shown in Fig. 3. These dosimeters are almost equivalent to muscle tissue in the sensitivity to radiations. Crystalline alanine—$C_3H_5O_2NH_2$— is sensitive to neutron, gamma-ray and any other radiation. On the other hand, DLTB—$^7Li_2{}^{11}B_4O_7$— is insensitive to neutron. Moreover, DLTB equals well alanine in the sensitivity to gamma-ray because of the similar effective atomic number. It would be consequently possible to measure respective neutron and gamma-ray absorbed doses by using a set of the dosimeters.

Alanine dosimeter DLTB dosimeter

Fig. 3. Alanine and DLTB Dosimeters

Several sets of the dosimeters were positioned vertically on the surface of the core tank and horizontally at different distances in the reactor room as shown in Fig. 4.



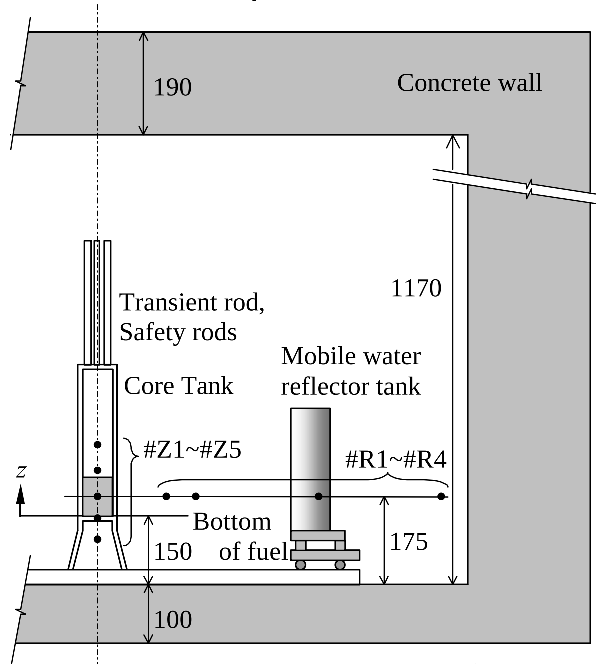
Horizontal layout

• #Z or R : Measuring point

Coordinates of measuring points (cm)

ID	*r* *	*x*	*y*	*z*
#Z1	0	0	+26.1	-30
#Z2	0	0	+26.1	0
#Z3	0	0	+26.1	+25
#Z4	0	0	+26.1	+50
#Z5	0	0	+26.1	+80
#R1	100	+126	0	+25
#R2	189	+171	+128	+25
#R3	403	+385	+188	+25
#R4	743	+615	+462	+25

* distance from the surface of the core tank



Vertical layout

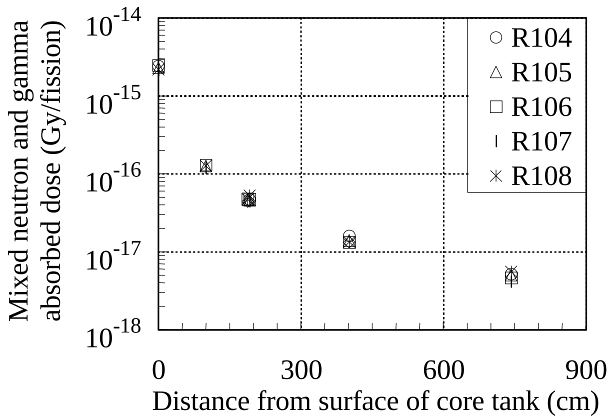
(Unit : cm)

Fig. 4. Arrangement of Dosimeters

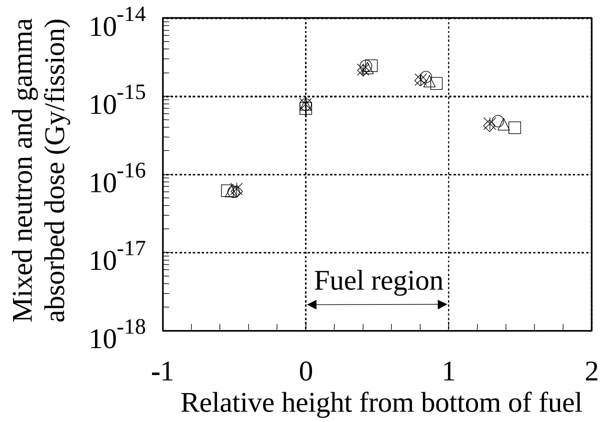
2.3 RESULTS OF MEASUREMENT

Figure 5 shows the spatial distribution of absorbed doses per fission in the alanine and the DLTB dosimeters. Since the solution level was different among the operations, the x-axis in the vertical distribution represented relative positions: zero indicated the bottom of fuel, and unity the top.

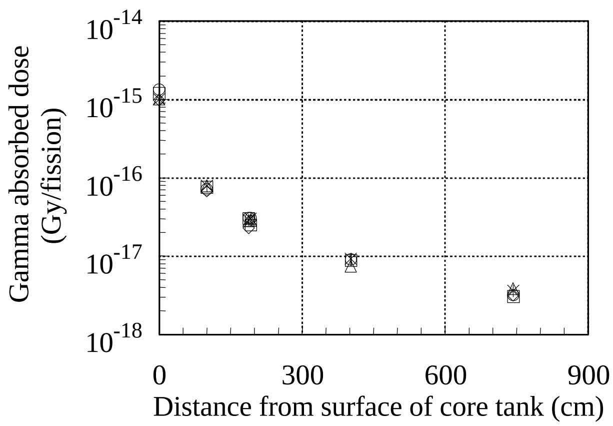
As shown in Fig. 5, the distribution profiles became the same shape in spite of different transient conditions. This result also shows that the dosimeters have excellent linearity between dose level and released energy up to 21 MJ (approximately 7×10^{17} fissions). Thereupon the following prospects are suggested. The distribution of absorbed doses is insensitive to the transient conditions, and the doses are proportional to the number of fissions. In other words, the number of fissions on a criticality accident could be estimated from dose level measured with at least one calibrated dosimeter, even if its transient condition were unknown.



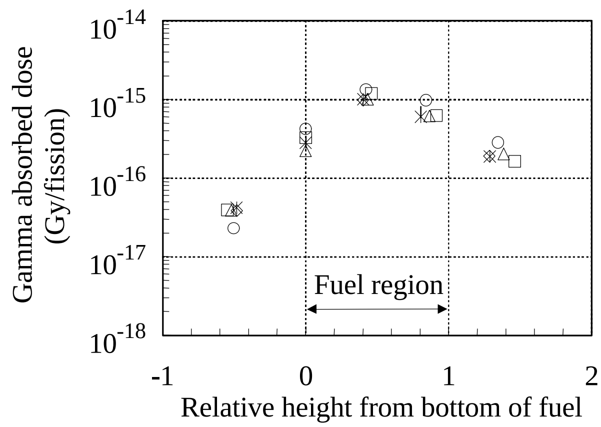
(a) Horizontal distribution of sum of neutron and gamma-ray absorbed doses in alanine dosimeter



(b) Vertical distribution of sum of neutron and gamma-ray absorbed doses in alanine dosimeter



(c) Horizontal distribution of gamma-ray absorbed dose in DLTB dosimeter



(d) Vertical distribution of gamma-ray absorbed dose in DLTB dosimeter

Fig. 5. Spatial Distribution of Neutron and Gamma-ray Absorbed Doses

3. ANALYSIS

3.1 COMPUTATIONAL CODES AND CONDITIONS

Neutron and gamma-ray absorbed doses were calculated with two continuous-energy Monte Carlo codes, the MCNP version 4B (MCNP4B) [6] and the MVP (Ref. 7, 8). The cross-section libraries for neutron and photon were the Japanese evaluated nuclear data library JENDL version 3.2 (Ref. 9) and the photon interaction table for MCNP,[6] respectively. The kerma approximation was adopted in the calculations to convert fluences into absorbed doses. The kerma factors for neutron and photon were cited from the reports by Caswell et al.[10] and Hubbell,[11] respectively.

Everything except the core tank, fuel solution and concrete surroundings was disregarded in the geometrical system for practical modeling. To calculate vertical dose distribution, 5-cm-wide and 1-mm-thick rings were coiled on the surface of the core tank for flux tallies of the track-length estimator. For horizontal distribution, the point-detector estimators (for MCNP4B) or 30-cm-diameter-spheres of the track-length estimator (for MVP, in which the point-detector estimator was not available) were positioned at the measuring points.

The calculations were made as an eigenvalue problem of neutron and photon transportation. The effective number of neutron histories was 1 million. The neutron and gamma-ray absorbed doses per fission were also calculated using the number of fissions simultaneously done.

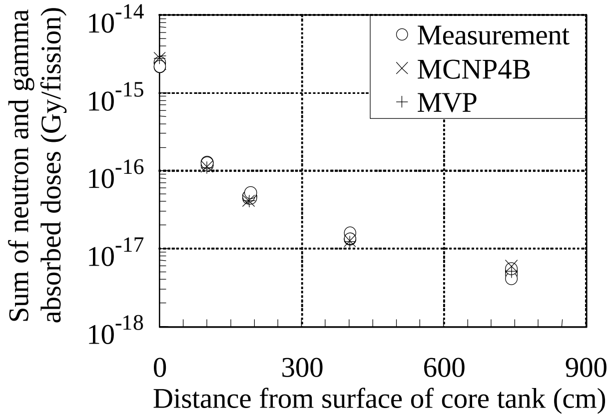
3.2 MODELING OF SOLUTION DURING POWER EXCURSION

In the case of criticality accidents of solution fuel, a power excursion is accompanied with not only rapid rise of solution temperature but also variation of solution density due to radiolytic gas void. It is, however, extremely difficult to measure such data during the accidents. In this report, neutron and gamma-ray absorbed doses were calculated under the static conditions at the beginning of the transient operations.

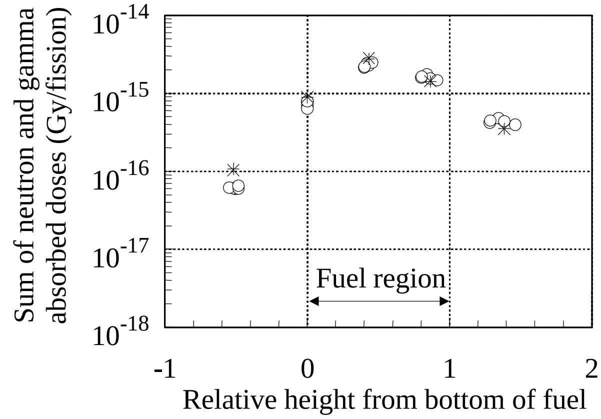
3.3 COMPARISON WITH MEASUREMENT

Figure 6 shows the comparison between the measurements and the calculations on the distribution of dose per fission. The calculations were represented by the case of R105 operation, since there was little difference in the calculated doses per fission among the operations.

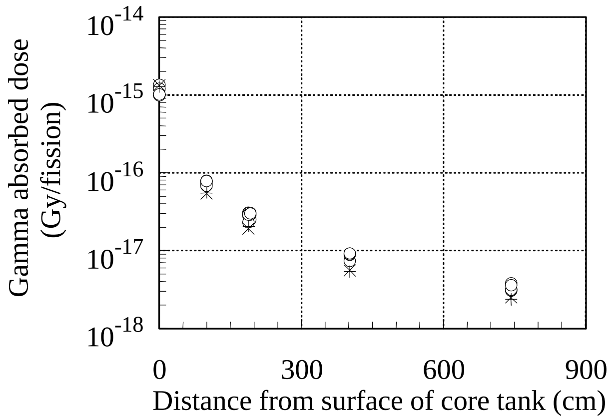
As shown in the vertical distribution in Fig. 6, the neutron and gamma-ray doses calculated with MCNP4B or MVP agreed well with the measurements. On the contrary, the calculations in the horizontal distribution were slightly underestimated at the positions off the core tank. However, the discrepancy between the measurements and the calculations was not more than 30 %. This discrepancy would be minimized by detailed modeling of the geometry system and time-dependent treatment of solution behavior during power excursions.



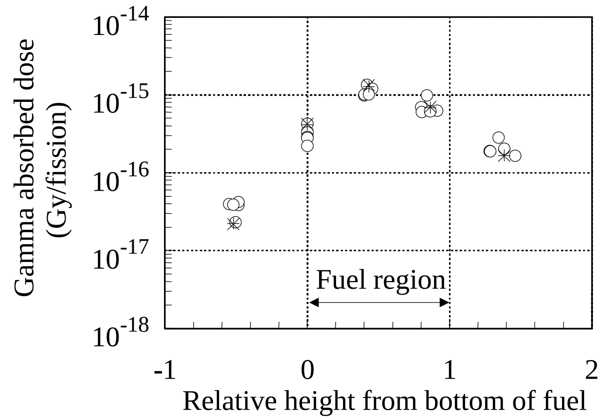
(a) Horizontal distribution of sum of neutron and gamma-ray absorbed doses in alanine dosimeter



(b) Vertical distribution of sum of neutron and Gamma-ray absorbed doses in alanine dosimeter



(c) Horizontal distribution of gamma-ray absorbed dose in DLTB dosimeter



(d) Vertical distribution of gamma-ray absorbed dose in DLTB dosimeter

Fig. 6. Comparison between Measurements and Calculations in Dose Distribution

CONCLUSIONS

A series of dosimetry experiments was initiated using TRACY in 1998 to obtain dosimetry data on hypothetical criticality accidents of low-enriched uranyl nitrate solution. In the experiments, an alanine and a DLTB dosimeters were used to measure neutron and/or gamma-ray absorbed doses in human bodies.

As the result of the experiments, the dosimeters showed excellent linearity between dose level and released energy. Moreover, spatial distribution of dose per fission was insensitive to transient conditions such as inserted excess reactivity and reactivity insertion rate. Thus the number of fissions could be estimated from the dose level on the criticality accident, even if its transient condition were unknown.

In comparison with the measurements, both MCNP4B and MVP calculated the neutron and gamma-ray absorbed doses with accuracy enough to estimate exposure risks in spite of a simple and static model.

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