Critical Experiments of Basic Heterogeneous Core Composed of LWR Fuel Rod Array and Low Enriched Uranyl Nitrate Solution

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A series of critical experiments in STACY was performed for the purpose of obtaining the benchmark data which are concerned to the dissolver of the reprocessing facility for LWR spent fuel.

A basic heterogeneous core was constituted in a cylindrical core tank, by arranging an assembly of 5 wt% enriched UO_2 fuel rods in the 6 wt% enriched uranyl-nitrate-solution. The fuel rod had basically the same dimension as that of PWR. The volume ratio of the solution to the pellet in the unit cell was 7.0 and the lattice pitch was 2.1 cm. The uranium concentrations of the uranyl-nitrate solution was adjusted to 363-51g/L, and the critical solution heights of 41-66cm were measured for both the unreflected and the water-reflected conditions.

Sensitivity analysis of effective-neutron-multiplication-factors (k_{eff}'s) for the uncertainty of experiment conditions was performed using SRAC and two-dimensional transport calculation code TWODANT. The effect of the measurement errors on k_{eff} was evaluated to be about 0.1 %Δk_{eff}. The benchmark calculations of k_{eff}'s were also carried out using Monte Carlo code MCNP4B with nuclear data library, JENDL-3.2. The eleven critical configurations are judged to be acceptable as the benchmark data.

KEYWORDS: STACY, Critical Experiment, Heterogeneous Core, Low Enriched, Dissolver, Uranyl-Nitrate, Unreflected, Water-reflected, MCNP4B, JENDL3.2

1. Introduction

The critical experiments of basic heterogeneous STACY core started in 2002 for the purpose of obtaining the benchmark data concerning the dissolver of the reprocessing facility for LWR spent fuel.[1) The core composed of LWR-type fuel rod array and low-enriched uranyl-nitrate-solution.

Before the STACY experiments, very few kinds of critical experiments had been reported for the heterogeneous cores in which fuel rod array and fuel solution were intermixed simulating the dissolver of reprocessing facility. One was for the uranium fuel configuration, and the other was for the FBR fuel. However, it was found that a mismatching of data existed in the former, and the latter was far different from the reprocessing conditions of the LWR spent fuel due to the high enrichment of plutonium in the MOX fuel. So, it was regarded that they were not sufficient for the benchmark data, and it was a subject to acquire new benchmark experiments.

Based on the above background, the heterogeneous STACY core was constituted for the purpose of clarifying the limiting value of the k_{eff} used for the criticality safety control of the dissolver. A series of critical experiments was performed, by changing uranium concentrations parametrically for both unreflected and water-reflected conditions.

The measurement error is a factor which influences the safety margin for determining the limiting value of k_{eff}. The uncertainties of the measured values such as dimensions, compositions etc. were evaluated in detail, and the effects of the measurement errors on k_{eff} were evaluated using SRAC[2)] and two dimensional transport calculation code TWODANT. Benchmark calculations of k_{eff}'s for both detailed- and benchmark-model were also carried out using Monte Carlo code MCNP4B[3)] with JENDL-3.2.

2. Description of Experimental Configuration

2.1 Criticality Facility

The STACY facility consisted of the core tank containing fuel solution or fuel rod array filled by fuel solution, a solution transfer system, a fuel treatment system, and a fuel storage system. The reactivity was controlled by adjusting the fuel solution level in the core tank. Initially, a fast-feed pump was used to feed the fuel solution to just below half of the predicted critical height. After that, a slow feed pump was used to feed the fuel solution to the near critical state. The maximum excess reactivity and the maximum reactivity addition rate were adjusted by limiting the position of the contact-type level gauge and the feed speed of the slow-feed pump. The level gauge consisted of a needle to detect the surface of solution, an electric motor for changing the vertical

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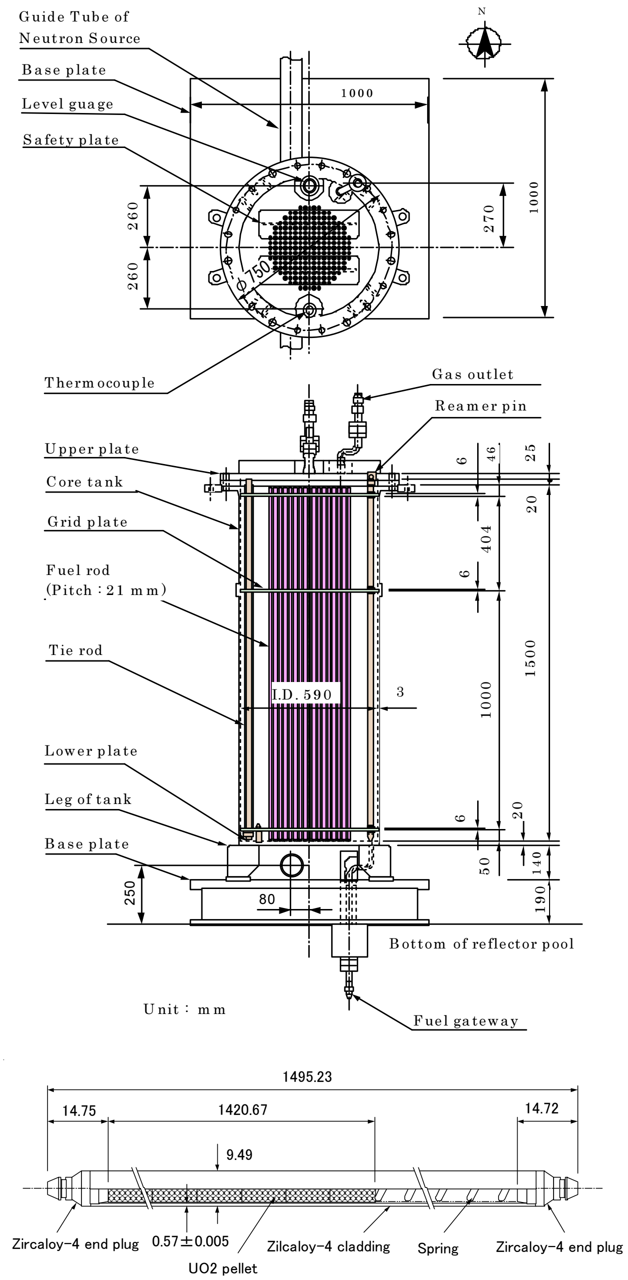

Fig.1 Schematic of the STACY core tank and fuel rod

position of the needle, and an encoder indicating the vertical position. The accuracy of the level gauge was 0.2 mm.

2.2 Core Configuration

The schematic of the STACY core tank and fuel rod are shown in Fig.1. The core tank was made of stainless steel, S.S.304. The basic heterogeneous core was constituted in the core tank with 59 cm inner diameter and 0.3 cm tank thickness, by arranging the assembly of 5 wt% enriched UO_2 fuel rods in the 6 wt% enriched uranyl-nitrate-solution. The assembly was supported with three grid plates held to the grid-plate housing, and the housing was fixed to the inner-side-wall of the core tank. The grid plate was made of zircaloy-4. The core consisted of two radial regions as follows: the assembly of the 221 UO_2 fuel rods with square lattice filled by the uranyl-nitrate solution was arranged in the center of the core tank, and the uranyl-nitrate-solution region surrounded the assembly. The fuel rod had the same dimension as that of PWR as shown in Fig. 1. The outer diameter of the clad made of zircaloy-4 and the diameter of the uranium dioxide pellet were 0.95 cm and 0.82 cm, respectively. The volume ratio of the solution to the pellet in the unit cell was 7.0 and the lattice pitch was 2.1 cm. The effective diameter of the fuel assembly was 35 cm.

Table 1 Dimensions of the STACY core (unit: mm)

Item		Value
(Tank)	Inner diameter	590.1±0.1
	Inner height	1522.5±0.5
	Thickness of side wall	3.2±0.1
	Thickness of lower plate	20.1±0.1
	Thickness of upper plate	105.1±0.1
(Grid plate)	Outer diameter	581.0
	Thickness	6.0±0.1
	Hole diameter	9.91±0.01
	Lattice pitch	21.00±0.01
(UO_2 rod)	Outer diameter	9.49±0.005
	Total length	1495.23±0.1
	Stack length	1420.67±0.1
	Clad thickness	0.57±0.005
	Length of upper end plug	14.72±0.01
	Length of lower end plug	14.75±0.01
	Pellet diameter	8.20±0.005

The core tank was supported by four stainless steel feet, which were fixed on the base plate. Each support was 14 cm-high, and the base plate was 3 cm-thick. The base plate was supported by 16 cm-high stainless steel beams, which were fixed on the bottom of the water-reflector pool tank. The base plate was a square stainless steel with 100 cm length at each side. The neutron source guide tube was set so that the center was 10 cm below the top surface of the bottom plate of the tank. The dimensions of the core tank, grid plate and the fuel rod are summarized in Table 1. The uncertainties include measurement error and accuracy of measurement.

As is shown in Fig.2, the core tank was set in the water-reflector pool tank, which was 202 cm wide, 402 cm long, and 240 cm high. The side walls and the bottom plate of the reflector tank was 33 cm above the bottom of the reflector tank. The shortest distance between the side wall of the core tank and the inner surface of the pool tank was 70 cm. The tank was filled with water up to 15 cm above the top of the core tank for the water-reflected condition.

The arrangement of the neutron detectors is shown in Fig. 2. The positions of the neutron detectors were variable depending on the experimental requirements. The figure shows the arrangement for Run No.176 as an example. Two ^{10}B-lined proportional counters

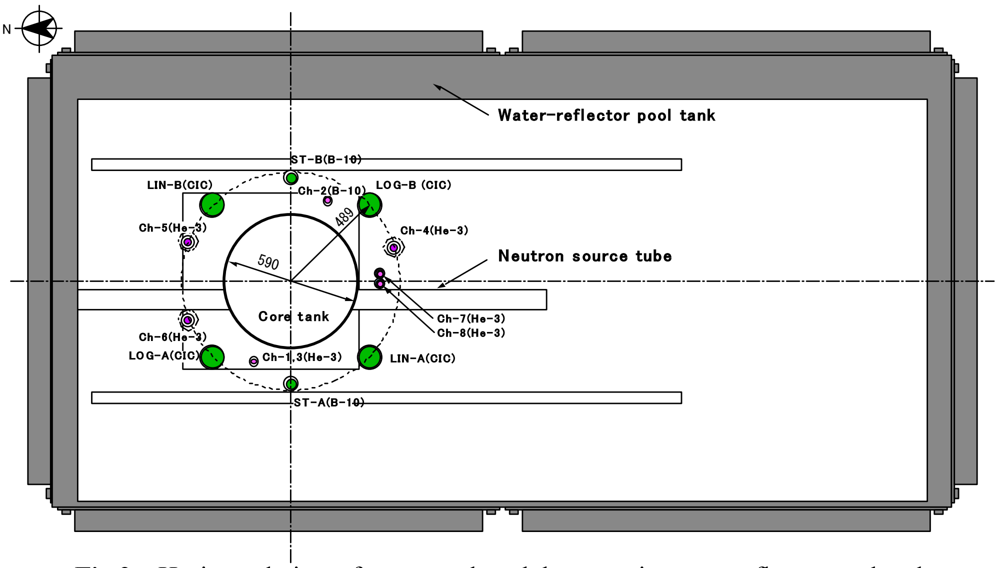

Fig.2 Horizontal view of a core tank and detectors in water-reflector pool tank

(ST-A and B) and four gamma-ray compensated ionization chambers (LIN-A, B, LOG-A and B) were located around the core tank to measure the neutron flux level for the start-up power range and the operational power range, respectively. Maximum power was limited to 200W. Nine additional experimental neutron detectors were also located around the core tank: two ^{3}He proportional counters (Ch-1 and 3), one gamma-ray compensated ionization chamber (Ch-6) used as input to a digital reactivity meter, one ^{3}He proportional counter (Ch-2), one gamma-ray compensated ionization chamber (Ch-5), one ^{3}He proportional counter (Ch-7) and one ^{10}B-lined proportional counter (Ch-4), two ^{3}He proportional counters (Ch-8 and Ch-9). For improving the neutron efficiency, the detectors were covered with polyethylene.

Table 2 Critical condition of STACY

Run No	Reflector condition	Condition of fuel solution at 25 ℃			Solution temp. (℃)	Critical height (cm)
		Uranium Concentration (g/L)	Acid molarity (N)	Solution Density (g/cm3)		
368	Un-reflected	362.6 ±0.7	2.49 ±0.04	1.5605 ±0.0004	24.7	42.55
376		284.5 ±0.7	2.52 ±0.02	1.4591 ±0.0007	24.5	44.35
379		214.2 ±0.6	2.54 ±0.02	1.3678 ±0.0006	24.8	47.43
385		154.5 ±0.4	2.61 ±0.03	1.2901 ±0.0005	24.1	52.07
397		101.8 ±0.6	2.54 ±0.03	1.2174 ±0.0007	24.7	57.78
400		51.2 ±0.5	2.53 ±0.03	1.1495 ±0.0007	24.7	66.39
375	Water-reflected	284.3 ±0.7	2.51 ±0.02	1.4588 ±0.0007	24.5	41.38
380		214.4 ±0.6	2.54 ±0.02	1.3681 ±0.0006	24.6	44.90
386		154.7 ±0.4	2.61 ±0.02	1.2903 ±0.0005	24.4	49.84
398		102.0 ±0.6	2.54 ±0.03	1.2177 ±0.0007	23.6	55.73
401		51.7 ±0.5	2.54 ±0.03	1.1505 ±0.0007	22.9	64.69

2.3 Fuel Material Description

The enrichments of the fuel pellet and fuel solution was were 4.98±0.01 and 6.02±0.01 wt%, respectively.

Uranyl-nitrate-solution was used as the fuel solution, which consisted of uranyl nitrate [$UO_2(NO)_3$], free nitric acid (HNO_3), and water (H_2O). Chemical analyses were carried out about once a week during the series of experiments, and by the interpolation of these analysis data, the uranium concentration, the free nitric acid concentration, and the solution density at the time of each experiment were evaluated. The results of the analysis are shown in Table 2. The temperature of the sample was kept at 25 ^{o}C during the analysis. The uranium concentrations of the uranyl-nitrate-solution were parametrically adjusted to 363-51g/L by successive steps. The free nitric acid was kept at about 2.5 mol/L. The solution density was in the range of 1.5605 –1.1495 g/cm^3**.**

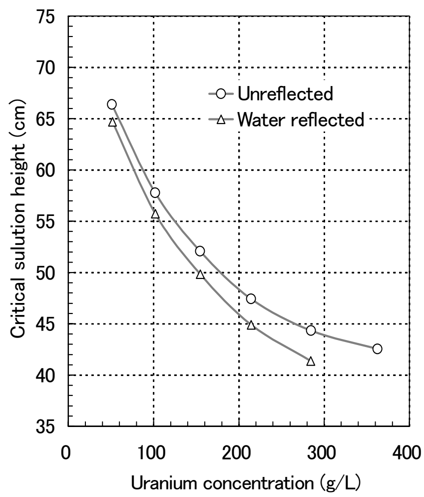

Fig.3 Variation of critical solution height

2.4 Critical Conditions

The eleven critical conditions for both the unreflected and the water-reflected conditions are summarized in Table 2. The critical solution heights of 41-66cm were measured for the uranium concentrations of 363-51g/L. The variation of critical solution heights with the uranium concentration is shown in Fig. 3.

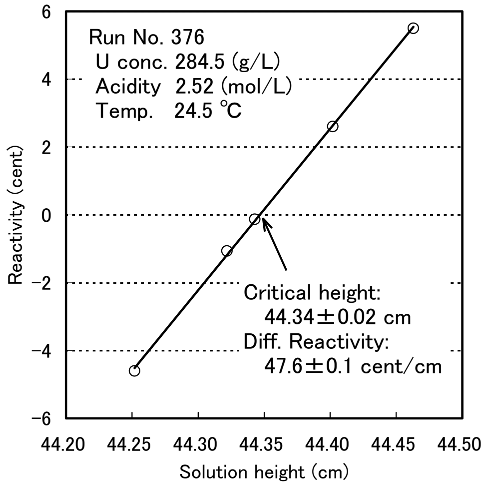

Fig.4 Determinations of critical height and differential reactivity worth (Run376)

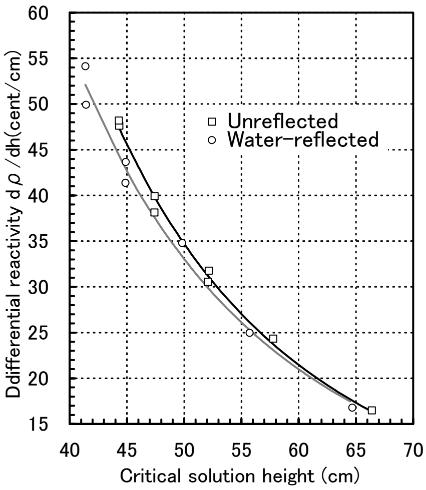

Fig.5 Differential reactivity worth vs. critical solution height

To obtain the critical height, first a critical solution height was confirmed by observing the steady-state neutron flux level. Then the final critical height was determined by a series of reactivity measurements for which the fuel solution was repeatedly drained and fed near the critical state. In the measurements, subcritical and supercritical conditions were repeated, not only subcritical conditions. The reactivities were measured, employing a digital reactivity meter. A digital reactivity meter calculates reactivities by solving the reactor kinetics equation in real time, using an analog signal from a neutron detector. Near the critical state, reactivity ρ is linear with solution height H, and ρ is expressed by

$$\rho = (d\rho / dh) \times (H - H_c) \qquad (1)$$

where, H_c is a critical solution height (cm), and dρ/dh is a differential reactivity at critical height (cent/cm). Figure.4 shows the result for Run No.176 as an example, and dρ/dh and Hc were estimated to be 47.6 cent/cm and 44.34 cm, respectively.

The differential reactivity worth with respect to the solution height was estimated from the measurement. The differential reactivity dρ/dh based on the one group theory is expressed by

$$d\rho / dh = C/(H+\lambda)^3 \qquad (2)$$

where C is a conversion factor (cent-cm^2), λ is an extrapolation length (cm) along the vertical direction, and H is a solution height (cm). The obtained differential reactivity worth for unreflected and water-reflected conditions are shown in Fig. 5. The function (2) was fitted to the measured values with two parameters, C and λ, and their best estimation values were obtained. The values for the water-reflected core are larger than those of the unreflected condition. C and λ for the unreflected condition were estimated to be 662 cent-cm^2 and 7.6 cm, respectively. And these for the water-reflected condition were estimated to be 760 cent-cm^2 and 11.2 cm, respectively.

3. Evaluation of Experimental Data

The effects on k_{eff} of uncertainties in measured data were estimated by sensitivity studies. The sensitivity studies were performed with a two-dimensional transport code, TWODANT, and a 16-energy-group cross section set collapsed from the 107-energy-group SRAC public library based on the evaluated nuclear data library, JENDL-3.2. For the sensitivity studies, a density formula for uranyl-nitrate-solution developed at JAERI was used. This formula gives the density of uranyl-nitrate-solution as a function of uranium concentration, free nitric acid concentration, and solution temperature.[4] Table 3 summarizes the effects on k_{eff} for typical parameters. The sub-total value is given as the square root of the sum of individual effects' squares. The sub-total includes those for other parameters which are not shown in the table. The sum-total was obtained by the square root of the sum of individual sub-totals' squares.

3.1 Uncertainties for each item

As to the fuel solution, the uncertainties of uranium enrichment, uranium concentration, free nitric acid concentration, and solution density were determined to be 0.01 wt.%, 0.7 g/L, 0.04 mol/L, and 0.0007 g/cm^3, respectively. The effects on k_{eff} of uncertainties pertaining to the fuel solution were calculated using these uncertainties as variations, and the results are given in Table 3. The values are shown at the minimum and the maximum uranium concentrations. The main factors to the total uncertainty of k_{eff} are found to be the free-nitric acid concentration, the solution density and the uranium enrichment in order,

Table 3 Summary of effects on k_{eff} for materials and geometrical uncertainties (%Δk_{eff})

Item	Parameter	Uncertainty	U concentration					
			Unreflected			Water-reflected		
			51.2(g/L)	~	362.6(g/L)	51.7(g/L)	~	284.3(g/L)
Fuel solution	U-235 enrichment (wt%)	0.01	0.006	~	0.022	0.007	~	0.022
	U concenration (g/L)	0.7	0.020	~	0.005	0.022	~	0.013
	Acidity (mol/L)	0.04	-0.050	~	-0.061	-0.050	~	-0.056
	Solution density (g/cm^3)	0.0007	0.017	~	0.027	0.016	~	0.022
	Sub total		**0.057**	**~**	**0.070**	**0.057**	**~**	**0.065**
Fuel rod	U-235 enrichment (wt%)	0.01	0.043	~	0.023	0.043	~	0.025
	Pellet diameter (cm)	0.0005	0.049	~	0.023	0.049	~	0.027
	Clad thickness (cm)	0.001	-0.010	~	-0.016	-0.009	~	-0.014
	Clad outer diameter (cm)	0.0005	-0.006	~	-0.010	-0.006	~	-0.009
	Pitch of fuel rod (cm)	0.01	-0.018	~	0.014	-0.015	~	0.016
	Sub total		**0.068**	**~**	**0.041**	**0.067**	**~**	**0.043**
Core tank	Tank inner diameter (cm)	0.11	0.004	~	0.029	0.002	~	0.015
	Thickness of side wall (cm)	0.03	0.003	~	0.018	-0.003	~	-0.020
	Sub total		**0.005**	**~**	**0.034**	**0.003**	**~**	**0.025**
Core	Fuel temparature (℃)	0.3	-0.002	~	-0.008	-0.002	~	-0.007
	Critical solution height (cm)	0.02	0.003	~	0.009	0.003	~	0.009
	Sub total		**0.004**	**~**	**0.012**	**0.004**	**~**	**0.011**
Sum total			**0.089**	**~**	**0.089**	**0.089**	**~**	**0.083**

and their magnitudes increase with the uranium concentration.

As to the fuel rod, uncertainties of uranium enrichment, fuel pellet diameter, outer diameter and thickness of the cladding, and lattice pitch of the unit cell were determined to be 0.01 wt.%, 0.0005 cm, 0.0005 cm, 0.001 cm and 0.01 cm, respectively. The effects on k_{eff} of uncertainties pertaining to the fuel rod are given in Table 3. The main factors to the total uncertainty of k_{eff} were found to be the pellet diameter and the uranium enrichment in order, and their magnitudes decrease with the uranium concentration.

As to the core tank, the uncertainties of the inner diameter and side-wall thickness were 0.11 cm and 0.03 cm, respectively. The effects on k_{eff} of uncertainties pertaining to the core tank are given in Table 3. The main factors to the total uncertainty of k_{eff} were found to be the tank inner diameter and the thickness of side wall in order, and their magnitudes increase with the uranium concentration.

The effects on k_{eff}'s of uncertainties pertaining to the fuel temperature and the solution height were evaluated as the "core uncertainties". The solution height was measured with a contact-type level gauge, of which the accuracy was 0.2 mm. The solution temperature was measured with a thermocouple as core temperature. The temperature change during the operation was estimated to be within 0.3 ^{o}C, including accuracy of the device. The effects on k_{eff} of uncertainties pertaining to the core are given in Table 3. Both the temperature effect and the solution height effect increase with uranium concentration.

3.2 Total Uncertainties

The calculated total effects on k_{eff}'s of material and geometrical uncertainties are summarized in Table 3 as sub-totals with respect to the fuel solution, the fuel rod, the core tank and the core. These effects for unreflected condition are shown in Fig. 6 as an

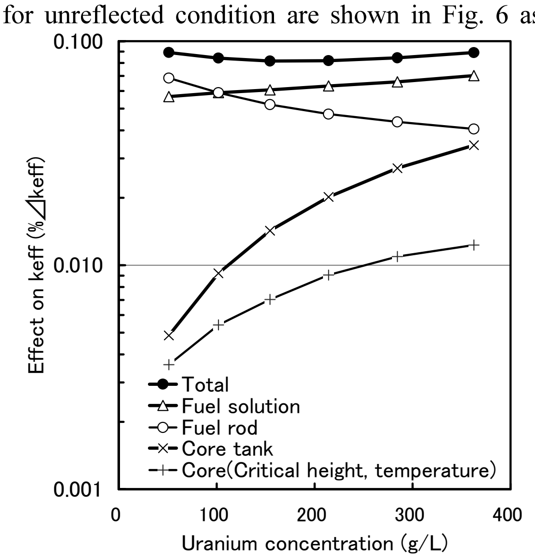

Fig.6 Effects on k_{eff} vs. uranium concentration for unreflected condition

example. As to the fuel solution, the effect increased with the uranium concentration while it decreased in case of the fuel rod. This tendency can be related to that the contribution of the fuel rods to neutron multiplication (i.e. fission fraction) decreased with the uranium concentration, which might resulted in the reduction of the effect on k_{eff}.

As to the core tank with the uranium concentration. which acts as neutron reflector for unreflected condition and absorber for water-reflected condition, the effect increased with the uranium concentration. As to the core, the effect also increased with the uranium concentration.

The total effect on k_{eff}'s shown as the sum-total changed up to 0.089 $\%\Delta k_{eff}$ with the uranium concentration for both the unreflected and the water-reflected condition. As result, it can be concluded that the total effect on k_{eff} due to all the experimental errors was evaluated to be about 0.1 $\%\Delta k_{eff}$.

4. Benchmark Calculation

4.1 Benchmark Model

The benchmark model is shown in Fig. 7. The model consists of the fuel solution, the fuel rods, the grid plates, the core tank, and the water or air around the core tank. The structures, the devices around the core tank shown in Figs 1 and 2 and the concrete wall of the room are not included in the benchmark model for simplification, while they are considered in the detailed model. The dimensions of the benchmark model are given in Fig. 7. The critical solution heights for each case are given in Table 3.

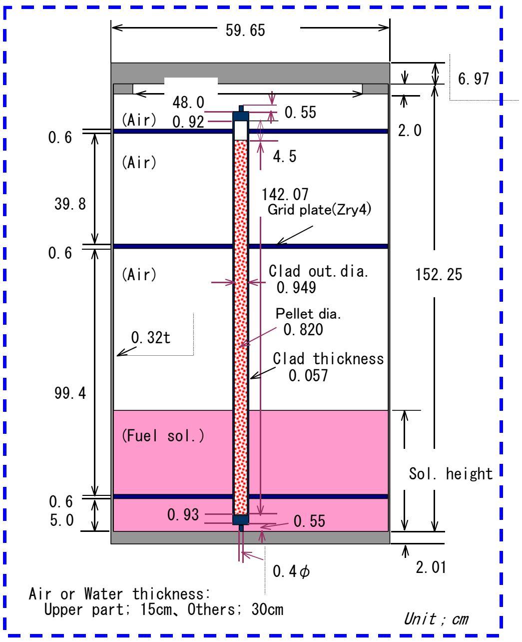

Fig.7 Benchmark model (elevation view)

4.2 Results of Calculation

The effects of model simplifications on k_{eff}'s were estimated, since a lot of structures in or around the core tank were omitted in the benchmark model. The model simplification effect is defined as the difference of k_{eff}'s between the benchmark model and the detailed model. To estimate the model simplification effect for each core configuration, a detailed model which includes the structures in or around the core tank were constructed. The calculations of k_{eff}'s were carried out by MCNP4B with JENDL-3.2 (10^7 neutron histories). The calculated k_{eff}'s for both the benchmark model and the detailed model are shown in Fig. 8

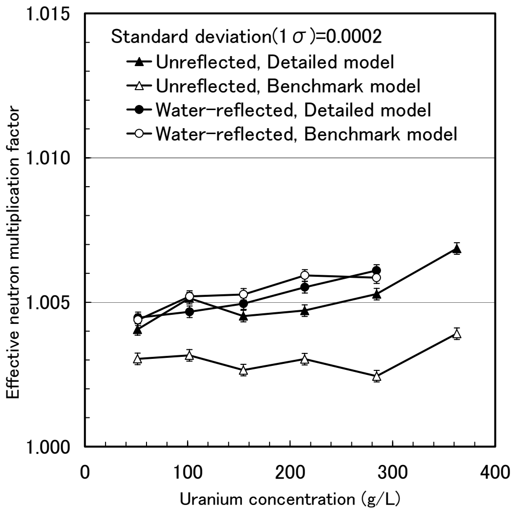

Fig.8 Calculated k_{eff}'s vs. uranium concentration

The model simplification effects are included as biases in the benchmark model k_{eff}'s. The k_{eff}'s for the detailed model, and the benchmark model with water-reflected condition were shown to be in the range of 1.004 to 1.007. The over-predicted k_{eff} values with JENDL-3.2 are mainly attributed to the fission cross section of ^{235}U which is larger than that of other libraries, and the fission spectrum of ^{235}U which is more softened than that of other libraries.

As to the benchmark model for unreflected condition, the model simplification effects are within 0.3 $\%\Delta k_{eff}$.

Because the chemical analysis of the fuel solution was carried out at a room temperature 25 ^{o}C, it is necessary to account for the temperature difference between calculations and experiments. The fuel temperatures and the effects on k_{eff} of uncertainties pertaining to the fuel temperature are shown in Tables 2 and 3, respectively. Based on these results, the temperature effects for each case were estimated by using the temperature difference between the experimental- and the adopted-temperature, and the sensitivities for k_{eff} by the temperature change. As the result, it was found that the temperature difference affects k_{eff} by only within ~0.03 $\%\Delta k_{eff}$.

5. Summary

The critical experiments of basic heterogeneous STACY core were carried out for the purpose of obtaining the benchmark data concerning the dissolver of the reprocessing facility for LWR spent fuel. The criticality benchmark data with high accuracy were obtained in the cylindrical fuel rods array with 2.1-cm square-lattice-pitch core configurations

The sensitivity analysis of k_{eff} for the uncertainty of experiment conditions showed the following results:

(1) The sensitivity to k_{eff} for each experiment condition depends on uranium concentration.

(2) The main factors to the total uncertainty of k_{eff} are found to be the free nitric acid concentration, the pellet diameter and the U-235 enrichment in that order.
(3) The total effect on k_{eff} by all the experimental errors is evaluated to be about 0.1 %Δk_{eff}.

The benchmark calculations of k_{eff}'s for both detailed- and benchmark-model were carried out using MCNP4B with JENDL-3.2. The k_{eff}'s for the detailed model were showed to be in the range of 1.004 to 1.007, and the benchmark model of water-reflected condition also showed the same result. As to the benchmark model for unreflected condition, the model simplification effects was within 0.3 %Δk_{eff}.

Because the experimental conditions are obviously known and the uncertainties of those have been sufficiently quantified, these critical configurations are judged to be acceptable as the benchmark data.

As the following phase, the critical experiments of the heterogeneous STACY core of lattice pitch 1.5 cm are undergoing for the purpose of expanding the benchmark data concerning the dissolver.

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