JAERI 1160

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June 1968

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編集兼発行　日本原子力研究所
印　　　刷　科学図書印刷株式会社

The Effective Resonance Integral of Thorium Oxide in Homogeneous Mixture

Summary

The effective ^{232}Th resonance capture integrals have been measured for homogeneous mixtures containing thorium oxide, by the activation technique. Experiments were made in the blanket region with a thorium oxide-heavy water slurry, using the Aqueous Homogeneous Critical Facility (AHCF). In the blanket, the slurry was circulated with a circulation pump so that the concentration distribution was uniform. A correction was made by calculations for the deviation of the epithermal neutron flux from the $1/E$ distribution.

The resonance integrals of the homogeneous dilution were found for the scattering cross secton per absorbing atom in the range 160~4400 ($10^{-24}\cdot cm^2$).

December 1967

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均質混合系における^{232} Th の実効共鳴捕獲積分

要　旨

放射化法により均質混合系における ^{232}Th の実効共鳴捕獲積分を測定した．実験は水性均質臨界実験装置 (AHCF) を使い，そのブランケット領域に酸化トリウム-重水スラリーを満たした中で行なった．ブランケット内ではスラリーが均一の濃度を保つように循環ポンプを使って循環させた．熱外中性子束の $1/E$ からのずれは計算で補正した．

均質混合稀釈の共鳴積分については，吸収性物質原子核当りの散乱断面積が 165~4400($10^{-24}\cdot cm^2$) の範囲で値を求めた．

1967 年 12 月

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1. Introduction

Several measurements have been reported on the infinite dilution resonance integral for thorium[1~5]. However, no data on the homogeneous dilution case of thorium have been obtained recently. It was shown by DRESNER[6] that the experimental values were somewhat larger than the calculated ones at the low dilutions.

In the studies with the AHCF, a slurry of ThO_2-D_2O is introduced into the blanket, and by changing gradually the concentration, series of critical experiments were performed. In this connection, it was thought that it was necessary in such studies to measure the effective resonance integrals of thorium. In the present study, experiments were made in the blanket with various thorium oxide concentrations.

The experimental procedure employed is in detail in section 3 and 4. The measurments were made by the cadmium-ratio technique, comparing the activities of a thin circular disk of thorium oxide and of the standard gold foil. Fabrication of the disk is described in section 4. A comparison with experimental values obtained and the theoretical ones by DRESNER and by BLAKE[7] are discussed in the last section 5.

2. Method

The effective resonance integral of resonance materials by the foil activation method is measured relative to the known resonance integral of the standard material. The cadmium ratio of the detector foil material i in the medium r is written as

$$CR^i(r)=\frac{\int_{u_c}^{\infty}\phi(u,r)\sigma^i(u)du+\int_0^{u_c}\phi(u,r)\sigma^i(u)du}{\int_0^{u_c}\phi(u,r)\sigma^i(u)du} \quad (1)$$

where

$\sigma^i(u)$: activation cross section of the detector material i,

$\phi(u,r)$: neutron flux at a lethargy u in the medium r, and

u_c : lethargy at an appropriate Cd cut-off energy.

If $\sigma^i(u)$ is assumed to have a $1/v$ charactor below the Cd cut-off energy, then the following relation can be obtained between the cadmium ratio of the detector material i, and j.

$$\frac{CR^j(r)-1}{CR^i(r)-1}=\frac{\int_{u_c}^{\infty}\phi(u,r)\sigma^j(u)du}{\int_{u_c}^{\infty}\phi(u,r)\sigma^i(u)du}\cdot\frac{I^i}{I^j}\cdot\frac{\alpha^i(r)}{\alpha^j(r)}$$

$$=\frac{\sigma_0{}^j}{\sigma_0{}^i}\cdot\frac{I^i}{I^j}\cdot\frac{\alpha^i(r)}{\alpha^j(r)} \quad (2)$$

where

$\sigma_0{}^i, \sigma_0{}^j$: activation cross sections of i and j for neutron velocity 2200 m/sec,

I^i, I^j : resonance integrals of i and j, and

$\alpha^i(r), \alpha^j(r)$: correction factors for i and j.

The correction factor is defined as

$$\alpha^i(r)=\frac{\int_0^{u_c}\phi(u,r)\sigma^i(u)du}{\int_0^{u_c}\sigma^i(u)du} \tag{3}$$

$\alpha(r)$ representing the correction factor for the deviation of the epithermal neutron flux from an ideal $1/E$ shape. This value is obtained by appropriate numerical calculation. Thus, the unknown I is determined by comparing with the known I of the standard detector, using in eq. (2). Thin circular disks of ThO_2 detectors and Au foils as the standard were prepared for the purpose of experiments.

3. Experimental Apparatus

The arrangement of the core and blanket in the apparatus is shown in **Fig. 1**. The spherical core was filled with 20% enriched (uranium) UO_2SO_4–D_2O solution, and the blanket with

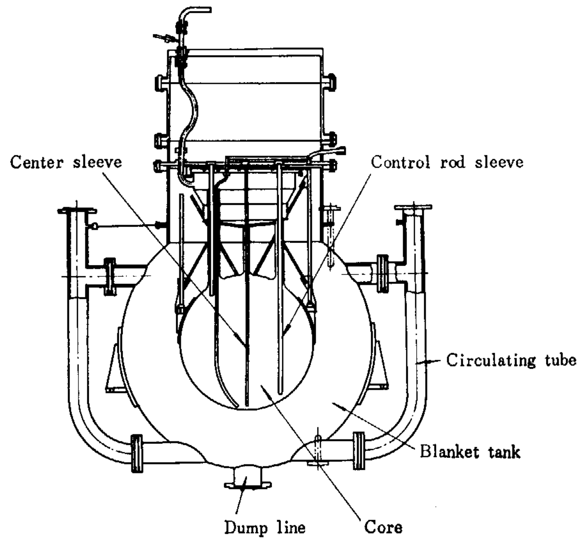

Fig. 1 Core and blanket assembly.

ThO_2–D_2O slurry. Three different cores of inner diameters 800, 600, and 530 mm, named A, B, and C′, were provided. The blanket tank had 1, 500 mm inner diameter and the blanket was changed by the various outer diameters of the core.

In order to prevent ThO_2 sedimentation, a circulating system was used, as shown in **Fig. 2**. The slurry in the dump tank was first stirred a mixer and then pushed into the blanket through the dump-tank line by N_2 gas. When the slurry filled the blanket, the dump-tank line was

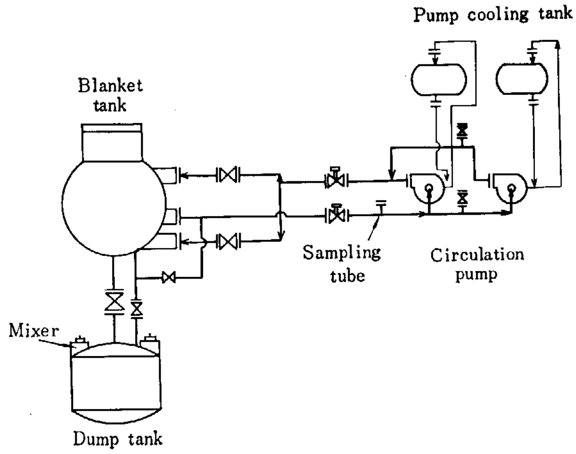

Fig. 2 Flow chart of ThO_2–D_2O slurry.

closed. Then, the circulating line was operated. The ThO_2–D_2O slurry was circulated by two canned water pumps connected in parallel and the distribution of ThO_2 grains was kept uniform in the blanket.

The critical characteristics of the apparatus are listed in TABLE 1. The concentration of the core solutions depend on the core diameter and blanket thickness. The blankets with ThO_2

TABLE 1. Data of critical characteristics

Core No.	Core volume (l)	Blanket thickness (cm)	Fuel concentration (^{235}U g/l)	ThO_2 concentration (ThO_2 g/l)
C′	75.5±0.5	48.5	25.28±0.07	36±3
B	142.1±0.3	42.0	13.10±0.04	122±3
A-2	262.1±0.5	35.0	7.09±0.02	378±4
A-3	262.1±0.5	35.0	7.46±0.02	450±5
A-4	262.1±0.5	35.0	9.02±0.03	630±6

various concentrations were provided for the experiments. The thorium concentrations were obtained by the specific-gravity measurement samples of ThO_2–D_2O slurry were taken from the the blanket in polyethilene vessels, and the weight of samples was measured with a chemical balance. There was some error in the measured thorium-oxide concentrations; and the specific gravities measured were accurate within 3%. Althouth slight ThO_2 sedimentation was observed at the high concentrations, it was considered that the effect was small.

4. Foils and Measurements

The Au foils used in the experiment were circular in shape, 8 mm in diameter and 45 mm /cm^2 thick. ThO_2 discs were prepared, as follows. ThO_2 powder was mixed uniformly with Araldite, and then diluted with a small amount of solvent. After solidification, the (thin) circular discs of 8 mm in diameter were punched out. The ThO_2 content in the discs was about 0.1 ThO_2-mg/cm^2. The discs of various thicknesses were irradiated in the apparatus reactor, i.e. in order to check the value for the self-shielding effect; the correction was small in all cases. The cadmium covers, used for Au and ^{232}Th cadmium-ratio measurements, were square cases of 11 mm×11 mm×1.8 mm, with 0.8 mm thickness.

The foils were irradiated in a central sleeve, penetrating through the blanket. Thin Au foils of 5 mm in diameter were used as the power monitor foils. After irradiation, the activities of Au and Th were measured with an end window type G-M counter. In the ThO_2, the (n, γ) reaction of ^{232}Th gives rise to the 22.2 min β decay of ^{233}Th; this activity was used for the measurements. There was a statistical counting error of about 1.5% in each case.

5. Experimental Results and Discussion

The cadmium ratios and the correction factors in the center of the core are given in TABLE 2. The deviation from the $1/E$ spectrum depend on the neutron leakage and absorption occurring in the system. For small reactor-systems, this deviation is generally remarkable. For the AHCF, with an absorbing reflector, it was found that a wide-range $1/E$ neutron spectrum occurs in the blanket a small distance outside the boundary[8]. The irradiations were made with this knowledge. The correction factor, $\alpha(r)$ in eq. (3), were obtained from the calculation.

TABLE 2. Cadmium ratios and correction factors at core center

Core No.	$CR_{Au}(O)$	$CR_{Th}(O)$	$\alpha^{Au}(O)/\alpha^{Th}(O)$
C′	1. 22±0. 01	1. 27±0. 01	0. 801
B	1. 38±0. 02	1. 46±0. 02	0. 859
A-4	1. 55±0. 02	1. 66±0. 03	0. 878
A-3	1. 64±0. 03	1. 81±0. 03	0. 886
A-2	1. 66±0. 03	1. 86±0. 03	0. 892

The SEFAC-1 code[9)] was used for this purpose. The calculation was done by using this code to estimate the spatial dependence of 54-group epithermal spectra. In this code, the spatial distribution of the fission source in the core was given by the measured value (the traverse with a micro-fission counter). The accuracy of the calculation was about ±10%.

The positions of samples in the blanket were, from the core center,

A core 50∼55 cm
B core 40∼45 cm
C′ core 33 cm.

The experimental values of the effective resonance integral as a function of the scattering cross section per absorbing atom are given in **TABLE 3.** In the table, the quantity σ_p denotes the

TABLE 3. Effective resonance capture integral of ^{232}Th as a function of potential scattering cross section

σ_p	162	332	717	1253	4345	∞
I_{eff}	32. 4	38. 7	45. 6	57. 4	72. 9	83

Results include $1/V$ contribution

scattering cross section per absorbing atom of the thorium oxide-heavy water mixture. The error in the results were caused mainly by the uncertainty in the cross sections used and in the correction factor for the departure of the neutron energy distribution from the $1/E$ shape. The values used in eq. (2) are

I^{Au} : 1535±40 barns[10)]
$\sigma_0{}^{Au}$: 98. 8±0. 3 〃
$\sigma_0{}^{Th}$: 7. 56 〃

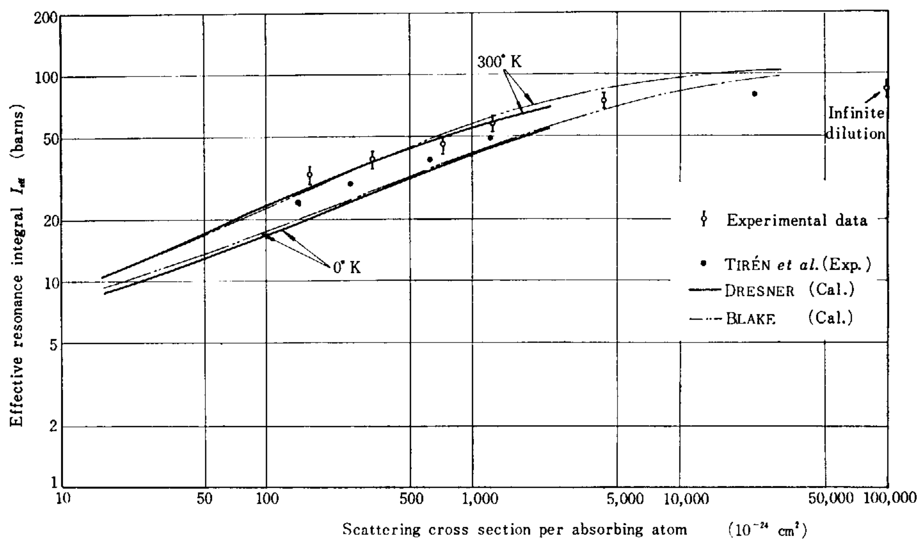

Fig. 3 Effective resonance capture integrals of ^{232}Th diluted with heavy water.

The σ_0^{Au}, σ^{Th} were from BNL-325 (2nd edi.).

The values of I_{eff} are plotted in **Fig. 3**. As seen in the figure, the experimental values obtained agreed with those calculated by DRESNER at the high dilutions. For the low dilutions, however, the experimental values were slightly higher than the DRESNER's curve. The values by TIŘEN and JENKINS[11]), for themetalic thorium powder diluted with graphite, were lower than the present ones.

The infinite dilution resonance integral of of 83±8 barns was obtained; in recent years, this resonance integral has been obtained in good agreement by many authors. BROSE obtained the experimental value of 82. 7±1. 8 barns and calculated 96 barns. On the other hand, the values by FOELL and CONNOLY[12]) are : 81. 2±3. 4 barns (measured), 82. 3 barns (calculated). It is evident therefore that the agreement between the theoretical and the experimental values is good for the infinite dilutions.

Acknowledgments

The author would like to thank the AHCF staff for their many helpful discussions and the technical assistance in performing the experiments.

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