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Doppler effect measurement on resonance materials for rock-like oxide fuels in an intermediate neutron spectrum

M. Andoh^{*}, Y. Nakano, S. Okajima, K. Kawasaki

Department of Nuclear Energy System, Japan Atomic Energy Research Institute, 2-4 Shirakata Shirane, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan

Abstract

Doppler effect experiments on resonance materials for rock-like oxide (ROX) fuels were carried out to examine the calculation accuracy in the intermediate neutron spectrum of the Fast Critical Assembly (FCA) of the Japan Atomic Energy Research Institute. This study is the second phase of a series of Doppler effect experiments on the resonance materials, which follows the measurements in the fast neutron spectrum. The Doppler effect was measured as the sample reactivity change between the heated and unheated samples. Cylindrical samples of resonance materials such as erbium metal (Er), tungsten metal (W) and thorium dioxide (ThO₂) were used. The sample was heated up to 1073 K at the center of the FCA core. The experiments were analyzed by the SRAC code system with the use of the JENDL-3.2 cross-section library. The calculated Doppler effects of the W and ThO₂ samples agreed with the measured values within the experimental error, while the calculation overestimated the measured Doppler effect of the Er sample by 12%.

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1. Introduction

As a rock-like oxide (ROX) fuel contains only small amounts of fertile materials, the ROX fueled Light Water Reactor (LWR) may have reactivity coefficients problems such as insufficient Doppler effect. The Doppler effect is one of the most important nuclear characteristics in the case of reactivity accidents. By adding resonance materials (thorium, erbium, etc.) to the ROX fuel, the Doppler reactivity of the ROX fueled LWR was improved [1]. The effects of adding the resonance materials to the ROX fuel were analyzed [2], from the point of view of burnup characteristics and radio toxicity hazards.

Doppler effect measurements have been carried out in various Fast Breeder Reactor (FBR) mock-up cores in the Fast Critical Assembly (FCA) of the Japan Atomic Energy Research Institute (JAERI) with the use of uranium samples [3]. To examine the calculation accu-

racy of the Doppler effect of resonance materials for ROX fuels, a series of the Doppler effect experiments on certain resonance materials has been carried out at the FCA. As the first phase of this study, the Doppler effect was measured in the fast neutron spectrum in the FBR simulated core (FCA XX-2 core) [4]. It was shown that measured Doppler reactivity worths of erbium, thorium and tungsten samples were comparable to that of uranium and that the SRAC code system [5] predicted the measurements within the experimental error, except for the tungsten sample where the calculation underestimated the measurement by about 10%.

The purpose of the present, second, phase of this study, is to obtain experimental data of the Doppler effect in an intermediate neutron spectrum and to examine the calculation accuracy of the SRAC system.

2. Experiment

2.1. Core description

The Doppler effect was measured in the FCA XXI-1 core. The fuel element used at FCA is plate- or

^{*} Corresponding author. Tel.: +81-29 282 5329; fax: +81-29 282 6181.

E-mail address: andoh@fca001.tokai.jaeri.go.jp (M. Andoh).

block-shaped and is 5.08×5.08 cm in size. An intermediate neutron spectrum is formed at the center of the test zone that consists of plates of enriched uranium, natural uranium, polystyrene and stainless steel. The averaged ^{235}U enrichment of the test zone is 15%. The atomic number ratio of ^1H to ^{235}U of the test zone is 3.3. The test zone is surrounded by a driver zone that con-

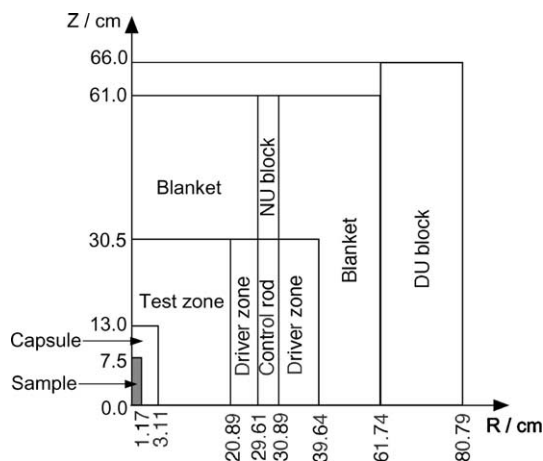


Fig. 1. R - Z model of the FCA XXI-1 core with the Doppler sample.

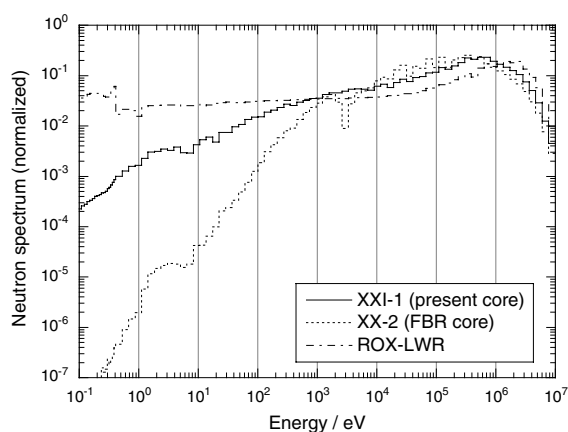


Fig. 2. Comparison of the calculated neutron spectrum between the FCA XXI-1 and XX-2 cores.

sists of enriched uranium and stainless steel plates and by an axial blanket that consists of depleted uranium dioxide and sodium plates. Fig. 1 shows the R - Z geometry of the core with a Doppler sample in the center of the test zone. The calculated neutron spectra in the core are compared between both the FCA cores and a typical ROX fueled LWR with weapon-grade plutonium [6] in Fig. 2. This shows that the spectrum in the core of the present phase of this study is intermediate one between those in the FBR and LWR.

2.2. Measurement of the Doppler effect

The Doppler effect was measured as the sample reactivity change between the heated and unheated samples. Cylindrical Doppler samples of the resonance materials, such as erbium metal (Er), tungsten metal (W) and thorium dioxide (ThO_2), were used. The natural uranium metal and dioxide (U-metal and UO_2 , respectively) samples were also used, to obtain reference data. The Doppler samples are 150 mm in stack length and 23 (25 for the uranium samples) mm in diameter. The sample is enclosed in a stainless steel capsule and is inserted into the center of the FCA core. Each Doppler sample is heated up to 1073 K by an electric heater set in the capsule.

Table 1 shows the measured Doppler reactivity worths (ρ_{Dop}) of each sample, where the temperature change is from room temperature to 573, 823 and 1073 K. To compare the measured ρ_{Dop} between both the FCA cores, the ρ_{Dop} of the resonance materials are normalized to the values of the uranium samples; the Er and W samples are normalized to the U-metal sample and the ThO_2 sample is normalized to the UO_2 sample. The normalized worths (Norm. ρ_{Dop}) from room temperature to 1073 K are listed in Table 2. The Norm. ρ_{Dop}

Table 2

Comparison of the measured Doppler reactivity worths normalized to the values of the uranium samples (293–1073 K)

Sample	XXI-1	XX-2
Er	1.10 ± 0.06^a	0.86 ± 0.08
W	0.78 ± 0.05	0.68 ± 0.08
ThO_2	1.13 ± 0.09	0.98 ± 0.16

^a Experimental error.

Table 1

Measured Doppler reactivity worths of each sample ($\times 10^{-5} \Delta k k^{-1}$) at the sample temperatures (T) raised from room temperature

T (K)	Er	W	ThO_2	U	UO_2
573	-2.51 ± 0.16^a	-1.73 ± 0.17	-1.53 ± 0.16	-2.14 ± 0.18	-1.31 ± 0.17
823	-3.93 ± 0.16	-2.75 ± 0.17	-2.36 ± 0.15	-3.43 ± 0.16	-2.03 ± 0.16
1073	-5.07 ± 0.17	-3.59 ± 0.18	-3.11 ± 0.16	-4.60 ± 0.18	-2.74 ± 0.16

^a Experimental error.

of the resonance material samples are increased in comparison with those in the XX-2 core by up to 25%. As shown in this table, it was found that the intermediate neutron spectrum benefited the Doppler effect of those resonance materials that have resonance reactions in the lower energy range.

3. Analysis

3.1. Calculation method

Calculations of the ρ_{Dop} were performed using the SRAC system with a 101-group energy structure. The nuclear data library used for the calculations is JENDL-3.2 [7]. As for the Er sample, nuclear data of the JENDL-3.3 library were used. The effective macroscopic cross-sections of each zone of the core were calculated by a collision probability method with one-dimensional slab geometry. The effective cross-sections of the Doppler samples were calculated with one-dimensional cylindrical geometry using PEACO, a collision probability routine with an ultra fine energy structure, to

consider resonance interaction effect between the sample and the adjacent core.

Neutron flux distributions with the Doppler samples at room temperature (293 K) at the core center were obtained by diffusion calculations in two-dimensional R - Z geometry using the CITATION module of the SRAC system. The ρ_{Dop} were calculated by first-order perturbation theory by replacing the cross-section of the Doppler sample at 293 K with the cross-sections at the other temperatures.

3.2. Results and discussions

The comparison of the energy breakdown of the calculated Norm. ρ_{Dop} for the resonance material samples and the calculated ρ_{Dop} of a typical uranium (UO_2) sample between both the FCA cores are shown in Fig. 3. This shows that the difference of the neutron spectra shown in Fig. 2 affects the energy dependency of the ρ_{Dop} very strongly.

The C/E values of the ρ_{Dop} (from 293 to 1073 K) are listed in Table 3 together with those in the XX-2 core [4]. The calculated ρ_{Dop} of the W and ThO_2 samples agreed

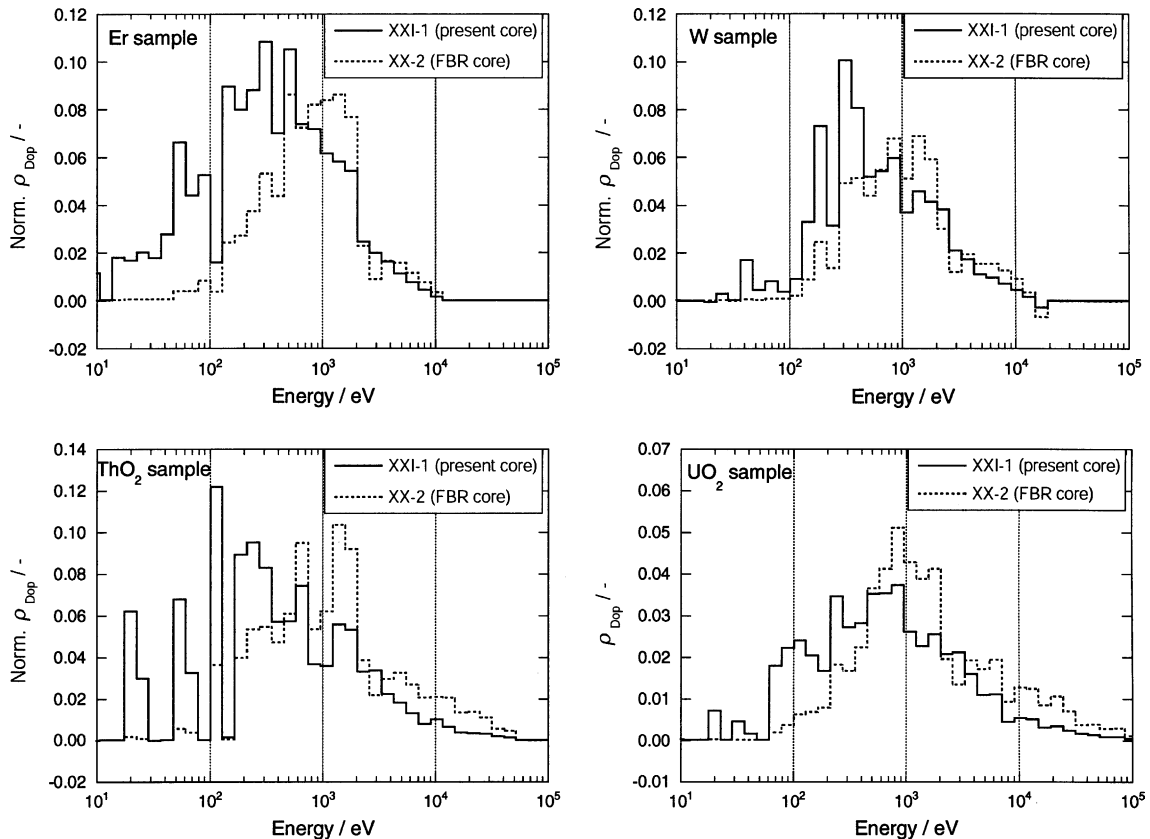


Fig. 3. Comparison of the energy breakdown of the calculated Doppler reactivity worth (293–1073 K) between the FCA XXI-1 and XX-2 cores.

Table 3
C/E values of the Doppler reactivity worths (293–1073 K)

Sample	XXI-1 (%)	XX-2 (%)
Er	1.12 ± 3.3 ^a	0.98 ± 6.6
W	0.99 ± 5.0	0.89 ± 8.9
ThO ₂	1.04 ± 5.1	0.99 ± 12
U	1.05 ± 3.9	1.06 ± 6.6
UO ₂	1.07 ± 5.8	1.03 ± 11

^a Experimental error (relative error).

with the measured values within the experimental error. There is a tendency for the C/E values for the resonance material samples in the present core to be bigger than those in the XX-2 core. The calculation overestimated the experiment for the Er sample by 12%, while it showed good agreement with the measurement in the XX-2 core.

To investigate the result of the Er sample, the sensitivity analysis and transport calculation were carried out using the SAGEP [8] and TWODANT [9] codes, respectively. JAERI's 70-group constant set for fast reactor analysis based on JENDL-3.2 (JFS-3-J3.2) was used for the calculations.

Fig. 4 shows the calculated sensitivity coefficients of the capture reactions of two dominant nuclides, ¹⁶⁶Er and ¹⁶⁷Er, to the ρ_{Dop} (from 293 to 1073 K). The figure shows that ¹⁶⁷Er is more dominant for the ρ_{Dop} in the present core. The calculated sensitivity coefficients of the capture reaction of ¹⁶⁷Er to the ρ_{Dop} are compared between the XXI-1 and XX-2 cores in Fig. 5. This figure shows that the capture reaction around 200 and 50 eV is more important in the present core than in the XX-2 core.

The transport calculation gave the result that the transport effect, the ratio of the calculated ρ_{Dop} between

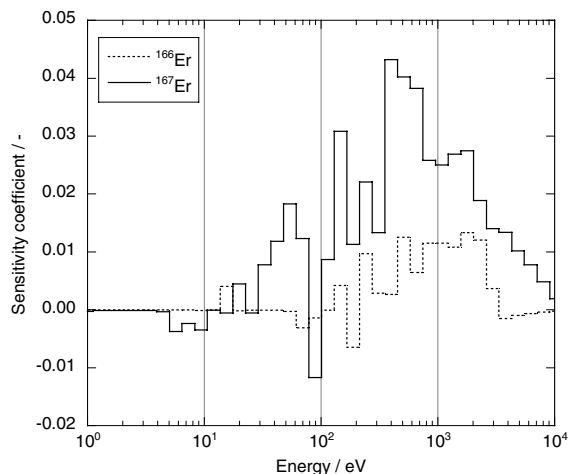


Fig. 4. Sensitivity of the capture reaction of ¹⁶⁶Er and of ¹⁶⁷Er on the Doppler reactivity worth (293–1073 K).

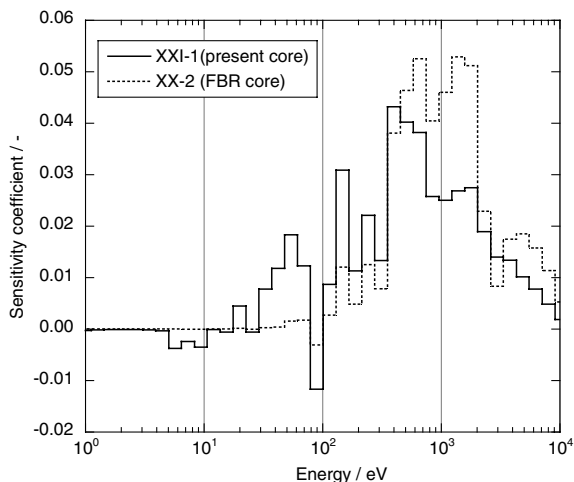


Fig. 5. Comparison of the sensitivity of the capture reaction of ¹⁶⁷Er between the FCA XXI-1 and XX-2 cores.

the transport and diffusion calculations, on the Er sample was –12.7%, while the effects on the other samples were about –7%. The C/E value of the Er sample was improved to be 0.98 with the correction of the transport effect. The ratio of the neutron flux between the transport and diffusion calculation is shown in Fig. 6 for the Er and ThO₂ samples. This figure shows that the change of the neutron spectrum in the Er sample below 10 keV is bigger than that in the ThO₂ sample, which means that the diffusion calculation underestimates the neutron flux depression in the resonance energy region in the sample. It can be said that the calculation accuracy of the diffusion theory is insufficient for the core calculation with the Er sample.

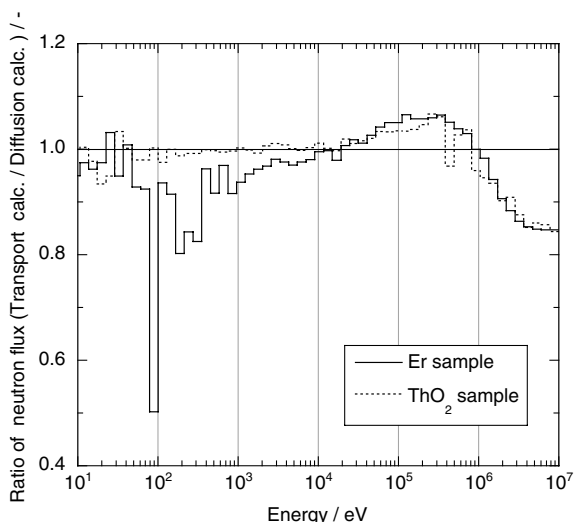


Fig. 6. Ratio of the calculated neutron flux between the transport and diffusion calculations.

There is a tendency that the calculations give smaller C/E values for the W sample relative to the other samples in both the cores. The transport corrected C/E value for the W sample in the XXI-1 core is 0.92, which means that the calculation underestimates the measured value beyond the experimental error of 5%. Tungsten has a giant resonance cross-section around 6 eV. It is considered that there is a problem with the calculated effective cross-section of the W sample or with the nuclear data library itself.

It is concluded that the diffusion calculation using the SRAC system has good accuracy for the analysis of the Doppler experiments in the intermediates neutron spectrum, except for the Er sample.

4. Conclusions

Experimental data of the Doppler reactivity worths for erbium metal (Er), tungsten metal (W) and thorium dioxide (ThO_2) in an intermediate neutron spectrum were compared with those in a fast neutron spectrum. It was found that the Doppler reactivity worths, normalized with the uranium samples, were increased in the range from 15% to 25% in the intermediate neutron spectrum.

Calculations were performed using the diffusion calculation module, CITATION, and the first-order perturbation routine in the SRAC code system. The calculated Doppler reactivity worths of the W and ThO_2 samples agreed with measured values within the experimental error, while the calculation overestimated the experiment for the Er sample by 12%. From the comparison of the C/E values between both the cores, it was found that there was a tendency that the diffusion calculation gave bigger C/E values for the resonance material samples in the XXI-1 core than in the XX-2 core. The overestimate for the Er sample was improved by the transport correction. The transport correction for the W

sample, however, gave the result that the calculation underestimated the measured value beyond the experimental error. It has been confirmed that the calculation method had good accuracy for the analysis of the experiment both for the fast and the intermediate neutron spectrum throughout the series of the experiments except for the tendency to underestimate for the W sample in both spectra and the overestimate for the Er sample in the intermediate neutron spectrum.

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