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Effects of Neutron Resonance Interference between Ta and Au

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ABSTRACT

The neutron resonance mutual interference between Ta and Au has been experimentally investigated at the central graphite cavity of the UTR-KINKI, where the neutron spectrum shows a good 1/E standard neutron field at energies from about 1 eV to a few hundreds keV. Tantalum was selected as a shielding sample and used as a box. A gold foil was set inside the shielding box for the measurement of the resonance interference effects between Ta at 4.3 eV and Au at 4.9 eV by the neutron activation method. Instead of Au, copper was employed for the measurement without the mutual interference because Cu has no resonance in the relevant energy region. By changing the window thickness of the Ta box, the induced activities of Au and/or Cu foil were measured to investigate the mutual interferences. The calculations were performed using the Monte Carlo code VIM and compared with the measured data. A good agreement was seen between the calculations and the measurements.

KEYWORDS

UTR-KINKI, resonance interference, mutual interference, Ta, Au, shielding effect, Cu, induced activities, measurement, Monte Carlo calculation, VIM code.

Introduction

In the neutronic calculations for safety reactor design, it is of great importance to assess the resonance self-shielding[1),2)] and mutual shielding effects [3)-5)] in the fuels and reactor structural materials, especially for a reactor with an intermediate neutron spectrum such as high conversion light water reactor. In the energy region, the resonance shielding effects with fission products are also important for burnup calculation.[3)-5)] Takano et al. pointed out that some of the main resonance peaks for typical nuclides, such

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as ^{240}Pu, ^{155,157}Gd, ^{149,150,151}Sm, ^{133}Cs overlap with that of ^{238}U and that the mutual interference makes the resonance integral for these nuclides reduce.[5] The reactivity loss with burnup is caused predominantly by fission product absorption, which is about 60% of the total reactivity loss for the reactor in the resonance energy region.

The mutual interference effects of resonance overlapping among major actinides and fission products are required for the estimation of the reactivity change and performing the burnup calculations. Nevertheless, it is said that there still exist discrepancies between the nuclear data of fission product nuclides, even among the data in the evaluated nuclear data files.[5] To solve the discrepancies and/or problems, integral tests with the resonance interference and/or mutual shielding experiments would be useful for the investigation not only of the resonance parameters but also of the neutron resonance energy, because the mutual interference has high sensitivity in the resonance energy region.

As a research prior to the integral investigation of the mutual interference effects between ^{238}U and fission products and/or ^{238}U and actinide nuclides, the aim of this study is to develop an experimental method for investigating the mutual resonance shielding and/or interference effects and to verify the validity of the integral test for the evaluation of nuclear data in the intermediate neutron energy region. In the present work, gold and tantalum whose nuclear data are relatively well evaluated were selected for the mutual interference experiments at the resonances of 4.9 and 4.3 eV, respectively. Tantalum box was prepared for the shielding sample and gold foil inside the box was used to measure the mutual interference by the neutron-induced activities. Moreover, copper sample was used to investigate the results without resonance interference, because copper has not a resonance in the eV energy region. The experiments were made in the 1/E standard neutron spectrum field which had been obtained by the present authors.[6] Comparing the calculations using the Monte Carlo code VIM[7] with the measurements for the mutual interference effects, the integral tests of the resonance neutron energies and parameters for Ta and Au have been performed.

Experimental Method

1) Facility

The Kinki University Reactor, UTR-KINKI, is a light water-moderated and graphite-reflected research reactor,[8),9] which has the 46 cm separate cores. It has twelve fuel elements and each of them contains twelve aluminum-clad, flat ETR-type fuel plates, whose enrichment is 90%, and the critical mass is approximately 3,018 grams of ^{235}U. The nominal output power is 1 W. At the center of the internal graphite reflector between the two-divided cores as shown in Fig. 1, there is a graphite stringer of 9.6×9.6 cm square and 122 cm long, which can be withdrawn to make a void region or a central cavity for irradiation of large samples. An aluminum sample holder of 66 cm length with tantalum and gold samples was set at this central cavity, and sandwiched by two 28 cm long graphite stringers up and down, to save the neutron leakage from the void region, so that one can expect an uniform and flat distribution of neutrons in the internal graphite reflector. The experiment has been carried out at the central cavity, where the neutron spectrum shows a good 1/E standard neutron field in the energy region from about 1 eV to a few hundreds keV.[6]

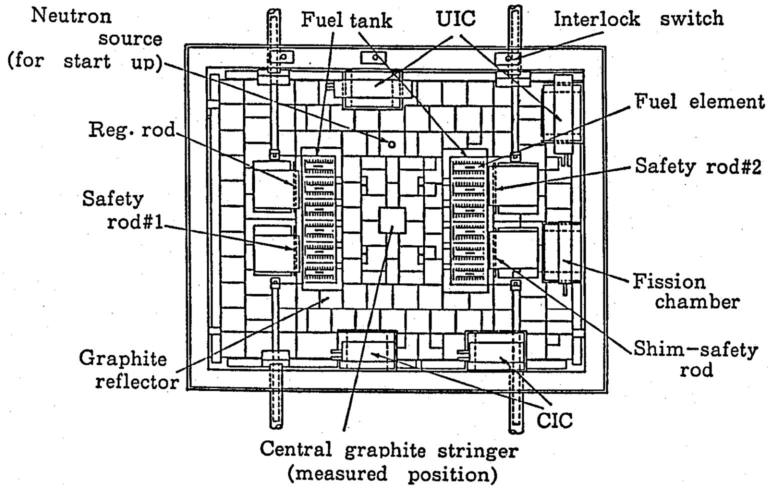

Fig. 1 Cross sectional view of UTR-KINKI.

2) Ta and Au Samples

Gold (Au) has a strong resonance at 4.9 eV and tantalum (Ta) has at 4.3 eV near the Au resonance, respectively, so that one can expect the mutual resonance interference between the nuclei. Copper (Cu) has no resonance structure in the relevant eV energy region. Then the Cu was selected as a sample which has no interference effect with Ta.

For the resonance neutron interference measurements between Ta and Au, a shielding box was prepared by Ta plates of 45 mm in diameter. Some of the plates have a hole of 15 mm in diameter at the plate center to put a Au foil together with Cd-cover of 0.5 mm thickness, as seen in Fig. 2. The size of the Au foil is 50 μm thick and 12.7 mm in diameter. The Ta box with the Au foil was set at the center of the sample holder which was inserted at the central graphite cavity. By changing the window thickness of the Ta

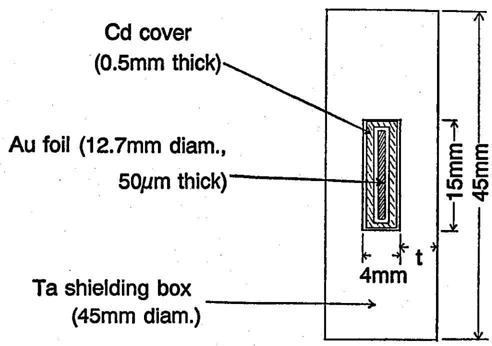

Fig. 2 Tantalum box for the present mutual experiment.
t=0, 0.5, 1.0, 2.0, 4.0, 6.0, 9.0 mm.

box, the resonance shielding and/or interference between Ta and Au samples has been measured by the activation method using the Au foil. For the experiment without the resonance interference and/or shielding effect, a copper foil was employed instead of the Au foil. The Cu foil was 0.3 mm thick and 12.7 mm in diameter.

3) Irradiation and Activity Measurement

The irradiations of Ta box with a Au foil or a Cu foil were performed for 30 minutes to two hours at the central graphite cavity of the UTR-KINKI with the nominal power of 1 W. The Ta plates were selected as a shielding sample and the activation foil for the $^{197}Au(n, \gamma)$ ^{198}Au or the $^{63}Cu(n, \gamma)$ ^{64}Cu reaction was used to measure the reaction rate caused by neutrons passed through the shielding Ta box window. In each irradiation run, two Au foils were also used without a Cd-cover to monitor the neutron intensity between the experimental runs, as done before.[6] Induced activities of 412 keV gamma-ray from the Au foil and of 511 keV from the Cu foil were measured with a HPGe detector. Calibration of gamma-ray energies and the detection efficiency was made with standard gamma-ray sources purchased from the Bureau National de Matrologie, France. The data reduction of the activation foil was similarly done as before.[6]

Calculation

The mutual interference calculations were performed with a continuous slowing down Monte Carlo code VIM,[7] using nuclear data libraries produced from the ENDF/B-IV file. In order to investigate the mutual resonance interferences, we calculated the reaction rates for the $^{197}Au(n, \gamma)$ ^{198}Au and the $^{63}Cu(n, \gamma)$ ^{64}Cu reactions which were induced by neutrons transmitted the window plates of the Ta shielding box, as seen in Fig. 2. In the calculation, the neutron energy spectrum used was taken from the literature,[6] which showed a good 1/E standard neutron field in the energy range from about 1 eV to a few hundreds keV. Between Ta and Au, strong resonance interference can be expected because the Au resonance at 4.9 eV is pretty close to the Ta resonance at 4.3 eV. Total number of energy mesh was 37 in the energy region from 1 meV to 1 keV, and the energy dependent interference spectra were calculated by changing the Ta window thickness. The reaction rates from the $^{197}Au(n, \gamma)$ ^{198}Au reaction show the results of the resonance interference with Ta. To compare and/or investigate the results for the case without the mutual interference, the reaction rates for the $^{63}Cu(n, \gamma)$ ^{64}Cu reaction were obtained, because Cu shows a 1/v energy dependent cross section and has no resonance structure in the relevant energy region. In the calculations for the mutual resonance interference and /or shielding effects using a set of Ta and Au and/or a set of Ta and Cu, the induced activities from the Au and/or Cu foil were obtained depending upon the Ta window thickness.

Results and Discussion

The reaction rate spectra near the resonance region for Au and Ta are calculated with the VIM code and shown in Fig. 3, where deep dips caused by the shielding of the Ta big resonance at 4.3 eV can be observed in the calculated reaction rate spectra for Au. Spectral changes due to the resonance interference between Ta and Au can be seen as a function of window thickness of the Ta box. Fractional distribution of the calculated reaction rates for the $^{197}Au(n,\gamma)^{198}Au$ reaction are given in Table 1. One can recognize that most of the reaction rates exist near the resonance region from 0.1 to 700 eV. So we have decided that we could evaluate the nuclear data for the Ta and Au cross sections even through the integral data such as the reaction rate measurement.

The reaction rate data for the $^{197}Au(n,\gamma)^{198}Au$ reaction have been measured by changing the window thickness of the Ta box, and the results are shown in Fig. 4. The reaction rates measured with the $^{63}Cu(n,\gamma)^{64}Cu$ reaction are also given in Fig. 4. Using the Monte Carlo code VIM, the calculated results for these reactions have been compared with the measurements. The experimental uncertainties are within 2 to 3% and the VIM calculations are in the several percent uncertainties at most, for the 10^6 histories run. It can be seen that the reaction rate data for Au are rapidly decreasing with the Ta window thickness due to the mutual interference. However, Cu data are changing slowly and following only neutron fluxes which are obtained by neutrons penetrating the Ta windows. That is, the difference of the slopes for the Ta-Au and Ta-Cu curves are due to the existence of the mutual interference effects between the Ta and Au samples.

For both cases with and without the interferences, a good agreement can be seen be-

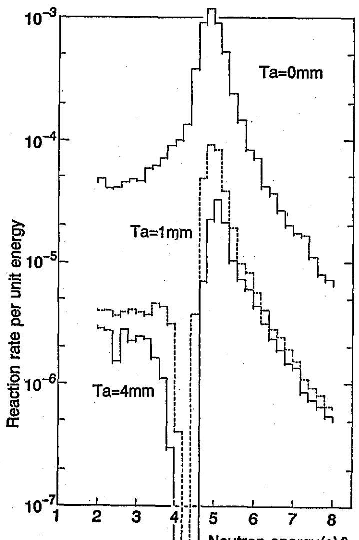

Fig. 3 Reaction rate spectra at Au and Ta resonances calculated with the VIM code.

Table 1 Fractional distributions of the reaction rates for the $^{197}Au(n, \gamma)^{198}Au$ reaction depending upon the window thickness of Ta box.

Ta thickness Energy (eV)	Fractional distribution of Au reaction rates		
	0 mm (%)	1 mm (%)	4 mm (%)
1,000 —700	0.06	0.26	0.36
700 —598	0.06	0.34	0.56
598 — 70	0.88	5.63	8.28
70 — 8	1.25	6.99	10.7
8 — 2	95.5	74.0	62.5
2 — 0.5	1.92	10.9	15.2
0.5— 0.1	0.31	1.74	2.21
0.1— 0.001	0.02	0.14	0.13

tween the measured and the calculation, as shown in Fig. 4. This implies that the existing nuclear data for Au, Ta and also Cu in ENDF/B-IV which was used in the calculations are relatively well established and evaluated. It is also said that the present technique and/or procedure for the investigation of the mutual resonance interferences are applicable and useful for the integral tests of the neutron resonance energies and resonance parameters, which would be interested in neutronic calculations, especially for the reactions with hard neutron spectrum such as high conversion light water reactors.

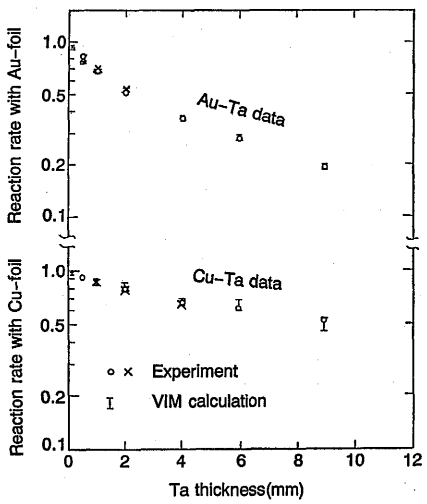

Fig. 4 Comparison of the measured and the calculated reaction rates by the resonance mutual interference effects between Ta and Au and between Ta and Cu.

Conclusion

The resonance mutual interference between Ta and Au have been investigated using a good 1/E standard neutron spectrum field at UTR-KINKI. The shielding sample was prepared by the Ta box which has a Au foil inside for the measurement of the interference effects by the neutron activation method. The Au activities were measured by changing the window thickness up to 9 mm of the Ta box. Instead of the Au foil, copper was selected for the measurement without the resonance mutual interference. Calculations were performed with the Monte Carlo code VIM and the results were compared with the measurements.

Most of the Au activities are derived from the energy region near the resonances of Ta at 4.3 eV and Au at 4.9 eV. The reaction rates from Au are rapidly decreasing due to the resonance interference between Ta and Au, as the Ta window thickness increases, while the Cu slope changes gently by the Ta shielding effects only. The fact that a good agreement between the calculations and the measurements can be seen would imply the validity of the nuclear data for Ta and Au, especially for the resonance parameters and the resonance energies. From the present results, it can be also said that the integral test can be applicable for the investigation of the resonance mutual interference effects.

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