Elemental distribution measurement of the Au-In-Cd alloy by neutron resonance imaging

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Abstract

The Japan Spallation Neutron Source (JSNS) at the Japan Proton Accelerator Research Complex (J-PARC) has been developed as a 1MW spallation neutron source. A Au-In-Cd alloy has been proposed and produced as a low activation decoupler material for two decoupled hydrogen moderators. Elemental distribution of the Au-In-Cd alloy was measured with SEM/EDX. However, due to very small amount of In, it was difficult to measure the distribution of the In. Then we utilize a pulsed neutron imaging tecunique. Measurement was performed at MLF BL10 in J-PARC. A Au-In-Cd specimen (0.9mm), In foil (10μm) and two Au foils (0.5mm and 1mm) are used as a sample. With the resonance neutron imaging, we can realize that the elemental distribution of the Au-In-Cd alloy was uniform.

1. Introduction

The Ag–In–Cd (AgIC) alloy was adopted as a decoupler material for two decoupled moderators of the J-PARC pulsed spallation neutron source JSNS [1]. A high decoupling energy at 1 eV was achieved for the first time in the world in MW-class spallation neutron sources[2,3]. Although the AgIC decoupler is superior in neutronic performance, its high residual radioactivity originating from Ag imposes a significant difficulty in handling the used moderators. To overcome this difficulty, a new alloy in which Ag in the AgIC alloy is replaced with Au, Au-In-Cd (AuIC) alloy, was proposed[4] by Harada et. al. Radioactivity in the AuIC decoupler can be reduced to about 1/1000 of that in the AgIC decoupler.

Accordingly, we have launched an R&D activity on the AuIC alloy production. As the first step, we made AuIC alloy specimens by melting the three elemental metal pieces in a small furnace and an infrared heater[5]. Target alloy compositions determined are 74.9, 0.5 and 24.6 atomic % for gold, indium and cadmium, respectively. The elemental distributions of gold and cadmium on a specimen surface were measured by a SEM-EDX (scanning electron microscope - energy dispersive X-ray spectroscopy) and were confirmed to be uniform. However, X-ray peak tails of cadmium overlapped X-ray peaks of indium because the indium composition was much smaller than the cadmium composition and the atomic number of indium (49) is next to that of cadmium (48). Therefore, it is difficult to see the indium distribution with the SEM-EDX method. In order to confirm the uniformity of the alloy including indium, we used a neutron resonance imaging technique. The resonance imaging is one of time-of-flight imaging technique aiming for neutron energy over eV regions.

Most of elements have resonance structure in their cross-sections as shown in Fig. 1. Neutron resonance energies are inherent for each nuclide. For example, when a sample is placed at 14 m from the neutron source and we see a transmission image of the AuIC specimen at t=475 μs after pulsed neutron generation, we can see an image contributed by the gold content only (see Fig. 1). Accordingly, we can distinguish certain elements in a sample by the neutron resonance imaging.

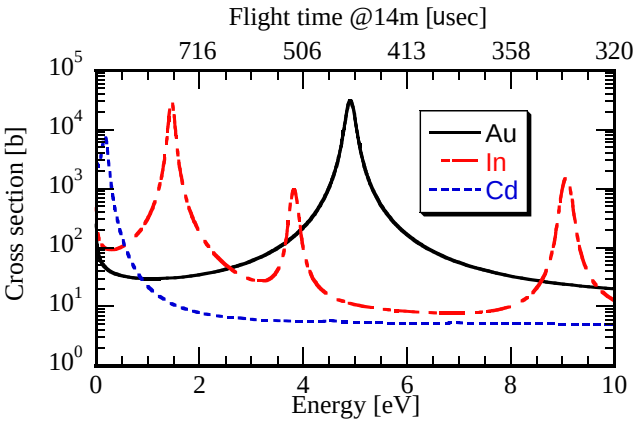

Figure 1. Neutron cross section of Au, In and Cd.

2. Experiment

Imaging experiment was performed at BL10[6] of the Materials and Life science experimental Facility (MLF). Epi-thermal neutrons needed for the neutron resonance imaging are available at BL10 because of its straight beam line configuration. One of the specimens was cut to a disk sample of 0.9 mm in thickness and 6.5 mm in diameter. The disk sample was set on a thin aluminum plate with other reference samples, i.e., indium (10 μm) and gold (0.5 mm and 1 mm), as shown in Fig. 2. Figure 3 shows an outline of a detector system which was developed by Kureta et al. A CMOS-type camera was coupled with an image intencifire (I.I.). A scintillator plate and an optical lens system were set on 45 degrees with respect to the neutron beam. Visible light from the scintillator plate was amplified by the I.I. and recorded by the camera. The I.I. was operated in synchronization with external signals to function as a shutter of the camera. By inputting a gate signal to the I.I. with a certain delay from the T0 timing, we could get an image of a certain energy range of interest. In order to obtain clear images, images for 4000 or 8000 neutron pulses were accumulated in one measurement. Neutron energies, trigger delay, gate width and number of accumulated images are summarized in Table 1.

Figure 2. Sample

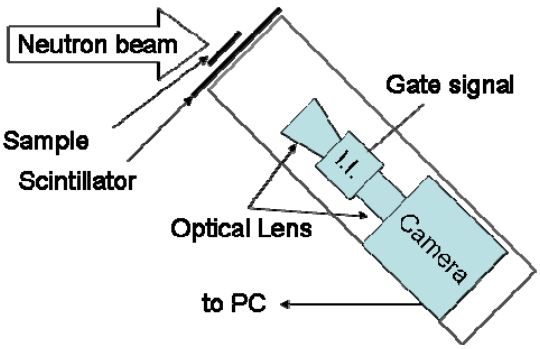

Figure 3. Outline of the detector system

Table 1 Measurement conditions.

Target material	Energy[eV]	Trigger delay [μsec]	Gate width [μsec]	Accumulation
Gold (Au)	3.88	500	20	8000
Indium (In)	1.44	812	20	8000
Cadmium (Cd)	0.47	1440	30	8000
Off resonance	2.23	660	50	4000

3. Results and discussions

Figure 4 shows the neutron transmission curve of AuIC sample. Figure 5 shows measured images around En=3.88 eV (Au), En=1.44 eV (In), 0.47 eV (Cd) and En=2.23 eV (off-resonance). To avoid saturation of the neutron absorption, the measured energy for Au was shifted from its peak energy (4.89 eV) to 3.88 eV. Even in the off-resonance energy regions we can see the AuIC and the gold sample images because these samples are relatively thick and the cross section values have some finite value in the off-resonance energy regions. The thickness of the In foil was only 10 μm while that of other samples were larger than 0.5mm. According to an estimation, neutron transmittion in the Au foil (0.5mm) is 91% while that in the In foil is 99.98% at En=2.23eV. In order to see the specific elenent in the alloy only, the off-resonance effect in the images has to be eliminated.

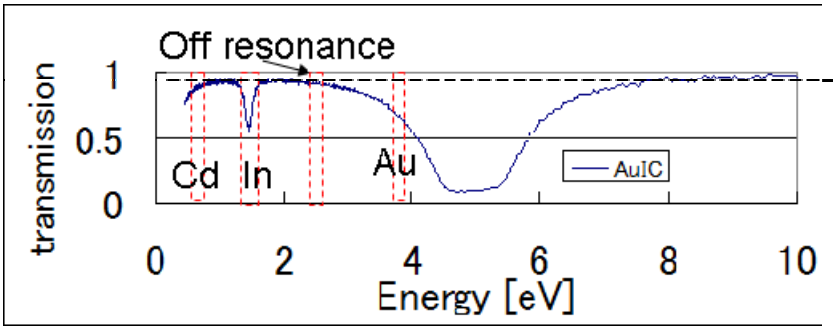

Figure 4. Neutron transmission curve of AuIC sample

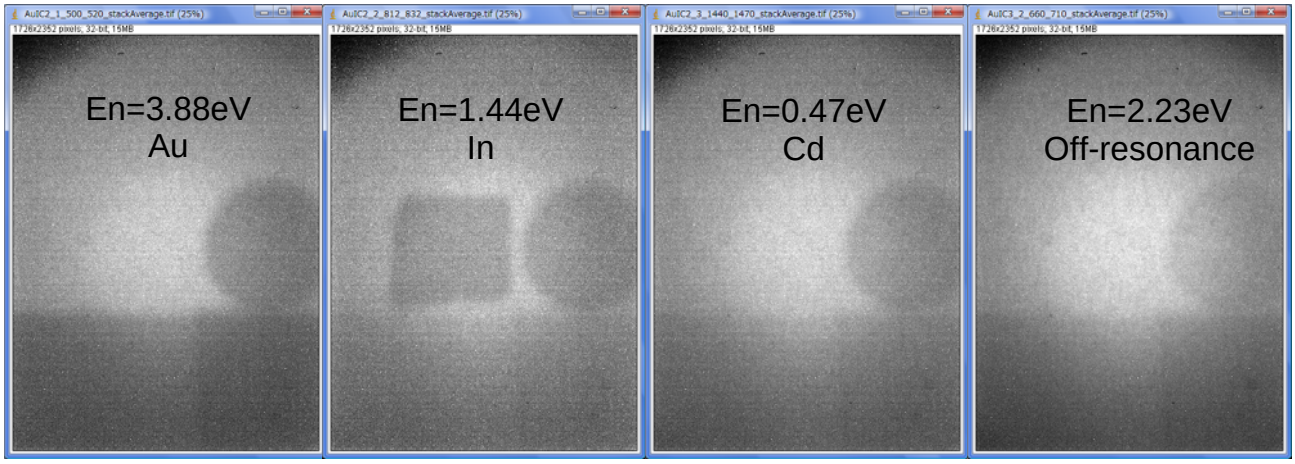

Figure 5. Raw images of the measured. Aiming material and energies are written in the figures.

The resonance peak cross section values of Au and In exceed 10000 barns. The off-resonance cross section of the two nuclides is almost constant at about 30 barns. To eliminate the off-resonance effect, we used the folloing equation.

$$I_r = I_{0r}\exp(-(\sigma_n + \sigma_r)Nt) \quad (1)$$

$$I_n = I_{0n}\exp(-\sigma_n Nt) \quad (2)$$

$$I_{0n} / I_{0r} = C \quad (3)$$

$$-\ln(C\, I_r / I_n) = \sigma_r Nt \quad (4)$$

Where I_r and I_n are transmitted neutron intensity at the resonance energy and off-resonance energy, respectively, I_{0r} and I_{0n} are neutron intensity without a sample, σ_n off-resonance cross section which is considered almost constant over the measured energy region, σ_r resonance cross section, N atomic density, and t thickness of the material. C is a correction factor of the neutron intensity, sintilator efficiency which depends on the neutron energy. Eq. (4) can be deduced from Eqs. (1), (2) and (3) to obtain the function of material thickness t.

Figure 6 shows the function of $\sigma_r Nt$ of gold, indium and cadmium deduced with Eq. (4). In each sub-figure, only the target element is seen. Figure 7 shows intensity profiles along lines A and B in Fig. 6. Fractuation is about 17% at the flat top. Table 2 shows calculated thickness from material composition of the sample and evaluated thickness from the images. The $\sigma_r Nt$ values in the gold and indium samples are almost proportional to their real thicknesses.
We could confirm that the distributions of indium in the AuIC alloy disk sample were almost uniform. These results indicate that the estimation of the thickness is possible in this way.

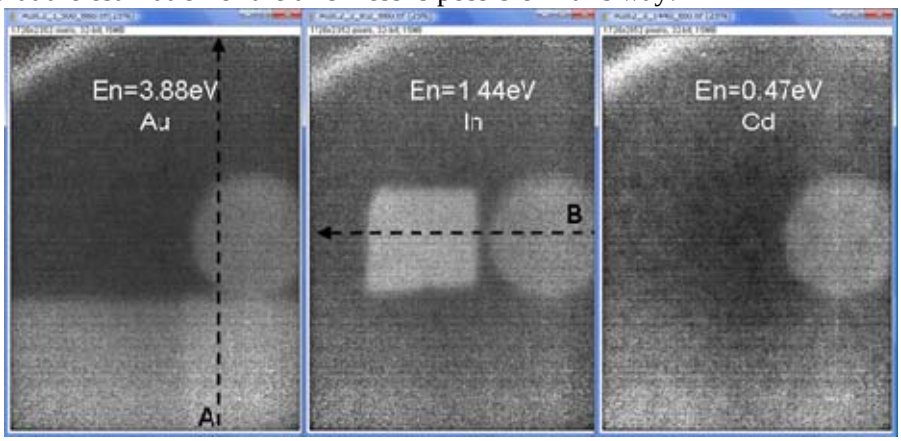

Figure 6. Analyzed images of Au (left), In (center) and Cd (right)

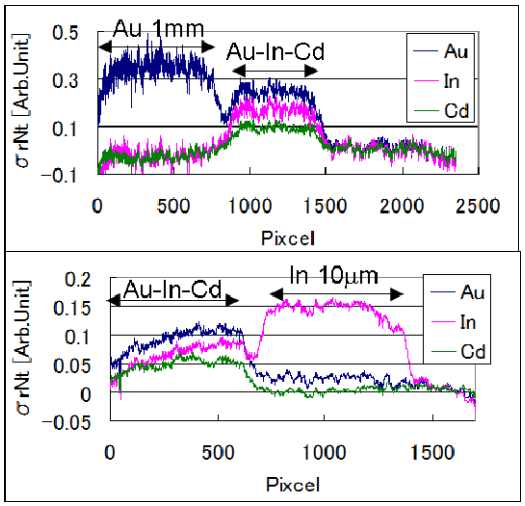

Figure 7. Profile of the analyzed images. Upper figure is line A. Lower figure is line B.

Table 2. Evaluated thickness of the elements in Au-In-Cd specimen.

Material	Calculated	Evaluated
Gold (Au)	1.29 g/cm^2	1.39 g/cm^2
Indium (In)	3.3 mg/cm^2	3.4 mg/cm^2

4. Summary

Elemental distribution of the Au-In-Cd alloy was measured with neutron imaging tecnique. From the difference of the images of the resonance energy and the off-resonance energy, we could get the transmission images of the specific element. From the profile of the image, it is also possible to estimate the thickness. And we confirmed that the elemental distribution of the Au-In-Cd alloy specimen was uniform.
This tecunique can be applied to other materials.

Refarence

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