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Cladding technique for development of Ag–In–Cd decoupler

M. Teshigawara [a,*], M. Harada [a], S. Saito [a], K. Kikuchi [a], H. Kogawa [a], Y. Ikeda [a], M. Kawai [b], H. Kurishita [c], K. Konashi [c]

[a] *Japan Atomic Energy Research Institute, 2-4 Shirakata-Shirane, Tokai-Mura, Naka-gun, Ibaraki-ken 319-1195, Japan*
[b] *High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba-shi, Ibaraki-ken, 305-0801, Japan*
[c] *Oarai Branch, Institute for Materials Research, Tohoku University, Oarai, Ibaraki-ken, 311-0013, Japan*

Abstract

To develop a Ag (silver)–In (indium)–Cd (cadmium) alloy decoupler, a method is needed to bond the decoupler between two plates of the Al alloy (A6061-T6). We found that a better HIP condition was temperature, pressure and holding time at 803 K, 100 MPa and 1 h, respectively, for small test pieces (ϕ 22 mm in diam. × 5 mm in height). Especially, a sandwich case (a Ag–In plate with thickness of 0.5 mm between two Ag–Cd plates with thickness of 1.25 mm) gave easier (or better) bonding results. Though a hardened layer is found in the bonding layer, the rupture strength of the bonding layer is more than 30 MPa, which is higher than the design stress in our application.

1. Introduction

Spallation neutron source facilities in J-PARC [1] (Japan Proton Accelerator Research Complex) project are under construction. Three kinds (coupled, decoupled and poisoned) of hydrogen moderator were adopted [2,3] to provide a pulsed neutron beam with higher neutronic performance as shown in Fig. 1. For the decoupled and poisoned moderators, a thermal neutron absorber, i.e., decoupler as shown in Fig. 2, is located around the moderator to prevent slow neutrons from flowing into the moderator and to give a neutron beam with a short decay time. Higher cut-off energy of the decoupler, which is called decoupling energy (E_d), results in a shorter decay time. The pulsed neutron beam with E_d of 1–2 eV is desired by neutron beam users. A boron carbide (B_4C) decoupler is already utilized for higher E_d which is controlled by the thickness of B_4C; however, it is difficult to use in a MW class source because of helium (He) void swelling and local heating by (n,α) reaction. Therefore, a Ag–In–Cd (AIC, 80Ag–15In–5Cd wt%) alloy which would give energy-dependence of macroscopic neutron cross-section like that of B_4C was chosen [4]. E_d of AIC of about 0.3 cm in thickness reaches at about 1 eV. AIC sheathed with stainless steel is already utilized for the control rod of PWR (pressurized water reactor). However, from a point of view of heat removal and corrosion protection, an AIC plate has to be bonded between two plates of the Al alloy (A6061-T6), which is the structural material of a moderator or reflector. The AIC plate is divided into Ag–In (15 wt%) and Ag–Cd (35 wt%) plates within the solubility limits of In and Cd in Ag in order to extend the life time, which is limited by burn up of Cd. Irradiation effects on Ag–In and

* Corresponding author. Tel.: +81 29282 6217; fax: +81 29282 6496.
E-mail address: teshigawara@linac.tokai.jaeri.go.jp (M. Teshigawara).

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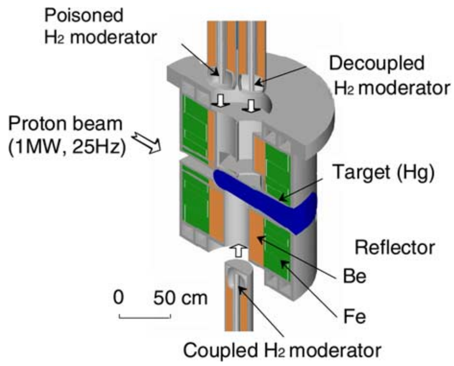


Fig. 1. Cross-sectional view of target–moderator–reflector system for spallation neutron source in J-PARC.

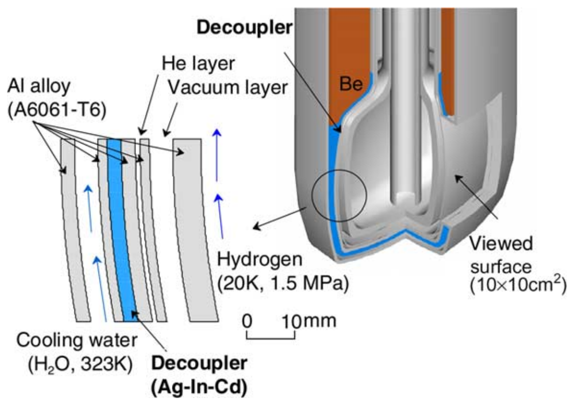


Fig. 2. Structure and location of Ag–In–Cd (AIC) decoupler in the moderator.

Ag–Cd alloys within the solubility limits in Ag were already studied to develop the AIC thermal neutron absorber [5–7] for the control rod of nuclear reactors. We performed bonding tests for the Al alloy, Ag–In, and Ag–Cd by HIP (hot isostatic pressing) to establish the bonding condition. Mechanical tests of the Al alloy and AIC were also performed to determine the basic properties after HIP.

2. Experimental

2.1. Materials

Two kinds of Ag–In–Cd alloy materials were used for the bonding tests by HIP. One is the Ag–In (85Ag–15In wt%) and Ag–Cd (70Ag–30Cd wt%) plates. The other is the Ag–In–Cd (80Ag–15In–5Cd wt%) plate. To make these Ag–In (AI), Ag–Cd (AC) and Ag–In–Cd (AIC) plates, Ag and necessary amount of In and Cd were melted at about 1373 K in an electric furnace and made into a rod with a diameter of 20 mm. The AI, AC and AIC were cut to the plates with the thickness of 2, 2 and 3 mm, respectively. AI/AC or AIC samples was stored in Al alloy (A6061-T6) housing capsule (shape: cylindrical, size: approximately 22 mm in outer diam. × 5 mm in height with thickness of 1 mm) sealed by using electron beam welding, where the evacuated pressure was about 10^{-3} Pa for each of sample. Before storing, samples and Al capsules were polished by an emery paper with up to 800 grit and cleaned in acetone using ultrasonic waves.

2.2. HIPing machine and condition

The HIPing condition is shown in Table 1. We used HIPing machine with the maximum pressure of 200 MPa and the temperature of 2273 K. The AI/AC or AIC sample stored in Al alloy capsules was inserted into the alumina crucible and pressurized up to 50 MPa by argon (Ar) gas with 99.9% purity at room temperature. It was heated up to about 773 K with temperature increasing at a rate of 500 K/h. The pressure was controlled up to 100 MPa with the temperature of HIP treatment. After holding for a certain time, the temperature was reduced to the room temperature with temperature decreasing at a rate of 250 K/h.

2.3. Microstructure observation and mechanical tests

After HIP treatment, we cut and polished the sample to observe microstructures of the interfaces between the Al alloy and AI/AC or AIC and to measure mechanical properties. Vicker's micro hardness tests were conducted with a load of 4.9 N at the outside interface region and 0.49 N at the interface region, respectively, both with a holding time of 15 s. Three-point bending tests were also performed to evaluate the mechanical strength of three kinds of samples (Al alloy matrix, AI–Al alloy interface, and AC–Al alloy interface region) with the volume of approximately $2 \times 2 \times 14$ mm^3. Tensile and shear tests were performed only for samples, having the best bonding results. The test samples including bonding layer

Table 1
HIP treatment condition (temperature, holding time and pressure) for the cases of HIPed Ag–In/Ag–Cd and Ag–In–Cd in the Al housing

Sample	Temperature (K)				
	713	743	773	803	833
Ag–In/Ag–Cd	60 min	60 min	60 min	10, 30 and 60 min	60 min
Ag–In–Cd	–	60 min	60 min	60 min	–

Pressure was fixed at 100 MPa. Increasing rate: 500 K/h, decreasing rate: 250 K/h.

were cut to the cylindrical shape with 5 mm in diam. by using a wire electrical discharge machining.

3. Experimental results

3.1. HIP treatment of Ag–In and Ag–Cd case

Fig. 3 is micrographs showing the HIP temperature dependence for the case of Ag–In (AI)/Ag–Cd (AC). Bonding tests were performed at temperatures from 713 K to 833 K with 100 MPa and 1 h holding time. The sample HIPed at 803 K was the only one successfully bonded. AI and AC were basically bonded up to 803 K in all cases. The sample HIPed at 833 K was melted. In this case the sample temperature might be over 840 K because the eutectic reaction between Al and Ag occurs at 840 K [8]. It was difficult to distinguish the reaction layer of AI and AC from the micrographs. Even the unsuccessful bonding, the reaction layers of both AI–Al alloy and AC–Al alloy were recognized as shown in Fig. 3. The thickness of reaction layers of AI–Al alloy and AC–Al alloy increased with increasing temperature as shown in Fig. 4. This result shows that a relatively thick reaction layer is needed for successful bonding. The thickness of reaction layer of AC and the Al alloy was about 3.5 times thicker than that of AI and Al alloy. In the case of successful bonding at 803 K, the thickness of reaction layer of AI and Al alloy, and AC and Al alloy were about 570 and 175 µm, respectively.

We examined the effect on the HIP treatment of varying the holding time from 10, 30 and 60 min at 803 K.

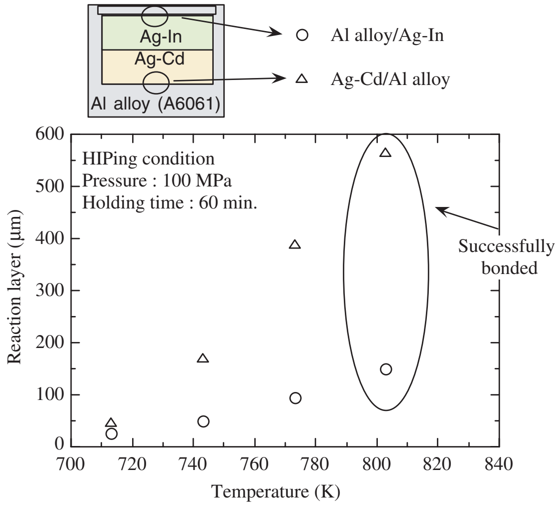


Fig. 4. Thickness of reaction layer after HIP treatment for Ag–In and Ag–Cd in the Al housing as a function of temperature.

Microstructures of HIPed interface with various holding time are shown in Fig. 5. All the samples appeared to be basically bonded. The reaction layer of AI and the Al alloy, and AC and the Al alloy consisted of 2 and 3 phases respectively as shown in Fig. 5. The reaction layer thickened with increasing holding time as shown in Fig. 6. The thickness of reaction layer of AC–Al alloy was also about 3–5 times thicker than that of AI–Al alloy. Better bonding was achieved at the HIP condition of 803 K, 100 MPa and 60 min holding time. The higher temperature (∼840 K) causes melting. The

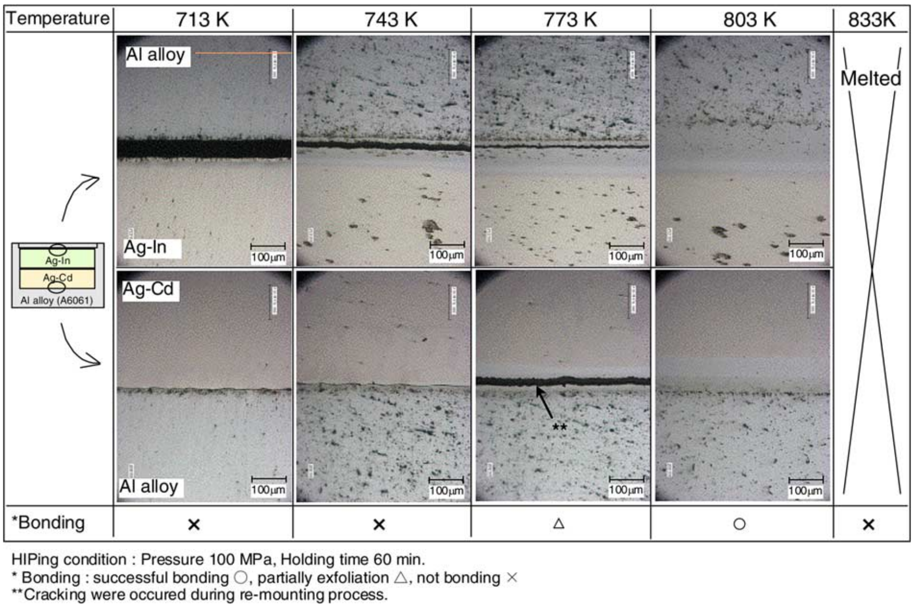


Fig. 3. Micrographs after HIP treatment for Ag–In and Ag–Cd in the Al housing at various temperature.

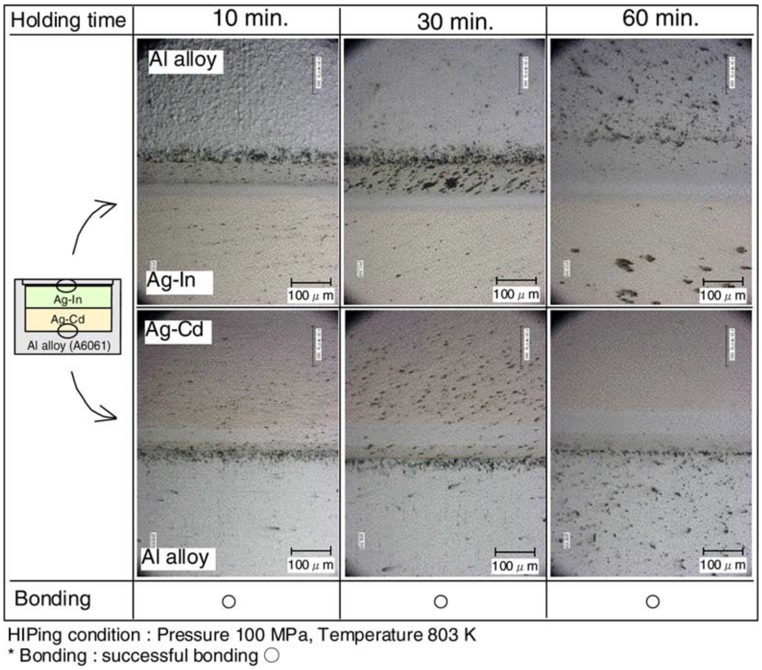


Fig. 5. Micrographs with various holding times after HIP treatment for Ag–In and Ag–Cd in the Al housing.

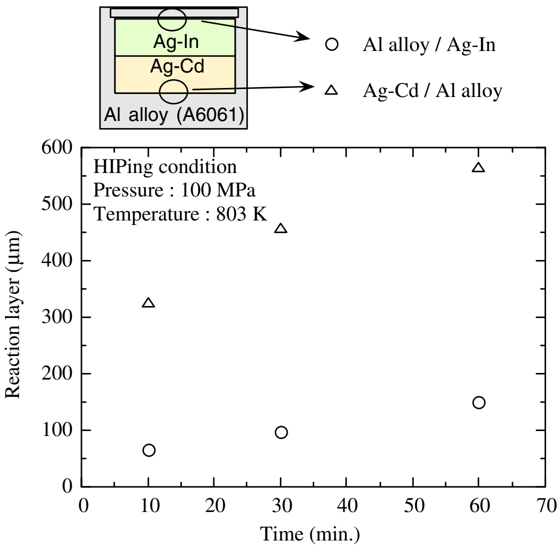


Fig. 6. Reaction layer of HIPed Ag–In and Ag–Cd in the Al housing as a function of holding time.

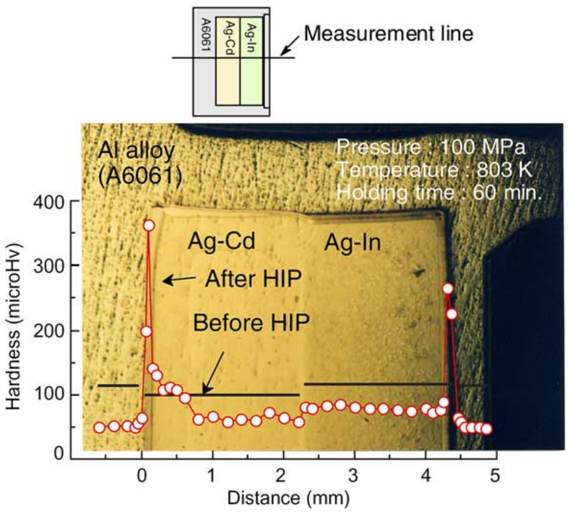


Fig. 7. Hardness profile at the bonding area of HIPed Ag–In and Ag–Cd in the Al housing.

lower temperature does not bring about enough thickness of reaction layer.

3.2. Vicker's micro hardness tests

We performed Vicker's micro hardness tests for the better bonded samples as shown in Fig. 7. The hardest layer could be recognized in each reaction layer of the AI–Al alloy and the AC–Al alloy. Especially, the hardness of the AC–Al interface was larger than that of the AI–Al interface. The other bonding samples showed the similar results. On the other hand, the hardness of HIPed AI, AC and the Al alloy matrix regions was reduced compared to that before HIP (as received), as shown in Fig. 8. The hardness of HIPed each sample

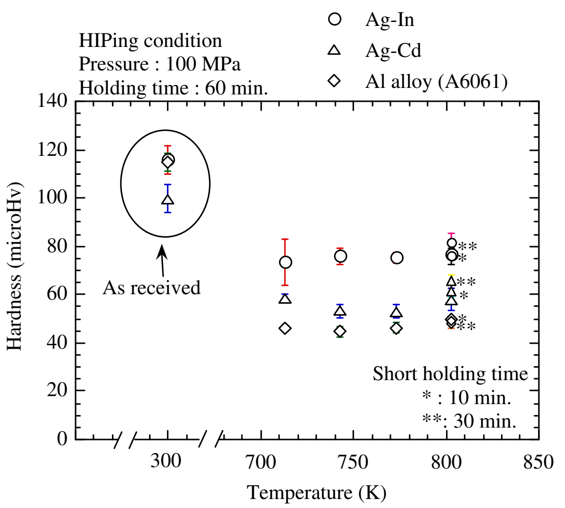


Fig. 8. Hardness measurement of matrix region (Ag–In, Ag–Cd and Al alloy) after HIP treatment for Ag–In and Ag–Cd in the Al alloy housing.

was not very different regardless of the different condition of temperature or holding time.

3.3. Three-point bending tests

The results of three-point bending tests for successfully bonded samples are shown in Fig. 9. Fracture occurred only at the interface between AC and the Al alloy over the yield strengths of AC–Al alloy. As descried above, Vicker's micro hardness of this interface layer was hardest over 350 micro Hv. The hardened layer of this interface shows cracking along the direction of circumference.

3.4. HIP treatment of AIC case

For the reference, we conducted HIP treatment of the AIC alloy under the condition as shown in Table 1. Samples HIPed at 773 and 803 K were bonded as shown in Fig. 10. Temperature dependence of reaction layer thickness was roughly the same as that of AC–Al alloy as shown in Fig. 11. The three-point bending test results in Fig. 12 shows that fracture was not observed.

4. Discussion

4.1. Cause of hardened interface layer

From the Vicker's micro hardness tests and the three-point bending tests, the hardest layer could be recognized in both reaction layers of the AI–Al alloy and the AC–Al alloy and fracture occurred only at the interface between AC and the Al alloy. The hardened layer may be responsible for embrittlement. In a Ag–Al system α phase (fcc) of Ag changes to β phase (bcc) with grater than 20.5 at.% Al [8]. The composition of Al in Ag was measured to be over 20.5 at.% using an energy dispersive X-ray analyzer. Phase changes by diffusion of Al into Ag might result in embrittlement.

4.2. Further improvement of bonding condition

Though a brittle and hardened layer was recognized in the AC–Al alloy, it was easy to make the bonding

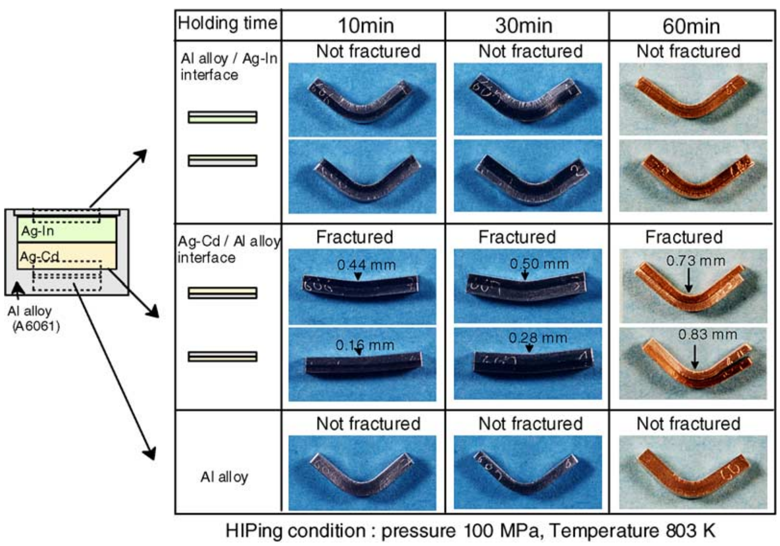


Fig. 9. Three-point bending tests after HIP treatment for Ag–In and Ag–Cd in the Al housing.

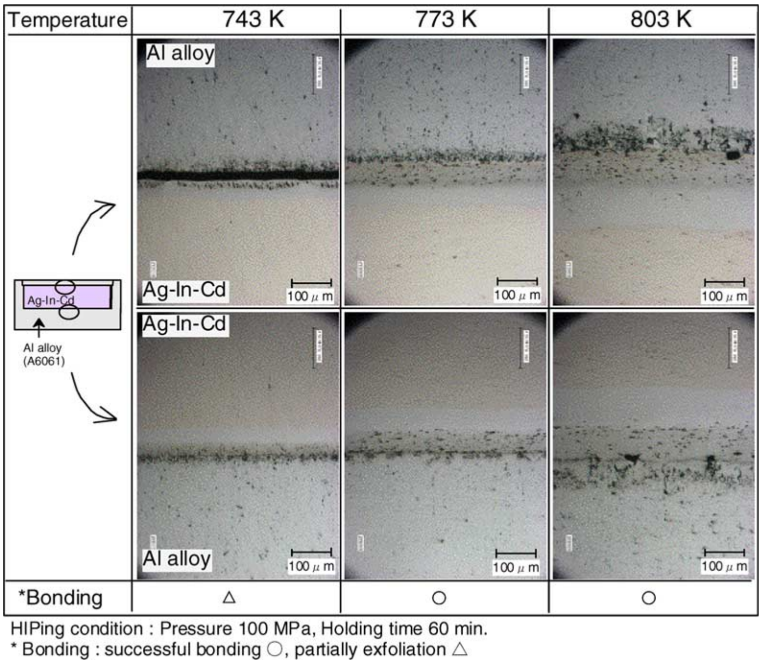


Fig. 10. Micrographs after HIP treatment for Ag–In–Cd in the Al housing at various temperature.

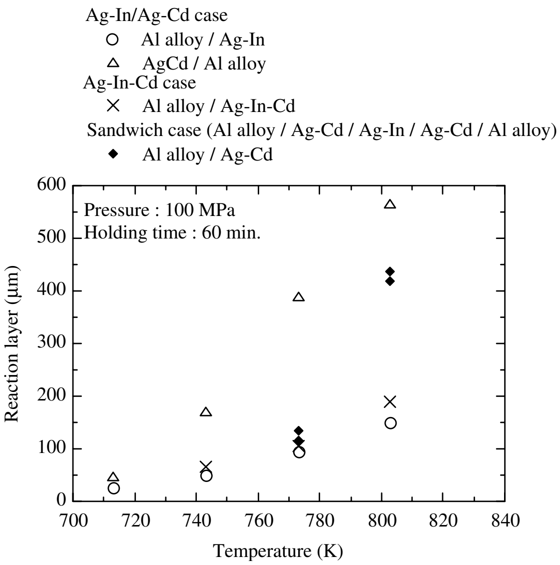


Fig. 11. Thickness of reaction layer after HIP treatment for the cases of Ag–In/Ag–Cd, Ag–In–Cd and Sandwich type (Ag–In inserted between Ag–Cd) as a function of temperature.

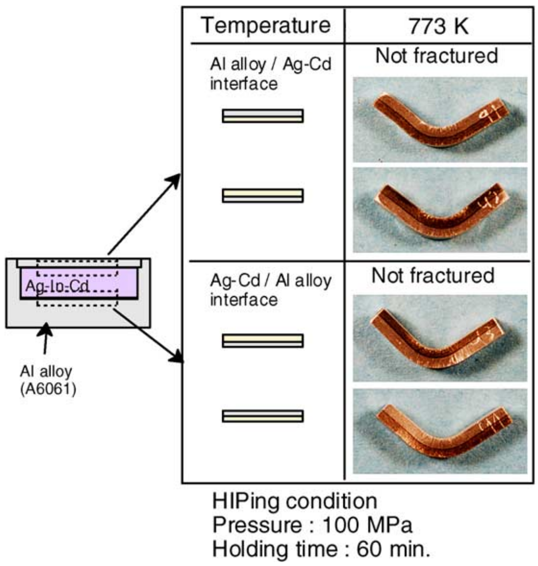


Fig. 12. Three-point bending tests after HIP treatment for Ag–In–Cd in the Al housing.

layer of the AC–Al alloy as compared to that of the AI–Al alloy. AI and AC were always bonded. Therefore, we considered sandwich type (an AI plate inserted between AC plates in an Al container) to seek even better bonding condition. We performed HIP treatment of the sandwich type at 773 and 803 K, at 100 MPa, for 60 min. The thicknesses of AI and AC plates were 0.5 and 2.5 mm in total, respectively, which were optimized so as to satisfy necessary neutronic performance for 6-year operation using neutronic calculation code [9–11]. Both materials

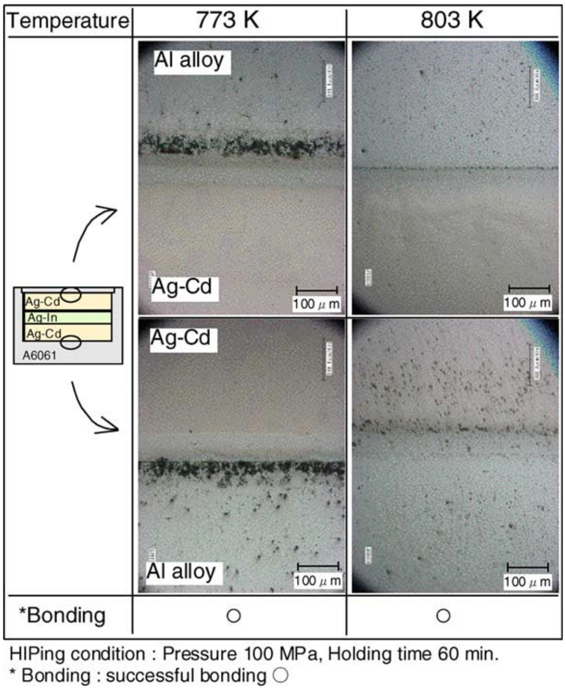


Fig. 13. Micrographs after HIP treatment for sandwich type (Ag–In plate inserted between Ag–Cd plates in the Al housing) at 773 and 803 K.

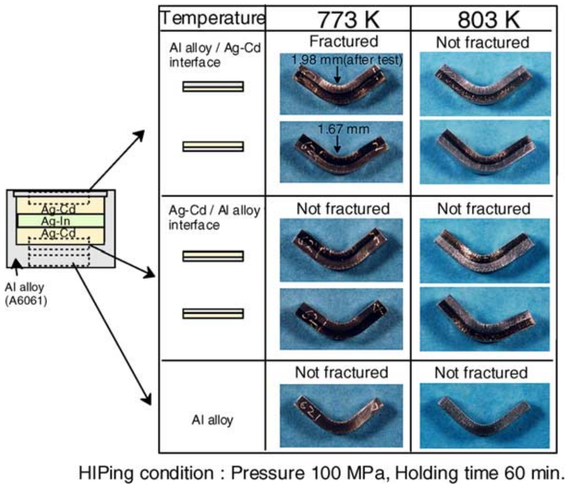


Fig. 14. Three-point bending tests after HIP treatment for sandwich type (Ag–In plate inserted between Ag–Cd plates in the Al housing).

were successfully bonded at 773 and 803 K as shown in Fig. 13. The thickness of the reaction layer of the AC–Al alloy for the sandwich type was relatively thinner than that of the AI/AC case as shown in Fig. 11. Especially, fracture was not observed in the sample HIPed at 803 K as shown in Fig. 14 for three-point bending tests. We performed tensile tests and shear tests to evaluate the mechanical strength of the sandwich type as shown in Fig. 15. Fracture was occurred in the interface layer of the AC–Al alloy with little elongation. The tensile strength was about 30–40 MPa. The shear strength of interface layer of the AC–Al alloy was about 80–100 MPa. These values were over the design stress of decoupler region, which were evaluated by ABAQUS, a finite element method (FEM) calculation code.

Easier bonding combination of AC and the Al alloy rather than that of AI and the Al alloy provided an opportunity of getting better bonding combination. We found that better bonding condition was achieved

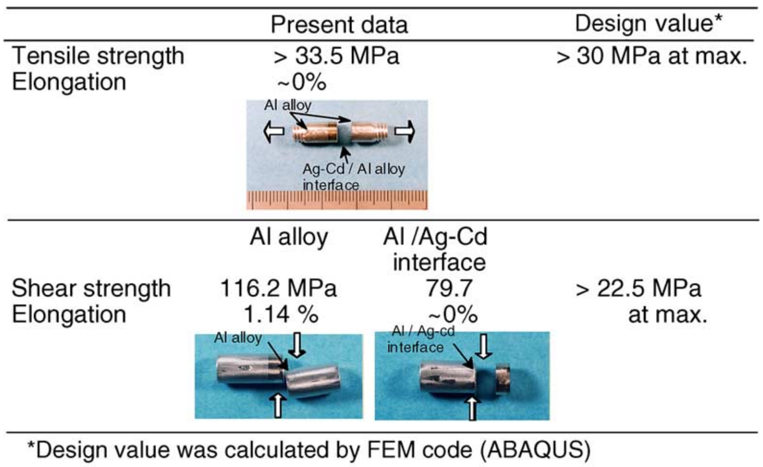


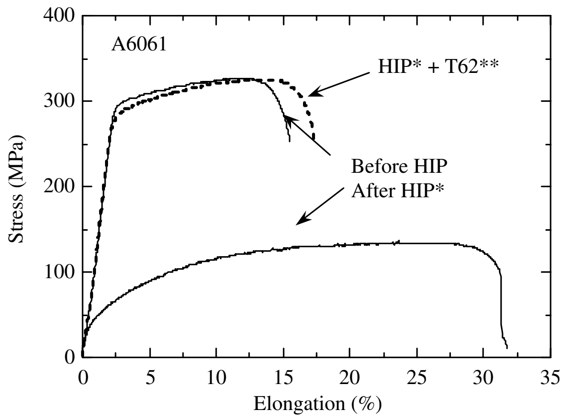
Fig. 15. Tensile and shear strength tests after HIP treatment for sandwich type (Ag–In plate inserted between Ag–Cd plates in the Al housing).

for the sandwich type, AC–AI–AC (AI was inserted between AC) at 803 K, 100 MPa with 1 h holding time. This HIPing condition for the sandwich type already satisfies required design stress. There is a possibility of getting bonding material with even higher strength by selecting another sandwich type, AI–AC–AI (AC is inserted between AI). From three-point bending tests the fracture in fractured samples always occurred at the AC–Al interface layer. Obtaining a good bonding of between AI–Al alloy in AI–AC–AI could give a bonding material with high strength in comparison with AC–AI–AC. However, it will not make bonding easily compared to the Al alloy and AC.

From a point of view of the fabrication and irradiation effect, it is desirable to minimize the number of bonding layer, because the sandwich type thermal neutron absorber gives more complicated structures to make curved shape of Al moderator vessel, which contains three layers of thermal neutron absorbers. Irradiation effects of matrix of AI and AC have already been revealed, however, there is no data on the irradiation effects of interfaces between bonding layers, especially between the Al alloy and AC or AI. Such engineering issues remain to be studied. R&D efforts should be under progress.

4.3. Improvement in the strength of Al alloy by heat treatment

HIP treatments affected the strength of Al alloy. The strength of the Al alloy (A6061-T6) was reduced about 1/3 after HIP treatment as shown in Fig. 16. This tensile test did not include the Ag–In–Cd alloy. The reduced strength affects the neutronic performance of moderator because thicker Al alloy is needed due to the required strength. We performed heat treatments called T62 to increase the strength. The strength was almost recovered as shown in Fig. 16, indicating that the heat treatment is very effective in recovering the reduced strength. The heat treatments including Ag–In–Cd alloy are in progress after HIP.



* : Pressure : 100 MPa, Temp. : 773 K, Holding time : 60 min.
** : Holding time :30 min. at 788K, water quentching, Holding time : 18 hr. at 438 K

Fig. 16. Improvement in the strength of Al alloy (A6061) by heat treatment, T62, after HIP treatment.

5. Conclusions

We performed bonding tests between the Al alloy (A6061-T6), Ag–In, and Ag–Cd by HIP treatment to establish the bonding condition.

A better bonding condition was achieved for the sandwich type (AI was inserted between AC) at 803 K and 100 MPa with 1 h holding time. Reaction layer between the Al alloy and AI or AC caused embrittlement, the mechanical strength of bonding layer of the Al alloy and AC was over the design stress. It is possible that we can get bonding materials with higher strength by selecting another sandwich type (AC is inserted between AIs).

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