Microstructural Characterization of Intermediate Layer Produced at Aluminum/Metallic Glass Interface Fabricated by Magnetic Pulse Welding

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Lap joining of crystalline pure aluminum and metallic glass (Zr₄₈Cu₃₆Al₈Ag₈ and Cu₅₀Zr₄₂.₅Al₇.₅) was carried out by using magnetic pulse welding. Transmission electron microscopy was performed for the welding interface in order to investigate microstructure of the intermediate layer produced at the welding interface. The welding interface exhibited characteristic wavy morphology as well as similar- and dissimilar-metal joints. The intermediate layer was produced along the wavy interface. TEM observation and electron diffraction pattern analysis revealed that the intermediate layer was not monolithic but had a multi-phase structure. A part of the layer consisted of the extremely fine grains and amorphous phase particles. The other part of the layer was composed of the stacked extremely thin amorphous layers. STEM-EDS analysis found that the chemical composition of the amorphous phase in the intermediate layer was not constant and was different from that of the metallic glass matrix.

Keywords: magnetic pulse welding, transmission electron microscopy, welding interface, intermediate layer, aluminum, metallic glass

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1. Introduction

Magnetic pulse welding is one of the solid-state welding processes, in which high-speed collision between two or multiple metal plates is utilized for lap welding. The impact energy is induced by electromagnetic force which is generated by interaction among discharge pulse current running through the coil, induced magnetic flux around the coil, and eddy current produced at the plate surface. The welding is normally achieved within 10 microseconds with a negligible temperature increase. This welding process can be applied to metal combinations widely differing in physical and mechanical properties. Sound metallurgical bonding can be obtained in a wide variety of similar and dissimilar metals and alloys without any external application of heat or the use of any inserted intermediate metals.¹)

Metallic glasses are known to have many unique mechanical and functional properties which are difficult to obtain with conventional crystalline alloys.²⁻⁴) This is attributed to the random atomic arrangement like those in a liquid. Recently, some bulk metallic glass with several tens millimeters thickness were produced directly from the melt with low cooling rates of the order of 10⁶ to 10⁷ K/s.²⁻⁵⁻⁷) The metallic glasses with such properties are worth for engineering materials. Therefore, weldability of the metallic glass to other engineering materials such as metals and ceramics is of great interest when we seek the usage of the metallic glass. Few studies have been done on the welding of metallic glasses to commercial alloys. Kawamura et al. investigated the electron beam welding of Zr and Ti to Zr-based metallic glasses, and defect-free joints with bending strengths higher than that of the crystalline metal matrix were obtained.⁸⁻¹⁰) Wang reported that strong Al alloy/Zr-based metallic glass butt joint was obtained by friction stir welding and no reaction layer was observed at the welding interface.¹¹) A part of the present authors performed magnetic pulse welding of pure Al plate to Zr-based and Cu-based metallic glasses.¹²) The metallic glasses were successfully magnetic pulse welded to the pure Al plate with no crystallization of the metallic glass.

In the present study, transmission electron microscopy was carried out for investigating microstructure of the welding interface. Based on the obtained results, formation mechanism of characteristic interfacial microstructure was discussed.

2. Experimental Procedures

2.1 Materials

A 0.5 mm thick crystalline pure aluminum plate (99.50 mass% Al) and two kinds of 50 µm thick metallic glass foils (Zr₄₈Cu₃₆Al₈Ag₈ alloy and Cu₅₀Zr₄₂.₅Al₇.₅ alloy) were prepared for the present study. The metallic glass foils were produced by rapid roll quench method. The formation of glassy structure in the Zr₄₈Cu₃₆Al₈Ag₈ alloy and the Cu₅₀Zr₄₂.₅Al₇.₅ alloy was confirmed by using X-ray diffraction. Surface of the samples subjected to the lap welding was cleaned with alcohol and dried.

2.2 Magnetic pulse welding

Figure 1 shows conceptual diagram of the impact welding method using electromagnetic force. A discharge circuit as shown in Fig. 1(a) is used for the magnetic pulse welding. The circuit consists of a capacitor for a supply of electrical energy, a discharge gap switch, and a special E-shaped one-turn flat coil. Two plates which have a little gap are set over the coil. The plate close to the coil is termed “flyer plate” and another plate above it, which is fixed firmly in place, is referred to as “parent plate”.

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The principle of generation of electromagnetic force is explained by using Fig. 1(b) which shows a close-up around a middle section of the coil. When a discharge pulse passes through the coil, high density magnetic flux lines are generated around the coil. The generated magnetic flux lines intersect with the flyer plate, and eddy currents are excited in the flyer plate surface adjacent to the coil in accordance with the principle of Lentz’s law. Since the discharge pulse is high-frequency, the eddy currents are generated at the plate surface due to the so-called surface effect. The eddy currents and the magnetic flux lines induce an electromagnetic force upward in accordance with the principle of Fleming’s left-hand rule. The generated electromagnetic force drives the flyer plate to the parent plate at a high velocity and the collision takes place between two plates. Metal plates with high electrical conductivity such as aluminum and copper are suitable for the flyer plate because they can generate larger electromagnetic force. In the present study, the aluminum plate was used as the flyer plate and the metallic glass foils were used as the parent plates.

In the previous study, when the plate layout, as shown in Fig. 1(b), was used for welding, the metallic glass fractured in brittle manner. However, we could prevent the metallic glass from such brittle fracture by using an aluminum plate as a shock absorber, as shown in Fig. 2. In the present study, the plate layout as shown in Fig. 2 was used. The initial gap between the plates was fixed to be 1.0 mm. The discharge energy was 2.5 kJ.

3. Results

3.1 Welding interface of lap joints

Figures 3(a) and 3(b) show backscattered electron images of welding interface in the aluminum/Zr48Cu36Al8Ag8 and the aluminum/Cu50Zr42.5Al7.5 joint, respectively. The upper bright-contrast area is metallic glass and the lower dark-contrast area is aluminum. The welding interface exhibited characteristic wavy morphology, as well as similar and dissimilar metal joints. A thin intermediate layer was also produced along the wavy interface. In the intermediate layer, several shadings of medium contrast between the aluminum and the metallic glass were observed. This indicates that the intermediate layer was not monolithic but had a multi-phase structure.

3.2 Microstructure of intermediate layer produced at welding interface

The thin intermediate layer with several shadings of gray contrast was clearly observed at the welding interface, as
shown in Fig. 3. In order to investigate microstructure of the intermediate layer, TEM observation and STEM-EDS analysis were carried out. The intermediate layer consists of two kinds of characteristic microstructure. The obtained results are shown as follows.

### 3.2.1 Fine grains and amorphous phase structure

Figure 4(a) shows a bright-field image of welding interface of the aluminum/Zr₄₈Cu₃₆Al₈Ag₈ joint. The upper dark-contrast area is Zr₄₈Cu₃₆Al₈Ag₈ metallic glass and the lower bright-contrast area is aluminum. The produced intermediate layer is sandwiched between them and located at the central position of the figure. Evidence of the crystalline such as dislocations and fringe was not observed in the Zr₄₈Cu₃₆Al₈Ag₈ matrix close to the welding interface. Figure 4(b) shows a schematic illustration of Fig. 4(a). Circles 1 to 4 in Fig. 4(b) indicate areas, from which we took selected area diffraction (SAD) patterns. The diffraction pattern taken from the area in the Zr₄₈Cu₃₆Al₈Ag₈ metallic glass matrix close to the intermediate layer consisted of only a diffuse halo ring, as shown in Fig. 4(c). This indicates that the Zr₄₈Cu₃₆Al₈Ag₈ metallic glass matrix kept the original amorphous structure after the welding. Figure 4(d) shows the diffraction pattern taken from the aluminum matrix close to the intermediate layer. This pattern was indexed as shown in Fig. 4(e), and the lattice constant calculated from this pattern corresponded to that of aluminum ($a = 0.40496$ nm). Two kinds of diffraction pattern were taken from the intermediate layer. One pattern was a halo ring, as shown in Fig. 4(f). The other pattern was a ring pattern which consisted of sharp spots, as shown in Fig. 4(g). The diameter of the ring pattern was measured, and the lattice constant was calculated. The ring pattern was determined to be the Debye ring of aluminum, as shown in Fig. 4(h). However, the lattice constant calculated from the ring pattern was larger several percent than that of the aluminum. These experimental results indicate that the intermediate layer produced at the welding interface consists of extremely fine aluminum grains and the amorphous phase.

In order to investigate chemical composition of the intermediate layer, STEM-EDS analysis was carried out. The analysis was performed for twenty points as indicated in Table 1(a). The positions of 1–10 are located in the amorphous phase and the positions of 11–20 are in the fine...
aluminum grains region. The obtained results are shown in Table 1(b). The chemical composition of dark-contrast area (position: 3–5) in the amorphous phase was Zr-rich. On the other hand, the chemical composition of bright-contrast area (position: 1, 2, and 6–10) was Al-rich. The atomic percentages of Zr, Cu, and Ag elements of the amorphous phases were lower than that of the metallic glass matrix. On the other hand, the atomic percentage of Al element was higher than that of the metallic glass matrix. In the region of fine aluminum grain (position: 11–20), the chemical composition was Al-rich but elements of Zr, Cu, and Ag were also detected. However, the diffraction pattern taken from this region did not contain spots of Zr, Cu, and Ag, as shown in Fig. 4(d). Therefore, the fine grains in the intermediate layer were considered to be Al–Zr–Cu–Ag solid solution. These experimental results revealed that a part of the intermediate layer produced at the welding interface consists of extremely fine grains of Al–Zr–Cu–Ag solid solution and Zr-based and Al-based amorphous phase.

3.2.2 Multiple amorphous layer structure

Figure 5(a) shows a bright-field image of welding interface in the aluminum/Cu50Zr42.5Al7.5 joint. The upper dark-contrast area is Cu50Zr42.5Al7.5 metallic glass and the lower bright-contrast area is aluminum. The intermediate layer is located between them. Extremely thin layers with different shadings were observed in the intermediate layer. Figure 5(b) shows a schematic illustration of Fig. 5(a). Circles 1 to 3 in Fig. 5(b) indicate the areas, from which we took SAD patterns. As well as the aluminum/Zr48Cu36Al8Ag8 joint, the diffraction pattern taken from the area in the Cu50Zr42.5Al7.5 metallic glass matrix close to the intermediate layer consisted of only a diffuse halo ring, as shown in Fig. 5(c), indicating that the Cu50Zr42.5Al7.5 metallic glass matrix also kept the original amorphous structure after welding. Figure 5(d) shows the diffraction pattern taken from the aluminum matrix close to the intermediate layer. This pattern was indexed as shown in Fig. 5(e), and the lattice constant calculated from this pattern corresponded to that of

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Fig. 4 TEM image of welding interface. (a) Bright-field image of welding interface. IML indicates intermediate layer. (b) Schematic illustration of (a). Circles indicate region which took diffraction pattern. (c) SAD pattern taken from region 1 in (b). (d) SAD pattern taken from region 2 in (b). (e) Key diagram to (b). Z = [112]. (f) SAD pattern taken from region 3 in (b). (g) SAD pattern taken from region 4 in (b). (h) Key diagram to (g).
aluminum. The diffraction pattern taken from the thin layers consisted of only diffuse halo ring, as shown in Fig. 5(f). A high-resolution TEM image of the interface between the aluminum and the intermediate layer which consisted of thin layers was shown in Fig. 6. The upper area is the intermediate layer and the lower area is the aluminum. The typical amorphous pattern was observed in the intermediate layer and no obvious fringe contrast corresponding to the crystalline structure can be observed. This indicates that the extremely thin layers produced in the intermediate layer were amorphous phase.

Figure 7 shows chemical composition of the amorphous layers produced in the intermediate layer. The chemical composition analysis was performed along the line indicated by an arrow in Fig. 7(a). The composition profile is shown in Fig. 7(b). The right-hand side is Cu50Zr42.5Al7.5 metallic glass and the left-hand side is aluminum. The area between the broken lines corresponds to the thin amorphous layers region. In the area of 50 nm and more away from the interfaces of amorphous layer/matrixes, chemical compositions were equivalent to those of the matrixes. On the other hand, chemical compositions of the amorphous layers were not constant and indicated several ratios of Al, Cu, and Zr elements. Both Zr-rich amorphous and Al-rich amorphous were produced, and the producing location of the each layer was random. These chemical compositions were different from that of the original metallic glass matrix. These experimental results revealed that a part of the intermediate layer produced at the welding interface consists of extremely thin amorphous layers.

4. Discussion

4.1 Formation mechanism of intermediate layer
The intermediate layer was produced along the wavy welding interface, as shown in Fig. 3. TEM observation and STEM-EDS analysis revealed that the intermediate layer consisted of extremely fine aluminum solid-solution grains and particles or extremely thin layers of amorphous phase which had different composition from that of the metallic glass matrix.

The welding interface exhibited characteristic wavy morphology as shown in Fig. 3, as well as similar and
dissimilar metal interface.\textsuperscript{1,13,14} There is considered to be a
high-speed oblique collision took place between the bulged aluminum plate and flat metallic glass and this collision process is considered to result in formation of the wavy interface.

In the magnetic pulse welding, the high-speed oblique collision takes place and the welding is achieved between the two plates. In order to understand the formation mechanism of the intermediate layer produced at the welding interface, it is necessary to know temperature generated by the high-speed oblique collision. Temperature change at the welding interface was investigated by numerical simulation using Euler–Lagrange coupling software.\textsuperscript{26} This report indicated that the maximum temperature at the welding interface was in the range from 320 to 430 K at the collision velocity of 100–500 m s\(^{-1}\) and the collision angle of 0.5–10 degree. This temperature is not enough to melt the aluminum (\(T_\text{mel}=933\) K) and the metallic glass (\(T_\text{f}=1143\) K (Zr\(_{48}\)Cu\(_{36}\)Al\(_{8}\)Ag\(_{8}\)),\textsuperscript{7}) 1149 K (Cu\(_{50}\)Zr\(_{42.5}\)Al\(_{7.5}\).\textsuperscript{27}) In addition, the melting point of metals increases with increasing pressure. The present authors estimated the collision pressure by using the travelling velocity of the flyer plate, which investigated by using electrical signals.\textsuperscript{13} It is found that the collision pressure was about 1 GPa at the magnetic pulse welded joint interface. The melting point of the aluminum increases up to 988 K at the pressure of 1 GPa.\textsuperscript{28} It is higher than the maximum temperature generated at the welding interface (430 K).\textsuperscript{29} For the metallic glasses, glass transition temperature (\(T_\text{g}\)) of 690 K (Zr\(_{48}\)Cu\(_{36}\)Al\(_{8}\)Ag\(_{8}\)),\textsuperscript{29} 701 K (Cu\(_{50}\)Zr\(_{42.5}\)Al\(_{7.5}\))\textsuperscript{27} is also higher than the maximum temperature generated at the welding interface (430 K).\textsuperscript{29} Consequently, the both matrixes of the aluminum and the metallic glasses close to the welding interface are considered to be solid-state during the welding process.

The mechanical alloying (MA) is solid-state powder processing technique involving repeated cold welding and fracture of powder particles in a high-energy ball mill. This process results in equilibrium and non-equilibrium phases such as supersaturated solid solutions, metastable crystalline, quasicrystalline phases, intermetallic compounds, nanostructures, lamella structure, and amorphous phase.\textsuperscript{30–33} As shown in Figs. 4 and 5, the intermediate layer consisted of fine grains of aluminum and particles or layers of amorphous phase. This characteristic microstructure is quite similar to the mechanical alloyed microstructure. Amorphous phase produced by the MA requires long milling time (about several ten hours). Process time of the magnetic pulse welding is within 10 \(\mu\)s and it is extremely shorter than the milling time in the MA for producing of the amorphous phase. It is considered that extremely high energy is considered to result in formation of amorphous phase during several microseconds at the magnetic pulse welded interface.

Generally, amorphous phase is produced at eutectic composition of the alloy by rapid quenching. The quaternary eutectic composition of Zr–Cu–Al–Ag system and the ternary eutectic composition of Zr–Cu–Al system are 48 at\%Zr–36 at\%Cu–8 at\%Al–8 at\%Ag\textsuperscript{3} and 50 at\%Zr–40 at\%Cu–10 at\%Al,\textsuperscript{34} respectively. The Zr\(_{48}\)Cu\(_{36}\)Al\(_{8}\)Ag\(_{8}\) alloy is a quaternary eutectic alloy but the Cu\(_{50}\)Zr\(_{42.5}\)Al\(_{7.5}\) alloy is not ternary eutectic alloy. The chemical composition of the amorphous phases in the intermediate layer differed from that...
of the metallic glass matrix, as shown in Table 1 and Fig. 7, and did not correspond to the eutectic composition. The Zr–Al–Cu–Ag and Al–Zr–Cu–Ag amorphous phase were produced at the aluminum/Zr₈₆Cu₃₀Al₀₅Ag₆ interface and the Zr–Cu–Al, Zr–Al–Cu, Al–Zr–Cu, and Al–Cu–Zr amorphous phase were produced at the aluminum/Cu₃₀Zr₁₂.₅Al₁₇.₅ interface. The amorphous phases having the chemical composition except the eutectic composition were produced by MA process. These reports are considered to support that MA behavior occurred at the magnetic pulse welded joint interface.

4.2 Position dependency of microstructure of intermediate layer

TEM observation and STEM-EDS analysis revealed that the intermediate layer consists of two kinds of characteristic microstructure. A part of the layer consisted of the extremely fine aluminum solid-solution grains and amorphous phase particles. The other part of the layer was composed of the stacked extremely thin amorphous layers.

At the magnetic pulse welded joint interface, the welding interface exhibited wavy morphology. Wavelength and amplitude of the interfacial wave were not uniform and gradually changed through the welding interface. The wavelength increased with increasing distance from the vertical centerline of the bulged region. The amplitude once increased toward outside, but it showed the maximum value at a certain position and then decreased. These gradual changes in the wavelength and the amplitude are considered to be due to the gradual change in oblique collision angle. These characteristics are common for the magnetic pulse welded lap joints including the present aluminum/metallic glass joint. The intermediate layer is produced through the welding interface. As well as change in the magnitude of the interfacial wave, the producing microstructure in the intermediate layer is also considered to change depending on the position of the welding interface, i.e., depending on the magnitude of the interfacial wave.

As mentioned in Section 4.1, the intermediate layer is considered to be produced by the MA process. The microstructure produced by the MA process changes depending on the MA parameter such as milling time and energy dose. Dougherty investigated microstructural evolution of Al–Ni–Fe–Gd alloy by changing the milling time. The microstructure of the sample milled at 2 h was lamella structure which consisted of Al layer, Gd layer, and Al–Gd amorphous layer including fine Ni and Fe particles. Then, Al–Gd amorphous matrix including fine crystal grain was produced with increasing milling time. After 80 h of ball milling, the powders were fully amorphous. Fadeeva reported that effect of energy dose on producing microstructure of Al–Fe alloy using the mechanical alloying. The Al–Fe solid solution was produced at the energy dose of 20 and 70 kJ/g. At the energy dose of 130 kJ/g, full amorphous was obtained. In the MA process, increase of the energy dose is considered to be equivalent to increase of the milling time because the input energy increases with increasing milling time. Consequently, amorphous phase is produced entirely with increasing milling time, i.e., increasing reaction time, in the MA process.

As mentioned above, magnitude of the interfacial wave is gradually changed through the welding interface and the wavelength of the interfacial wave increases with increasing distance from the vertical centerline of the bulged region. Miyazaki investigated travelling velocity of the collision point using numerical simulation. They reported that the collision point velocity decreased with increasing distance from the vertical centerline of the bulged region. In other words, the wavelength of the interfacial wave increases with increasing travelling velocity of the collision point. The reaction time at the welding interface decreases with increasing travelling velocity of the collision point. Therefore, it is assumed that the intermediate layer produced at smaller wavy interface is produced at shorter reaction time and the intermediate layer produced at larger wavy interface is produced at longer reaction time. Because amorphous phase is produced entirely with increasing reaction time in the MA process, it is considered that the intermediate layer produced at larger wavy interface, which located at the outer interface, consisted of amorphous layers (Fig. 5) and the intermediate layer produced at smaller wavy interface, which located at the inner interface, consisted of fine aluminum solid solution grains and amorphous phase (Fig. 4).

5. Conclusions

Microstructure of the intermediate layer produced at magnetic pulse welded aluminum/metallic glass interface was examined and following findings were obtained.

(1) The welding interface exhibited characteristic wavy morphology, as well as that of the metal/metal interface. A thin intermediate layer was produced along the wavy interface.

(2) TEM observation and analysis revealed that the intermediate layer consisted of extremely fine aluminum solid solution grains and extremely thin multiple layers of amorphous phase which had different composition from that of the metallic glass matrix.

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