Fabrication of Fe-Based Metallic Glass Particle Reinforced Al-Based Composite Materials by Friction Stir Processing

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Fe72B14Si9Nb4 metallic glass particles and pure Fe particles were separately dispersed into the pure aluminum by the friction stir processing. The microstructure and mechanical properties of the stir zone were analyzed using XRD, SEM, TEM and Vickers hardness tests. As a result, it was found that the Fe metallic glass and pure Fe particles can be uniformly dispersed into the pure aluminum by friction stir processing and the mechanical properties of the stir zone can be improved due to the formation of precipitates. [doi:10.2320/matertrans.M2011094]

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1. Introduction

Friction stir welding (FSW) is a welding technique that can plastically deform metallic materials and soften the materials around the probe due to the heat generated by the friction between the rotating tool and work-pieces. The combination of tool rotation and translation results in the movement of material from the front to the back of the probe, thereby producing a welded joint in the solid state. This method can be used to refine the microstructure and improve the mechanical properties of the welded material by heat treatment and severe plastic deformation. Friction stir processing (FSP) is a new technique for microstructural modification and was developed based on the basic principles of FSW. Recently, the surface modification of materials by FSP has been intensively studied. It was reported that the fabrication of metal matrix composites (MMCs) uniformly dispersed with ceramic, fullerene, SiC and carbon nanotube particles has been successfully developed using the FSP. Since the process is performed below the melting point of the materials, FSP has many advantages when fabricating MMCs compared to other melting methods. With the rapid development of bulk metallic glasses, metallic glass particles are attractive as reinforcement and have been successfully used in the fabrication of Al-based MMCs because of their remarkable mechanical properties including high yield strength and large elastic strain. In addition, they may yield an improved interface between the matrix and particles with respect to the conventional ceramic reinforcement.

In this study, the fabrication of an Al-based composite dispersed with Fe-based metallic glass particles was attempted using the FSP method. The welding temperature of the Al alloy is between 400°C and 450°C during the FSW. Accordingly, the Fe-based metallic glass particle was used because its glass transition temperature is higher than the welding temperature of the Al alloy and the oxidation does not easily occur. The base material was pure Al plates (1050-H24). For comparison, the fabrication of Al-based composite dispersed with pure Fe particles was also attempted using FSP.

2. Experimental Procedure

In this study, the base material was pure Al plates (1050-H24) with the dimensions of 300 × 70 × 5 mm³. The Fe72B14Si9Nb4 metallic glass particles with average size of 45 μm and pure Fe particles with average size of 150 μm were used for the dispersion purpose. The tools usually consist of a shoulder with a large diameter and a probe with a small diameter, which is shown in Fig. 1(c). The tool used in this study was made of SKD61 and had a shoulder of 15 mm diameter, a probe with 6 mm diameter and 4.3 mm length with a 10° recessed shoulder surface. The probe was the screw-type and the tool was tilted by 3° during the process. For the dispersion process, a gap was intentionally formed by placing 2 mm-thick shims between the two pure Al plates before the FSP was performed. Then, Fe-based metallic glass particles or pure Fe particles can be separately filled into the gap, as shown in Fig. 1(a). In order to prevent the particles from flying off and to increase the filling rate of the powder, the FSP was first performed with only the shoulder (without probe), as shown in Fig. 1(b). By this means, the particles can be sealed beneath the sample surface by the metallic materials. Next, the FSP was repeated using the same welding conditions, however, with rotation tools having a 4.3 mm long probe, as shown in Fig. 1(c). This is called the first pass processing. In order to distribute the

Fig. 1 Schematic illustration of friction stir processing. (a) filling powder into gap; (b) without probe; and (c) with probe (1st or 2nd pass).
particles more uniformly in the stir zone, the FSP under the same condition with that of the first pass processing was carried out again and is called the second pass processing. After the FSP, the phase identification and microstructures of the dispersed specimens were examined by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM) and electron back-scattering diffraction (EBSD). The XRD analysis was conducted (Bruker, AXS D8 DISCOVER) using Co-Kα radiation. The stir zone of the dispersed specimen was observed by SEM (Hitachi SU-70). The samples for the TEM observations were prepared using twin-jet electrolytic polishing in a solution with a mixed ratio of \( \text{HNO}_3 : \text{CH}_3\text{OH} = 7 : 3 \). The TEM observations were carried out using a Hitachi H800T microscope at 200 kV. In addition, the samples for the EBSD observations were prepared using electrolytic polishing in a solution with a mixed ratio of \( \text{HNO}_3 : \text{CH}_3\text{OH} = 7 : 3 \). The Vickers micro-hardness (HV) was measured using a digital tester (Akashi, AAV 501) with an applied load of 245.2 mN.

3. Results and Discussion

3.1 Dispersion of Fe-based metallic glass particles and pure Fe particles

Figure 2 shows the SEM images of the stir zone of the dispersed specimens after one pass processed at 1500 rpm and 100 mm/min. As shown in Fig. 2(a) and (b), the Fe-based metallic glass particles and pure Fe particles can be uniformly dispersed into the pure Al by FSP and no large defects or cracks are observed in the stir zones. When the Fe-based metallic glass particles were dispersed into the pure Al, the size and shape of the particles are maintained, as shown in Fig. 2(a). This indicates that the metallic glass particles were not deformed or pulverized by the stirring of the tool due to their high strength. However, when pure Fe particles were dispersed into the pure Al, the initial schistose state of the particles was destroyed and the particle size was less than its original value of 150 μm. This is because the pure Fe particles have been crushed by the rotation of the tools. In addition, a thin layer was observed at the interface between the pure Al and Fe, as shown in Fig. 2(b). This indicates that a reaction between Al (base material) and Fe particles occurred during the FSP.

Figure 3 shows the XRD results of the stir zone in the dispersed specimens. When the Fe-based metallic glass particles were dispersed into the pure Al, the Al peaks were observed, as shown in Fig. 3(a). However, when pure Fe particles were dispersed into the pure Al, the diffraction peaks of pure Al and \( \text{Al}_{13}\text{Fe}_4 \) intermetallic compound that formed due to reaction between Al and Fe, were observed as shown in Fig. 3(b).
3.2 Microstructure and mechanical properties

Figure 4 shows the results of EBSD mapping obtained in the stir zone of the dispersed specimens. As shown in Fig. 4(a) and (b), the average grain sizes of the stir zone without particles and with particles are about 33 µm and 17 µm, respectively. The coarsening of the Al grains was retarded due to the pinning effect by addition of the Fe-based metallic glass particles.

Figure 5 shows the hardness distribution along the cross-sectional centerline in the stir zone of the dispersed specimens. As shown in Fig. 5, when the Fe-based metallic glass particles are dispersed into the pure Al, the hardness of the stir zone slightly increased compared with that of the specimen without particles.

3.3 Effect of pass number and travel speed on the hardness

In order to investigate the effect of the heat input on the hardness and particle distribution of the Al FSP joint containing the Fe-based metallic glass, the FSP with various travel speeds were carried out. In addition, a second pass FSP process was repeated to enhance the particles distribution and hardness of the joints. Figure 6 shows the effect of the pass number and travel speed on the hardness of the Fe-based metallic glass particle dispersed specimens. During the FSP, the rotation speed was fixed at 1500 rpm. Generally, a low heat input is necessary during the FSP to prevent crystallization of the metallic glass and the hardness reduction in the stir zone. However, the joint with the travel...
speed of 10 mm/min, corresponding to the highest heat input, shows higher hardness value. In addition, the hardness in the stir zone shows a value higher than that of the base metal. The abnormal hardness increase of the sample with a high heat input requires further investigation. It is worth noting that the pass number also shows a different influence on the hardness distribution of the sample when processed at different travel speeds. When traveled at 100 mm/min, the hardness decreased after the 2 pass FSW process. In addition, for the sample welded at 10 mm/min, the hardness increased after the 2 pass FSW process. Therefore, a different hardening mechanism needs to be considered. For the sample processed at other travel speeds, it shows no significant changes in the hardness distribution of the stir zone.

Figure 7 shows the hardness distribution along the cross section perpendicular to the FSP direction of the pure Fe particles dispersed specimens. When the travel speed decreased to 400, 100 and 25 mm/min, the hardness of the stir zone increased with the increasing heat input. In addition, the hardness decreased with the increasing pass number. From this result, it indicates that when pure Fe particles are dispersed into pure Al, the hardness of the stir zone increased with the increasing heat input.

In order to understand the mechanism of the hardness distribution variations with different processing conditions, microstructural characterizations in the stir zone were carried out by TEM observation. Figure 8 shows the bright field TEM images and corresponding SAD (selected area diffraction) patterns of the stir zone for the Fe-based metallic glass particle dispersed specimens. When the travel speed was 100 mm/min (1 and 2 pass), the Fe-based metallic glass particles maintained their original amorphous structure. On the other hand, when the travel speed was 10 mm/min (1 and 2 pass), fine precipitates were observed. Accordingly, the hardness of the stir zone increased due to the precipitate, as shown in Fig. 6 (10 mm/min).

Figure 9 shows the bright field TEM images and corresponding selected area diffraction (SAD) patterns of the stir zone of the pure Fe particle dispersed specimens. No matter whether the heat input is high or low, intermetallic compounds formed in the dispersed specimen. It is postulated that the pure Fe particles react faster with the Al than the Fe-based metallic glass particles. Also, the amount of the intermetallic compounds increased with the increasing pass number. This indicates that the dispersibility and heat input increased with the increasing pass number, because the reaction between Al and the pure Fe particles are promoted. In addition, when the travel speed was 25 mm/min, the intermetallic compound of AlFe can be observed in the processed specimen after both 1 pass and 2 pass. Since the intermetallic compounds are of very small grain size of 200 nm, it is suggested that they are not from breaking of the pure Fe particles during the FSW, but precipitated from the Al matrix. For the pure Fe dispersed specimen, the hardness of the stir zone increased regardless of the processing condition due to precipitation hardening.
Fig. 8 TEM images of stir zone of Fe-based metallic glass particles dispersed specimens. (a) 1 pass and (b) 2 pass processed at 1500 rpm and 100 mm/min; (c) 1 pass and (d) 2 pass processed at 1500 rpm and 10 mm/min.

Fig. 9 TEM images of stir zone of pure Fe particles dispersed specimens. (a) 1 pass and (b) 2 pass processed at 1500 rpm and 400 mm/min; (c) 1 pass and (d) 2 pass processed at 1500 rpm and 25 mm/min.
3.4 Solid solubility of Fe metallic glass and pure Fe particles

Figure 10 shows the bright field TEM images and corresponding SAD patterns of the Fe-based metallic glass particle dispersed specimens processed at 10 mm/min (1st and 2nd pass). After the one-pass FSP, a reaction layer at the interface between Al and Fe-based metallic glass particle was observed. After the 2nd pass, the precipitate of Al$_{13}$Fe$_4$ was observed due to the reaction between the pure Al and Fe-based metallic glass, as shown in Fig. 10(b). The reaction of the Fe-based metallic glass particles with pure Al is weaker than that of pure Fe particles. When the travel speed is 10 mm/min, the reaction between the Al and Fe-based metallic glass particles was promoted due to the higher heat input. In addition, the amount of the precipitate increased with the increasing pass number. Because the intermetallic compounds have a very small grain size, it is postulated that the intermetallic compounds are not formed from breaking of the pure Fe particles during the FSP, but precipitated from the Al matrix. The hardness of the stir zone after the 2nd pass increased, because the movement of dislocations in the Al matrix was inhibited by the precipitate, as shown in Fig. 10(c).

Figure 11 show the temperature profile of the pure Fe particle dispersed specimens during processing. According to the profile, the peak temperature was 457°C, indicating that the FSP was performed below the melting point of the pure Al. The solid solubility of Fe in Al at the processing temperature of 457°C is low. The precipitates formed in the stir zone, since precipitation as well as dynamic recrystallization are specific phenomena of the FSP. According to the studies of Nagahama et al. on the relationship of recrystallization and precipitation of the Al-Fe alloy, when the precipitates are formed during the recrystallization process, large precipitates are formed in the subgrain boundary and small precipitates are formed in the internal subgrain boundary. On the other hand, precipitates formed after recrystallization are very small and the shape of precipitates is needle-like or discoid-
like. As shown in Fig. 9, large precipitates and small needle-like and discoid shaped precipitates are mixed together, which is consistent with Nagahama’s study.

4. Conclusions

The Fe-based metallic glass particles and pure Fe particles were separately dispersed into pure Al by friction stir processing. The obtained results are summarized as follows:

(1) Fe-based metallic glass particles and pure Fe particles can be uniformly dispersed into pure Al by friction stir processing.

(2) When Fe-based metallic glass particles were dispersed into pure Al, the coarsening of the Al grains retarded. However, the dispersion of the Fe-based metallic glass particles had a little influence on the hardness of the stir zone. When the heat input is increased at the travel speed of 10 mm/min, the reaction between the pure Al and Fe-based metallic glass particles occurred, and the mechanical properties of the stir zone are improved due to the formation of precipitates.

(3) For the pure Fe particle dispersed samples, the hardness of the stir zone increased with the decrease in the travel speed at 400, 100 and 25 mm/min due to the enhanced reaction between the Al and Fe. In addition, the hardness is improved with the increasing pass number. In the Al-Fe system, although the solid solubility of Fe in Al is low, the mechanical properties of the stir zone are improved due to the formation of Al<sub>13</sub>Fe<sub>4</sub> precipitates.

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REFERENCES