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Formation of Cu_2FeAl_7 phase in friction-stir-welded SiCp/Al–Cu–Mg composite

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Extruded 15 vol.% SiCp/Al–Cu–Mg composite was friction-stir-welded (FSW) and subjected to subsequent post-weld T4-treatment. The formation of Cu_2FeAl_7 phase with Fe resulting from steel tool wear was identified at or near the SiC particle interfaces by using transmission electron microscopy, high-resolution electron microscopy and differential scanning calorimetry. Two types of Cu_2FeAl_7 phases were observed: a single crystal phase and a polycrystalline nanostructured phase. No elemental iron was detected. The formation mechanism of the Cu_2FeAl_7 phase is discussed.

Keywords: Friction-stir-welding; Composite; Aluminum; Silicon carbide

Metal matrix composites (MMCs) exhibit improved stiffness, strength, wear resistance and a reduced coefficient of thermal expansion over monolithic matrix alloys, and have potential structural applications in the aerospace and automotive industries [1]. However, the weldability of MMCs is significantly reduced due to the addition of ceramic reinforcements [2]. This limits the widespread application of these composites [3].

Friction-stir-welding (FSW) is a relatively new solid-state joining technique developed for aluminum alloys [4,5]. This joining technique is energy efficient, environmentally friendly and versatile, characterized by not using filler metals or shielding gas, reducing undesirable chemical reactions, and offering low distortion, good dimensional stability and repeatability [4]. Recently, several studies have been conducted to evaluate the feasibility of FSW of aluminum matrix composites [6–10]. It was discovered that high-quality welds without visible defects could be generated by FSW. The homogeneous distribution of reinforcements and improved joint efficiencies were reported [11–13].

In assessing the challenges associated with FSW of MMCs, two issues are worth noting. First, the MMCs exhibit lower plasticity than monolithic alloys even at high temperatures [14]. Therefore, the optimum FSW parameters for producing sound welds were limited to lower tool traverse speeds [6]. For example, Nakata et al. reported that the optimum parameter range for achieving a sound weld of Al_2O_3/6061Al composite was obviously narrower than that of the parent material [12]. Second, when the tool steel was used as the tool material, severe tool wear occurred during FSW, in particular at higher tool rotation rates, due to the presence of hard ceramic reinforcements [2,6,15]. The tool wear not only decreases the lifetime of the FSW tool but also produces some deleterious effects on the properties of the FSW welds due to the existence of the wear debris [16]. Clearly, compared to monolithic alloys, it is difficult to achieve sound welds in MMCs via FSW.

Previous work on the tool wear of the MMCs focused on the effective wear vs. FSW parameters [17,18]. Special attention was given to a self-optimized wear concept in an attempt to assess tool wear. However, very limited literature is available on the formation of wear debris during FSW and subsequent T4-treatment. It was reported that the wear debris contains elements originating from the welding tool [19], e.g. deposits of Fe from the tool during FSW [2]. This leads to some interesting questions. Will the wear debris exist as elemental Fe? Can it react with other elements in the aluminum alloys to form some intermetallic compound?

In a previous paper [20], the present authors reported that 8 mm thick as-extruded SiCp/2009Al composite plates were successfully welded via FSW at a medium

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tool rotation rate of 600 rpm without severe tool wear. In this study, the FSW SiCp/2009Al composite was subjected to detailed microstructural examinations. The purpose of this work is to identify if the Fe debris resulting from tool wear reacts with the elements in the aluminum matrix and the formation mechanism of the reaction product.

15 vol.% SiCp/2009Al composite, produced by powder metallurgy and subsequent extrusion, was used in this study. The chemical composition of the 2009Al alloy was 4.26Cu–1.61Mg–0.01Si–0.009Fe (wt.%). Eight millimeter thick composite plates were cut from the extruded bars and FSW along the extrusion direction at a tool rotation rate of 600 rpm and a traverse speed of 50 mm min^{-1}. A tool with a shoulder of 24 mm in diameter and a cylindrical threaded pin of 8 mm in diameter was used. The tool was made of H13 tool steel with a hardness of HRC 45. Following the FSW, samples were kept at room temperature for one month to age naturally. In order to study the effect of the post-weld heat-treatment (PWHT), some of the FSW samples were subjected to T4-treatment (solutionized at 502 °C for 1 h, water quenched, and then aged at room temperature for one month).

The microstructure was examined using transmission electron microscopy (TEM) and high-resolution electron microscopy (HREM, JEOL 2010) equipped with energy-dispersive spectroscopy (EDS, Oxford). In addition, X-ray analysis was conducted on a D/max 2500PC diffractometer at 50 kV and 250 mA, using Cu K_α radiation ($\lambda = 1.5418$ Å). Differential scanning calorimetry (DSC) experiments were performed at a heating rate of 10 °C min^{-1} using STA449C DSC equipment.

Figure 1 shows the X-ray diffraction patterns of the as-extruded composite and the nugget zone of the FSW composites under the as-FSW and T4-treatment conditions, respectively. The Cu_2FeAl_7 diffraction lines were distinctly visible under both as-FSW and T4-treatment conditions (Fig. 1a and b), indicating the formation of Cu_2FeAl_7 phase in the nugget zone of the FSW composite. By comparison, it seems that no distinct Cu_2FeAl_7 peak was detected in the as-extruded composite (Fig. 1c). No Fe diffraction peak was detected in the FSW composites under both as-FSW and T4-treatment conditions.

TEM examinations revealed the existence of some black particles around the SiC particles in the nugget zones of the FSW composites under both as-FSW and T4-treatment conditions (Fig. 2a and b). EDS analyses indicated that these black particles contained 42.6 wt.% Cu, 13.5 wt.% Fe and 43.9 wt.% Al, and the compositions are very close to those of the Cu_2FeAl_7 phase. Cu_2FeAl_7 is tetragonal, space group *P*4/mnc with the lattice parameters $a = 6.336$ Å and $c = 14.870$ Å [21]. By comparison, the interface between SiC and aluminum matrix was relatively clean in the as-extruded composite and large Cu_2FeAl_7 particles were hardly observed around the SiC particles (Fig. 2c).

HREM observations revealed that two types of Cu_2FeAl_7 phase were generated in the FSW composite. The first was single-crystal Cu_2FeAl_7 phase, as shown in Figure 3. This type of Cu_2FeAl_7 particle was found in

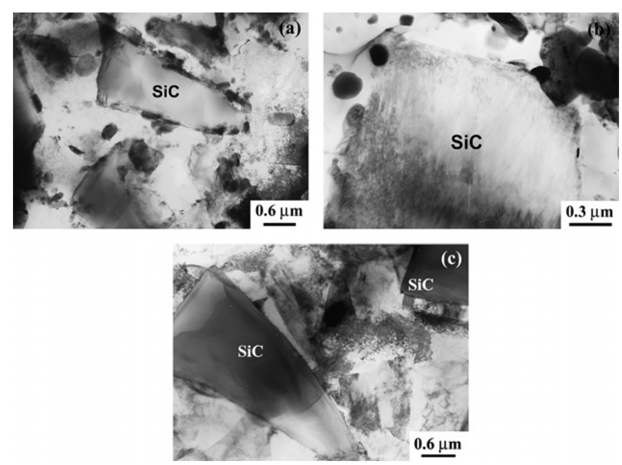

Figure 2. TEM images showing the Cu_2FeAl_7 phase around SiC particles in the nugget zones of FSW composites under (a) as-FSW and (b) T4-treatment conditions, and (c) the relatively clean interface in as-extruded composite.

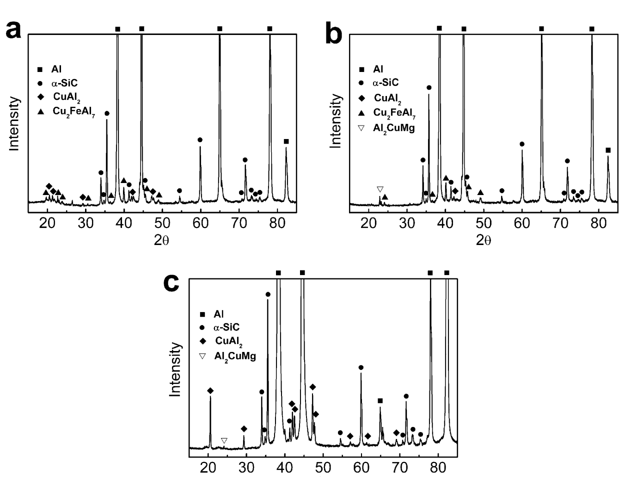

Figure 1. X-ray diffraction patterns of the nugget zones in FSW composites under (a) as-FSW and (b) T4-treatment conditions, and (c) the as-extruded composite.

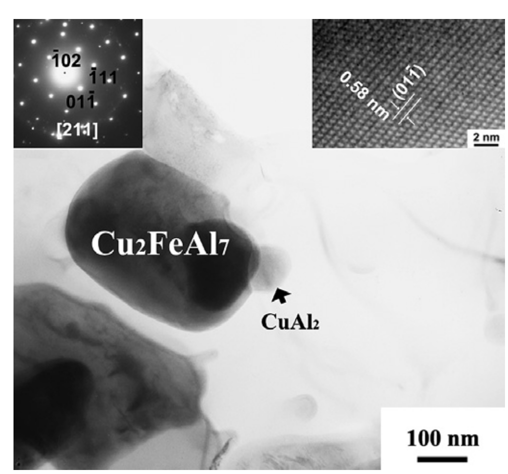

Figure 3. TEM image of nugget zone of T4-treated FSW composite with inserted micrographs showing a selected-area diffraction pattern and an HREM image of the Cu_2FeAl_7 phase.

the aluminum matrix near the SiC particles. A small particle adjacent to the Cu_2FeAl_7 particle was identified by EDS to be $CuAl_2$ phase. This implies that the formation of the Cu_2FeAl_7 phase was intimately associated with the existence of the $CuAl_2$ phase. The second was polycrystalline nanostructured Cu_2FeAl_7 phase, as shown in Figure 4. This type of Cu_2FeAl_7 particle was detected at the interface of the SiC particle with a specifically crystallographic orientation relationship $(10\bar{1}2)_{SiC}\|(212)_{Cu_2FeAl_7}$ between the SiC particle and Cu_2FeAl_7 phase. No interfacial transition layer was found between them.

The heat flow of the as-extruded and as-FSW composites, measured as a function of the temperature, is shown in Figure 5. The endothermic peak (632 °C) observed in the composite was due to the melting of aluminum matrix. However, an additional endothermic peak with an incipient melting temperature of 545 °C was detected in the nugget zone of the as-FSW SiCp/2009Al composite. This endothermic peak is associated with the Cu_2FeAl_7 phase.

According to an Al–Cu–Fe liquidus projection diagram, the projection of the liquidus valley onto a Gibb's triangle plane is shown in Figure 6. There is a ternary eutectic transformation at 545 °C (point E) [22]. The eutectic composition of liquid consists of 82.8% Al, 17.0% Cu and 0.2% Fe by atom ratio.

$$\text{L.q.} \rightarrow \text{Al} + \text{CuAl}_2 + \text{Cu}_2\text{FeAl}_7\ (545\ ^\circ\text{C}) \tag{1}$$

According to Figure 6, there are two prerequisites for the formation of the Cu_2FeAl_7 phase in the aluminum alloys. The first is a higher concentration of Cu and Fe, and the second is higher temperature. Because the 2009Al matrix has a very low Fe concentration of 0.009 wt.%, it seems impossible to form the Cu_2FeAl_7 phase in the alloy. However, the tool wear during FSW due to the presence of hard and sharp SiC particles might result in a significantly higher Fe concentration in the FSW composite. In several previous studies, it was reported that the tool wear occurred in FSW of ceramic-particle-reinforced aluminum matrix composites and generally increased with increasing tool rotation rate [2,15]. It is expected that there was higher Fe concentration around the SiC particles because the tool wear was caused by hard and sharp SiC particles. Therefore, the local high Fe concentration might form around the SiC particles due to the wear of SiC particles to the steel tool in the FSW SiCp/2009Al composite. On the other hand, the aggregation of $CuAl_2$ phase at the reinforcement/matrix interfaces has been reported in several discontinuously reinforced Al–Cu–Mg composites [23,24]. In this study, TEM examinations on the SiCp/2009Al composite also revealed the aggregation of $CuAl_2$ particles at SiC/Al interfaces (not shown). Therefore, it is very likely that local high concentrations of Fe and $CuAl_2$ will form around the SiC particles in the FSW SiCp/2009Al composite.

During FSW of aluminum alloys, peak temperatures of ~500 °C were recorded in the regions adjacent to the rotating pin [25]. The presence of SiC particles in the composite resulted in enhanced friction and reduced thermal dissipation during FSW. It is very likely that a peak temperature $\geqslant$500 °C was reached in the nugget zone of FSW composite.

FSW produces intense plastic deformation and material mixing with a strain rate of 10^0–10^2 s^{-1} and a strain of up to ~40 [26,27], resulting in significantly acceler-

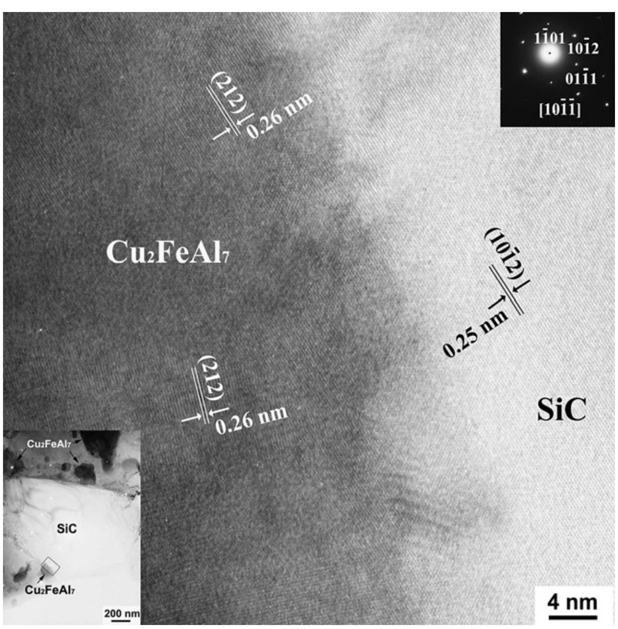

Figure 4. HREM image showing interface between SiC particle and Cu_2FeAl_7 phase in the nugget zone of T4-treated FSW composite.

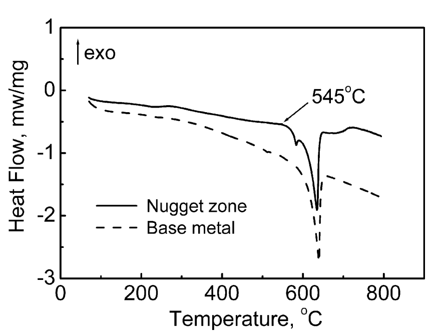

Figure 5. DSC curves for as-extruded and as-FSW SiCp/2009Al composites.

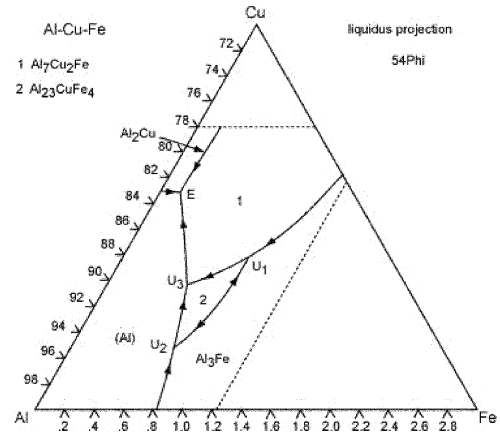

Figure 6. Liquidus projection of Al–Cu–Fe ternary system [22].

ated diffusion rate and shortened diffusion distance [28]. Biallas et al. suggested that the material flow around the pin is somewhat similar to that of regular milling of the metal [29]. Generally, mechanical alloying consists of repeated deformation (welding, fracturing and rewelding) of powder particles and can be used as a means of synthesizing nanostructured material [30]. Therefore, it is very likely that the ternary phase Cu_2FeAl_7 formed through the solid-state reaction between Fe, $CuAl_2$ and Al, under milling-like severe plastic deformation condition with higher concentrations of Fe and $CuAl_2$ and higher process temperatures. This suggestion is supported by the formation of the nanostructured Cu_2FeAl_7 phase.

It is generally believed that the solubility of Fe in aluminum alloy is small and it is usually present in the form of $FeAl_3$. However, when the aluminum alloy has a higher content of Cu, Fe will form the Cu_2FeAl_7 phase, which is insoluble via solution treatment [31]. This is because the dissolution temperature of Cu_2FeAl_7 phase (545 °C) is much higher than the solution treatment temperature (~500 °C for Al–Cu–Mg alloys). Therefore, the post-weld T4-treatment did not change the Cu_2FeAl_7 phase in the FSW SiCp/2009Al composite.

The formation of the Cu_2FeAl_7 phase in the FSW SiCp/Al composite will influence the mechanical properties in two ways. First, the Cu_2FeAl_7 particle formed at the interface of the SiC particle might reduce the interfacial bonding between SiC and aluminum matrix. Second, the formation of Cu_2FeAl_7 phase reduced the amount of the precipitates due to the dilution of Cu. In this case, the precipitation-strengthening effect from the fine precipitates was reduced. Our research on the effect of the formation of the Cu_2FeAl_7 phase on the mechanical properties of the FSW SiCp/2009Al composite will be reported elsewhere.

In summary, the formation of the Cu_2FeAl_7 phase was identified at or near SiC particle interfaces in the FSW SiCp/2009Al composites under both as-FSW and T4 conditions. Two types of Cu_2FeAl_7 phase were generated in the FSW composite: a single-crystal Cu_2FeAl_7 phase and a polycrystalline nanostructured Cu_2FeAl_7 phase.

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