Microstructure and Properties of In Situ Si and Fe-rich Particles Reinforced Al Matrix Composites Assisted with Ultrasonic Vibration

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Abstract In this research, the in situ Si and Fe-rich particles reinforced Al matrix composites were fabricated by rheocasting (RC) process assisted with ultrasonic vibration (USV). After USV treatment, the polygonal primary Si crystals were refined into particles with average diameter of about 15–23 μm , and the fraction of primary Si declined to about 5.4–6.5 vol%. The coarse plate-like $\delta\text{-Al}_4(\text{Fe},\text{Mn})\text{Si}_2$ phase was transformed into fine particles with average diameter of about 17–20 μm , and the fraction of particle-like Fe-bearing particles is about 3.6–5.3 vol%. The ultimate tensile strength of the RC composites increases with the increase of Fe content at 350 °C. The increase of the elevated temperature strength of the composites is mainly attributed to the refinement of $\delta\text{-Al}_4(\text{Fe},\text{Mn})\text{Si}_2$ phase and the increase of the volume fraction of the Fe-bearing compounds. Compared with the composites without USV, the RC composites assisted with USV have thinner mechanical mixing layer in wear test, which corresponds to smaller wear rate.

KEY WORDS: Fe-bearing phase; Metal matrix composites; Microstructure; Properties; Ultrasound

1 Introduction

The particulate-reinforced Al matrix composites are widely used in aerospace, automotive manufactory, and other fields due to their light density, high specific strength and stiffness, good wear resistance, and favorable coefficient of thermal expansion [1]. The parts manufactured with these composites often serve at high-temperature circumstance. For instance, the operating temperature of Al matrix composites piston is approximately up to 350 °C [2]. Therefore, the improvement of the elevated temperature strength of the Al matrix composites has been attracting a lot of attention.

Iron is the most harmful impurity element in cast Al alloys. Because the equilibrium solid solubility of Fe in aluminum is very low, Fe segregates during solidification and tends to form intermetallic compounds. The coarse Fe-bearing compounds, which have relatively low bonding strength with the matrix, are detrimental to the mechanical properties. Nevertheless, it has been proven that Fe is an effective element for improving the high-temperature strength of Al alloys [3]. For this reason, refining of Fe-bearing compound particles in Al alloys is very important for the improvement of their mechanical properties.

Many techniques have been proposed to change the morphology of the coarse Fe-bearing compounds, such as rapid solidification, melt superheating, and addition of neutralizer elements. However, the application of these methods is limited because of their costly equipment or complicated processes. Recent studies have shown that ultrasonic vibration (USV) treatment can effectively modify the Fe-bearing compounds of hypereutectic Al–Si alloys [4, 5]. Hence, the high Fe content hypereutectic Al–Si alloy has the prospect of becoming an in situ composite

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(the silicon and the Fe-rich phases acting as the reinforcing phase) with specific properties.

In this research, the in situ Si and Fe-rich particles reinforced Al matrix composites were fabricated by rheocasting (RC) process assisted with USV. The microstructure, mechanical properties, and wear behavior of the composites were investigated. The reinforcing mechanism of in situ primary Si particles and Fe-bearing compounds was also discussed.

2 Experimental

The in situ Si and Fe-bearing particles reinforced Al matrix composites with different Fe additions were prepared under USV, and their chemical compositions are listed in Table 1. Differential scanning calorimetry (DSC) (NET-ZSCH STA449F3) was employed to determine the liquidus temperatures of the composites. The liquidus temperatures of C1, C2, and C3 composites are about 650, 655, and 674 °C, respectively. The installation of USV in Ref. [4] was employed in this experiment. The applied ultrasonic power was 1.6 kW, and the frequency of USV was 20 kHz.

The raw materials were melted in a resistance furnace at 820–850 °C. Then, the melt was degassed for 10 min with argon gas, and cooled down to a temperature of 750–780 °C. Subsequently, about 600 g melt was poured into a preheated metal cup. Then, the melt of C1 and C2 composites was treated by USV in the temperature range of 665–645 °C, and C3 composite was treated at 682–662 °C. After the melt was treated with USV for 1.5 min, the semi-solid slurry with a certain solid fraction was obtained. The slurry was immediately poured into an iron mold preheated at 200 °C to get tensile samples with diameter of 8 mm. For comparison, conventional gravity casting (GC) specimens were also made without USV treatment under a pouring temperature of 750 °C. Half of the samples were heat treated with T6 process (solution treated at 510 °C for 7 h, followed by water quenching, and then artificially aged at 190 °C for 10 h).

Room temperature and elevated temperature tensile tests were performed on a SHIMADZU AG-IC tester. The average Brinell hardness of the specimens was tested by a BH3000/T electronic Brinell hardness tester. Specimens

for the metallographic examination were cut from the ends of tensile test specimens, then grinded, polished, and etched by solution with 5% sodium hydroxide. The microstructures were examined using an Axiovert 200MAT optical microscope. Six micrographs for each specimen were analyzed by a self-developed software system Solidvf 3.0 which can calculate the size and volume fraction of the reinforcing particles. The size of reinforcing particles was characterized by average particle diameter, which was measured by Heyn's linear intercept method. Phase constitution and composition of different phases were studied by Quanta 200 environmental scanning electron microscope (SEM) fitted with an Energy Dispersive X-ray Spectroscopy (EDX). X-ray diffraction (XRD) analysis was carried out with an X'Pert PRO diffractometer.

Dry sliding wear tests were carried out on a pin-on-disk wear tester to evaluate room temperature (i.e., 25 °C) wear behavior of the composites. The pins, 6 mm in diameter and 12 mm in length, were machined from the RC or conventional GC samples. The disks, with a diameter of 70 mm and a thickness of 10 mm, were made of GCr15 steel. The pins in the horizontal orientation were loaded against the rotating steel disk through a dead weight loading system. The tests were conducted at four different loads of 50, 100, 150, and 200 N. Sliding speed and distance were kept constant at 0.75 m/s and 1,000 m, respectively. The wear rate values were determined from the measured weight loss and expressed in terms of volume loss per unit sliding distance (m^3/m).

3 Results and Discussion

3.1 Microstructures of the Composites

Figure 1 shows the microstructures of as-cast C1, C2, and C3 composites produced by conventional GC. Because of low Fe content, Fe-bearing compounds were hardly observed in C1 composite; therefore, the reinforcing particles of this composite mainly consisted of polygonal primary Si (A), as shown in Fig. 1a. Apart from polygonal primary Si (A), the reinforcing particles of C2 composite consisted of coarse plate-like compound (B) and long needle-like compound (C), but a small quantity of dendritic compound (D) was observed occasionally, as shown in Fig. 1b. EDX analysis indicated that Al, Si, Fe, and Mn elements were observed in compound B, C, and D, as shown in Table 2. XRD analysis showed that C2 composite contains $\delta\text{-Al}_4(\text{Fe,Mn})\text{Si}_2$, $\beta\text{-Al}_5(\text{Fe,Mn})\text{Si}$, AlNi_3 , and $\text{Al}_8\text{FeMg}_3\text{Si}_6$ phase, as shown in Fig. 2a. It can be concluded that compound B and C should be $\delta\text{-Al}_4(\text{Fe,Mn})\text{Si}_2$ and $\beta\text{-Al}_5(\text{Fe,Mn})\text{Si}$ phase, respectively. The compound D

Table 1 Chemical compositions of the in situ particles reinforced Al composites (wt%)

Composites	Si	Fe	Cu	Ni	Mn	Mg	Al
C1	17	0.23	2	1	0.4	0.4	Balance
C2	17	2	2	1	0.8	0.4	Balance
C3	17	3	2	1	0.8	0.4	Balance

Fig. 1 Microstructures of the as-cast composites produced by conventional GC: **a** C1; **b** C2; **c** C3

Table 2 EDX results of intermetallic compounds in C2 composite produced by conventional GC (at%)

Compounds	Al	Si	Fe	Mn	Ni	Cu	Mg
B	50.96	30.77	14.48	2.63	0.82	0.33	–
C	75.69	13.78	7.43	1.44	1.05	0.62	–
D	68.01	11.36	11.89	6.29	0.69	1.1	0.66

was close to α -Al₁₅(Fe,Mn)₃Si₂ based on the ratio of compositions of the compounds, as shown in Table 2. Some researches confirmed that the dendritic α phase usually appears in the high Fe content Al–Si alloys [6]. Therefore, the compound D should be α phase. As for the absence of the diffraction peaks of α phase shown in Fig. 2a, it may be due to a barely detectable volume fraction of the phase in C2 composite. In the microstructure of C3 composite, primary Si, δ and β compounds appeared as the reinforcements, as shown in Figs. 1c and 2b.

Figure 3 shows the microstructures of the as-cast C1, C2, and C3 composites produced by RC process assisted with USV. The compounds in C2 and C3 composites were also determined by EDX and XRD analysis, and XRD patterns are

shown in Fig. 4. The polygonal primary Si crystals in C1, C2, and C3 composites were refined into particles with average diameters of about 27, 22, and 23 μ m, respectively. The coarse plate-like δ phase was also refined into fine block-like particles, and only a small amount of needle-like β phase remained. Comparing Figs. 2a and 4a, and Figs. 2b and 4b, the phase constitutions of C2 and C3 were invariant after USV, meaning that USV could change the morphologies of the Fe-bearing compounds but not the phase constitutions of the composites. The size and volume fraction of the reinforcements in C1, C2, and C3 composites produced by RC process assisted with USV were measured and are shown in Table 3.

Compared with the GC composites without USV (Fig. 1), the primary Si and δ phases in the composites produced by RC process with USV (Fig. 3) were refined significantly. This could be mainly attributed to the cavitation and acoustic streaming induced by USV. The energy of the collapsing cavitation bubbles is transformed into pressure pulses up to 1,000 MPa [7]. According to the Clausius–Clapeyron equation, the pressure pulse arising from the collapse of bubbles alters the equilibrium melting point (T_m). An increase in T_m is equivalent to the increase of the undercooling, so that an enhanced nucleation event

Fig. 2 XRD patterns of the as-cast C2 **a**; C3 **b** composites produced by conventional GC

Fig. 3 Microstructures of the as-cast composites produced by RC: **a** C1; **b** C2; **c** C3

Fig. 4 XRD patterns of the as-cast C2 **a**, C3 **b** composites produced by RC

Table 3 Average diameters and volume fractions of reinforcements in C1, C2, and C3 composites with RC

Composites	Average diameter of particle-like δ phase (μm)	Volume fraction of Fe-bearing phase (vol%)	Average diameter of primary Si (μm)	Volume fraction of primary Si (vol%)	Total volume fraction of particles (vol%)
C1	—	—	27	9.3	9.3
C2	18	3.6	22	6.5	10.1
C3	20	5.3	23	5.4	10.7

is expected. The acoustic streaming plays a very important role in homogenizing the solute and temperature fields of the melt, accelerating the heat and mass transfer. This is expected to slow down the preferential growth of primary Si and δ phases during the growth stage. As a result, the average grain diameter of them decreases significantly after USV.

3.2 Mechanical Properties of the Composites

The mechanical properties of C1, C2, and C3 composites produced by different processes are listed in Table 4. As it can be seen, in T6 condition, the ultimate tensile strengths

(UTS) of the RC samples of C2 and C3 composites were 289 and 262 MPa at room temperature, respectively. Compared with the GC samples, the UTS of RC samples of C2 and C3 composites were increased by 22.5 and 33.7% at room temperature, respectively.

Figure 5 shows the fractographs of the tensile specimens of C2 and C3 composites produced by different processes after T6 heat treatment. Without USV treatment, the crack mainly propagated along the boundaries between the δ phase platelets and the aluminum matrix, as shown in Fig. 5a and b. In contrast, in the case of the samples treated with USV, the crack mainly propagated through the primary Si, as shown in Fig. 5c and d. At room temperature,

Table 4 Mechanical properties of T6 treated C1, C2, and C3 composites produced by different processes

Process	Composites	At room temperature			At 350 °C	
		UTS (MPa)	Elongation (%)	Hardness (HB)	UTS (MPa)	Elongation (%)
GC	C1	278	0.72	112	110	1.41
	C2	236	0.53	127	123	0.90
	C3	196	0.28	141	120	0.45
RC	C1	306	0.93	119	116	1.55
	C2	289	0.70	139	132	1.18
	C3	262	0.35	150	135	0.55

UTS of the as-cast C1, C2, and C3 composites produced by GC process are 194, 175, and 167 MPa, respectively; UTS of the as-cast C1, C2, and C3 composites produced by RC process are 207, 190, and 177 MPa, respectively

Fig. 5 SEM fractographs of the tensile samples produced by different processes: **a** C2 by GC; **b** C2 by RC; **c** C3 by GC; **d** C3 by RC

the sharp tips of polygonal primary Si crystals and coarse plate-like Fe-bearing compounds acted as potential sites for crack initiation that may result in de-cohesion failures. With USV treatment, the primary Si and Fe-bearing compounds were refined and existed mainly as fine particles, which had less effect on crack initiation and could prevent dislocations from moving. Thus, the UTS of the RC samples at room temperature were improved significantly.

Nevertheless, the UTS of the RC samples at room temperature decreased with the increase of Fe content. As shown

in Table 3, the total volume fractions of the reinforcing particles (Si particles and Fe-bearing compounds) of the RC samples of C1, C2, and C3 are 9.3, 10.1, and 10.7 vol%, respectively. Although the primary Si and Fe-bearing compounds were refined, their inherent brittleness at room temperature was not changed. As a result, the room temperature UTS of the RC composites decreased with the increase of total volume fraction of the reinforcement particles.

The UTS at 350 °C of C2 and C3 composites with RC process were also improved, which are 132 and 135 MPa,

Fig. 6 Wear rates of the composites produced by different processes as a function of applied load: **a** C1; **b** C2; **c** C3

Fig. 7 SEM images showing the subsurface of the C1 **a, d**, C2 **b, e**, and C3 **c, f** composites with USV **a–c** treatment and without USV treatment **d–f** after wear tests under a load of 200 N (arrow marks indicate the sliding direction)

respectively. They are about 14% higher than that of C1 composite at 350 °C (Table 4). The UTS of the RC samples at 350 °C increased with the increase of Fe content. As it can be seen from Table 3, the volume fraction of particle-like Fe-bearing compounds increased from 3.6 (C2) to 5.3% (C3). In contrast, the volume fraction of primary Si decreased from 6.5% (C2) to 5.4% (C3). Hence, the fine Fe-bearing particles play an important role in improving the elevated temperature strength of the composites.

Although both copper and magnesium act as precipitation-hardening elements, their strengthening effect is diminished at elevated temperature because of the low thermal stability of the precipitates. However, at elevated

temperature, the Fe-bearing particles exhibit excellent thermal stability [8]. Moreover, the fine Fe-bearing particles, which were distributed uniformly in the matrix, have higher bonding strength with the matrix and can also prevent grain-boundary sliding. Consequently, the elevated temperature strength was improved after USV and increased with the increase of the volume fraction of the fine Fe-bearing compounds.

3.3 Wear Behavior of the Composites

Figure 6 represents the variation of wear rates as a function of applied loads for the composites with or without USV

treatment. The wear rate of conventional GC composites without USV was higher than that of RC composites with USV under all loads. It means that RC process and USV can enhance the wear properties of the composites. The wear behavior is closely related to the microstructure features of the materials [9, 10]. The superior wear resistance of the RC composites was attributed to the refinement of the primary Si and Fe-bearing reinforcement particles.

Figure 7 shows the subsurface microstructure of the composites tested under a load of 200 N. The mechanical mixing layer (MML) was the outermost layer in all these micrographs. The MML commonly formed on the sliding surface of the material when it slides against a steel counterpart [11, 12]. Furthermore, a deformation zone beneath the MML with material flow lines that progressively bend toward the sliding direction was observed. Finally, beneath the deformation zone, there was undeformed material.

It is obvious that the MML thicknesses of the composites were reduced by RC and USV. This can be attributed to the size and morphology of the primary Si and Fe-bearing phases. The long needle-like β and coarse plate-like δ phases, which are hard and brittle, have relatively low bonding strength with the matrix [13]. Hence, cracks initiated easily in these brittle phases during sliding. In addition, the Si particles in the composites existed as discrete grains. Larger Si grains lead greater microcracking tendency [10]. In Fig. 7a and c, some coarse primary Si grains in the deformation zone were broken toward the sliding direction. The long needle-like β and coarse plate-like δ phases were fractured and aligned along the sliding direction, as shown in Fig. 7b and c. The fragmented Fe-bearing phases weakened the MML and facilitated the initiation and propagation of the microcracks due to their poor bonding in the MML. However, the fine Si particles and Fe-bearing phases could act as effective barriers for dislocation motion in the subsurface region during sliding, as illustrated in Fig. 7d, e, and f. For this reason, USV-treated composites had smaller thicknesses of MML than those of the composites without USV. It can be concluded that the small wear rate corresponds to the thin MML. Therefore, the wear properties of the RC composites were better than that of the GC composites.

In addition, from Fig. 6, it was also obvious that with the increase of Fe content, the wear rate was decreased. This can be confirmed from Table 3 that the total volume fraction of hard particles increased with the increase of Fe content. From Table 4, the hardness of the matrix was also increased when Fe content was increased from 0.23 to 2 and 3%. These two factors caused the decrease of wear rate.

4 Conclusions

- (1) After USV treatment, the polygonal primary Si in Al matrix composites was refined into particles with average diameters of about 15–23 μm . The Fe-bearing reinforcement phases were also refined, and they mainly existed as particle-like $\delta\text{-Al}_4(\text{Fe,Mn})\text{Si}_2$ phase with average grain diameters of 17–20 μm . Cavitation and acoustic streaming caused by USV led to the refinement of primary Si and $\delta\text{-Al}_4(\text{Fe,Mn})\text{Si}_2$ phases.
- (2) Compared with the GC composites without USV, the UTS of RC composites C2 and C3 with USV treatment were increased by 22.5 and 33.7% at room temperature, respectively. The UTS of the RC composites increased with the increase of Fe content at 350 °C, although it decreased with the increase of Fe content at room temperature.
- (3) RC composites with USV had smaller thicknesses of MML than those of the GC composites without USV. Small wear rate corresponded to the thin MML. The decrease of wear rate was attributed to the fine Si particles and Fe-bearing reinforcements in RC composites.

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