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Improvement of particles distribution of in-situ 5 vol% TiB_2 particulates reinforced Al-4.5Cu alloy matrix composites with ultrasonic vibration treatment

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ABSTRACT

Ultrasonic vibration treatment is successfully applied in preparation of in-situ 5 vol% TiB_2 particulates reinforced Al-4.5Cu alloy matrix composites with salt-metal reaction route. Formation of TiB_2 phase is confirmed by X-ray diffraction analysis, and intermediate phases such as Al_3Ti are not detected. Agglomerations of particles in the melt are effectively eliminated by the cavitation and acoustic streaming affects in the melt with ultrasonic vibration. TiB_2 particles are uniformly distributed throughout the bulk melt after treated by ultrasonic vibration for 240 s. The tiny agglomerations formed by TiB_2 particles smaller than 100 nm are also broken by ultrasonic vibration. Some TiB_2 particles smaller than 400 nm are observed to dispersed near grain boundary in the matrix after solidification. The optimal improvements of yield strength and ultimate tensile strength are 114% and 50%, respectively, when composite treated by ultrasonic vibration for 240 s.

1. Introduction

As promising materials for application in structure, aerospace, the military and transportation, particulates reinforced aluminum matrix composites (PRAMCs) attracts a lot attentions since they have outstanding combination of mechanical properties like low density, high specific strength, specific modulus, hardness and low thermal expansion coefficient [1–4]. TiB_2 ceramic phase is an outstanding reinforcements in aluminum among various potential reinforcement particles like Al_2O_3, SiC, TiC, Si_3N_4, B_4C, ZrB_2 [5–11], since TiB_2 particles have high modulus, high hardness, high melting point, good thermodynamic stability, high corrosion resistance and low density [5,12–15]. TiB_2 also has no interface reaction with aluminum. As a widely used in-situ process, the salt-metal reaction route is based on the aluminothermic reaction between two kinds of potassium fluoride salts and aluminum to form in-situ TiB_2 particles. The following sequences are the exothermic processes of salt-metal reaction route [5,16]:

$$3K_2TiF_6 + 13Al \rightarrow 3KAlF_4 + K_3AlF_6 + 3Al_3Ti \quad (1)$$

$$2KBF_4 + 3Al \rightarrow 2KAlF_4 + AlB_2 \quad (2)$$

$$AlB_2 + Al_3Ti \rightarrow 4Al + TiB_2 \quad (3)$$

And a direct reaction is also believed to form in-situ TiB_2 particles [17]:

$$3K_2TiF_6 + 6KBF_4 + 10Al \rightarrow 3TiB_2 + 9KAlF_4 + K_3AlF_6 \quad (4)$$

Compared with external addition processes, TiB_2 particles formed by salt-metal reaction route are much well bonded with aluminum matrix, thus the particle-matrix system has clearer interface and better interfacial thermodynamic stability. More important, the in-situ process has no wetting problem between TiB_2 particles and molten aluminum which is the biggest difficulty in external addition processes [5]. Among various in-situ processes, salt-reaction route has much lower reaction temperature than other processes, which makes TiB_2 particles are smaller and can be submicron in size. Reactions of salt-reaction route are also more moderate, thus the reaction products are much controllable [5,12,20].

But there are also some shortages of salt-reaction route. First of

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all, the mass fraction of Ti and B in K_2TiF_6 and KBF_4 are 19.9% and 8.6%, respectively, so the utilization rate of K_2TiF_6 and KBF_4 is much lower comparing with the Al-Ti-B in-situ system. Therefore fabricating high TiB_2 particulate volume percentage aluminum matrix composites needs a very high weight ratio of salts to base metal, which may enlarge the possibility of inhibiting of subsequent reactions and form more TiB_2 agglomerations. Agglomerations may harm the mechanical properties [18], but more dispersed particles bring more effective reinforcements of Orowan strengthening [19]. In our previous work, the mechanical stirring is employed to reduce TiB_2 agglomerations [20]. The results show that mechanical stirring can effectively eliminate large agglomerations, but small agglomerations with size in 50–100 μm cannot be completely eliminated by mechanical stirring. And introducing mechanical stirring at a speed higher than 540 rpm may lead to the mixture of molten salts and the melt, which may cause a seriously increasing of rounded and large agglomerations instead of reducing or eliminating small agglomerations. Therefore better solution to further improve particles distribution is needed.

Ultrasonic vibration is a promising technology to treat molten aluminum alloys and other light alloys [21–23]. It is a relatively environment-friendly process with low cost and uncomplicated procedure. Ultrasonic vibration treatment (UVT) can clean and degas the melt and often be used to refine intermetallics or other metallic phases [24]. It is also used to improve particulate distribution of composites [11,23]. But researches about using UVT to improve particulate distribution of high volume percentage TiB_2 particles reinforced aluminum matrix composites are scarcer so far.

In this work, Al-Cu alloy is chosen to be the base metal. In practical experiments, TiB_2/Al-4.5Cu composites are successfully fabricated by in-situ salt-metal reaction route, and the particle volume percentage is set at 5 vol% (nominal). UVT is employed to treat the re-melt composites, and agglomerations in all size are effectively reduced by UVT. The improvements of particulate distribution and mechanical properties of the composites are discussed. The mechanism of UVT improving particulate distribution is also discussed.

2. Experiment procedures

2.1. Materials preparing and processing

For preparing TiB_{2p}/Al-4.5Cu composites, a resistance furnace was firstly employed to melt aluminum. A graphite crucible was used to contain aluminum (99.8%, wt%, the same below) ingots. Then Cu (99.9%) chips were added when aluminum melt at 700 °C. Reaction salts were mixture of K_2TiF_6, KBF_4 and Na_3AlF_6 (which was used to help the reactions as flux), and they were all chemically pure and thoroughly mixed. Mass of K_2TiF_6 and KBF_4 were controlled on a Ti/B molar ratio at 1/2, and mass of Na_3AlF_6 was 10% of total mass of K_2TiF_6 and KBF_4. Reaction salts were prepared at the mass could form 5 vol% (nominal) TiB_2 particles. They were preheated and wrapped by aluminum foil, then gradually added into molten aluminum at 830 °C and avoided to generate great fluctuation of reaction temperature. After addition of all the salts, the melt was held at 830 °C for 40 min with stirring to complete the reactions [20]. After that the melt was cast at 720 °C using a preheated permanent mould.

The UVT was employed in this study and sketch of UVT system was shown in Fig. 1 [24]. In this study, interval resting time Tr was set at 1 s and ultrasonic time Tw was also set at 1 s in an ultrasonic viberation cycle. Vibration power was set at 2.8 kw which is the biggest power of UVT system. The frequency of ultrasound was 20 kHz. The transforming rod was made with titanium alloy with a diameter of 25 mm, and metal cup was made with stainless steel

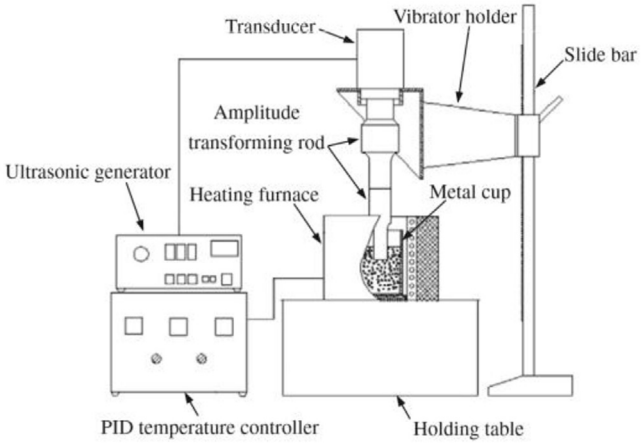

Fig. 1. Sketch of UVT system.

with a height of 130 mm and a diameter of 70 mm.

In ultrasonic vibration experiment, a resistance furnace was firstly employed to melt TiB_{2p}/Al-4.5Cu composite ingots and crucible was a graphite crucible. Then the melt of composite was poured into the metal cup at 760 °C when it was preheated up to 700 °C. After the melt was cooled down to 720 °C, ultrasonic vibrator was immersed into the melt below the surface at 10–15 mm and then start UVT. UVT time was set at 60, 120 and 240 s, respectively. The temperature of the melt was controlled in the range of 720–710 °C during UVT. After UVT, the slag was removed and the quenching samples were obtained by quenching water with a 6 mm diameter quartz tube. Then the molten composite was immediately cast into a preheated permanent mould.

2.2. Characterization

For metallographic examination, specimens of composites and base metal were cut from the ends of tensile test samples. Quenching samples and specimens were grinded and polished before etched by 0.5% HF solution. X-ray diffraction (XRD) examination was carried by a SHIMADZU XRD-7000S X-ray diffractometer with Cu Ka radiation operated at 40 kV and 30 mA. Microstructure analysis was carried by a JEOL JSM-7600F scanning electron microscope. A Tecnai G2 F30 (FEI, Holland) transmission electron microscope (TEM) was employed. A SHIMADZU AG-100KN tester was employed for tensile tests following the GB/T228.1–2010 standard, tests were proceeded at room temperature (25 °C) with crosshead speed at 1 mm/min. Four samples for each specific condition were tested to obtain the average mechanical properties, such as ultimate tensile strength (UTS). Fig. 2 shown the sketch of the tensile test specimen.

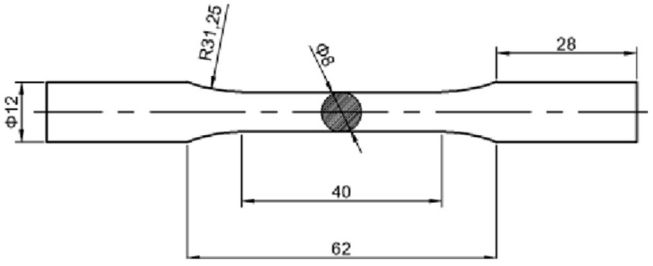

Fig. 2. The drafting of tensile test specimen.