The effect of rotating magnetic field on the microstructure of in situ TiB$_2$/Cu composites

C Zou, H Kang, R Li, M Li, W Wang, Z Chen and T Wang[a]
Key Laboratory of Materials Modification (Ministry of Education), School of Materials Science and Engineering, Dalian University of Technology, Dalian, 116024, China

E-mail: [a] tmwang@dlut.edu.cn

Abstract. Nano ceramic particulate reinforced metal matrix composites are confronted with the problem of particle aggregation emerging in the process of solidification. It sharply deteriorates the mechanical properties of the composites. In order to improve the microstructure and particle distribution, in situ TiB$_2$/Cu composites were prepared using Ti and Cu-B master alloys in a vacuum medium frequency induction furnace equipped with a rotating magnetic field (RMF). The effect of RMF magnetic field intensity employed on the microstructure and particles distribution of the TiB$_2$/Cu composites were investigated. The results show that with the applied RMF, TiB$_2$ particles are homogeneously distributed in the copper matrix, which significantly improves the mechanical properties of TiB$_2$/Cu composites. The mechanism of RMF may be ascribed to the following two aspects. On the one hand, the electromagnetic body force generated by appropriate RMF drives forced convection in the equatorial plane of composite melt during solidification. On the other hand, a secondary flow in the meridional plane is engendered by a radial pressure gradient, thus making a strong agitation in the melt. These two effects result in a homogenous dispersion of TiB$_2$ particles in the copper matrix, and hence excellent properties of TiB$_2$/Cu composites were obtained.

1. Introduction
Due to the good mechanical properties, high electrical conductivity and thermal conductivities in copper and copper alloy, it has been widely used as rail transit line contacts, integrated circuit lead frame, electrodes and heat exchangers [1]. The materials require both high conductivity, well thermal conductivity, and high strength. The major strengthening method of traditional copper alloy is solution strengthening. In those alloy, alloying elements solubilize in copper, and lead to lattice distortion the copper, which result in a big drop of conductivity in copper [2]. Therefore, the copper matrix composite with higher mechanical properties and thermo stability become the hotspot of current research.

Copper matrix composites can be fabricated via many routes, such as power metallurgy, self-propagating high-temperature synthesis (SHS), mechanical alloying, internal oxidation, etc [3-7]. Due to good wetting between ceramic particles and copper matrix by in situ reaction synthesis, preferable integrated performance (i.e. tensile strength and electrical conductivity) is more facilely obtained than the conventional methods [1, 8]. Among various in situ routes, the casting process is of particular interest owing to its lower cost and potential for mass production. Second phase particles dispersion strengthening copper composites are a good way to strengthen copper. Comparing to other ceramic particles, TiB$_2$ particles possess not only good conductivity, thermal conductivity, high hardness, but also excellent chemical stability, corrosion resistance and wear resistance. Moreover, the

standard Gibbs free energy of TiB_2 is low, and could synthesise from boric acid potassium fluoride and fluoride potassium titanate compound salt, or directly generated by titanium and boron at high temperature. However, the traditional methods, such as directly added enhance phase particles to the matrix, or B, Ti in situ reaction, face the same problem of TiB_2 particles aggregation, which will seriously deteriorate the mechanical properties of composites.

Rotating magnetic field (RMF) has been widely used in materials processing to overcome this shortcoming [9, 10]. The liquid metal in the RMF is subjected to electromagnetic stirring, which has the advantages of refining the internal structures of the ingot, increasing the fraction of equiaxed grains and reducing the segregation and shrinkage cavity [11]. It is also inferred from the literature that using electromagnetic stirring in copper melt, the TiB_2 particles may exhibit a homogeneous distribution in the copper matrix. However, until now, the effects of RMF on the particle distribution in copper matrix composites have not been reported.

In this study, the in situ synthesis of TiB_2/Cu composite from Cu-4.6B and Ti with different RMF implied during solidification were prepared. The effect of RMF on the microstructure and mechanical properties of in situ TiB_2/Cu composite were investigated.

2. Experimental procedure

The experiment was performed in a vacuum medium frequency induction melting furnace with a rotating magnetic field. Figure 1 shows a schematic diagram of the experimental apparatus. The raw materials used were electrolytic copper (99.96% purity), titanium sponge (98.5% purity) and Cu-4.6wt%B master alloy.
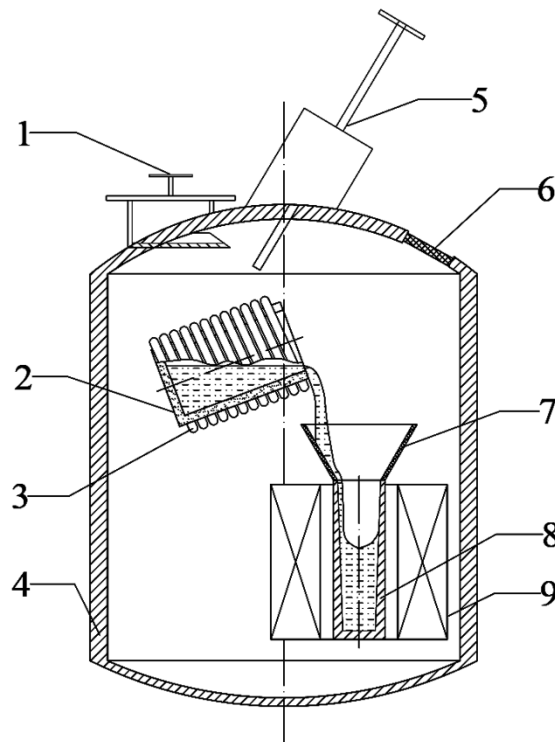

Figure 1. Schematic diagram of a vacuum induction heating furnace with RMF: (1) Hopper, (2) Graphite crucible, (3) Induction coil, (4) Vacuum furnace, (5) Material pounding rod, (6) Hagioscope, (7) Ceramic funnel, (8) Graphitic casting mould, (9) Rotating magnetic field generator.

Pure Cu was melted in a vacuum medium frequency induction melting furnace in an argon atmosphere, when the Cu melt in the furnace reached 1300 °C, Ti and Cu-B master alloys were incorporated into the pure Cu sequentially. After 5 minutes of holding, TiB_2 particles were formed via chemical reactions between Ti and B. Subsequently, the Cu-TiB_2 melt was poured into a cylindrical graphite mould (45 mm in diameter and 220 mm in height), which was inserted in a RMF generator for electromagnetic stirring. The RMF was charged with a frequency of 50 Hz shortly after pouring. The intensity of RMF was controlled by adjusting the alternating current. Four Cu-1wt.%TiB^2 samples were prepared with different current intensities (20 and 40 A, corresponding to the magnetic field intensity of 13 mT and 30 mT, respectively). The intensity of RMF was measured by Gauss/Teslameter Model 7030, Sypris Test & Measurement. For comparison, a reference sample without RMF was also prepared.

The as-cast samples were mechanically ground, polished and etched in a corrosive agent (3g $FeCl_3$, 2 mL HCl and 95 mL C_2H_5OH) for metallographic examination. The microstructure and TiB_2 distribution in the copper matrix were observed under a scanning electron microscopy (SEM, Zeiss supra 55) operated at secondary electron mode with an accelerating voltage of 15 kV. The tensile specimens, with a dimension of 40 mm gage length, 6 mm diameter, were machined from samples according to the GB/T 228-2002 standard. Tensile tests with cross head speed of 2 mm•min^{-1} were conducted at room temperature. Vickers hardness tests were performed under a load of 100 g for 10 s using a Vickers hardness tester (MH-6L). For each test, five measurements were performed and average experimental data was reported.

3. Results and discussion

3.1. Thermomechanical analysis

In order to determine the reaction in the composite melt, the Gibbs free energy of feasible chemical reactions in molten copper was calculated according to the thermodynamic data from thermodynamics manual [12]:
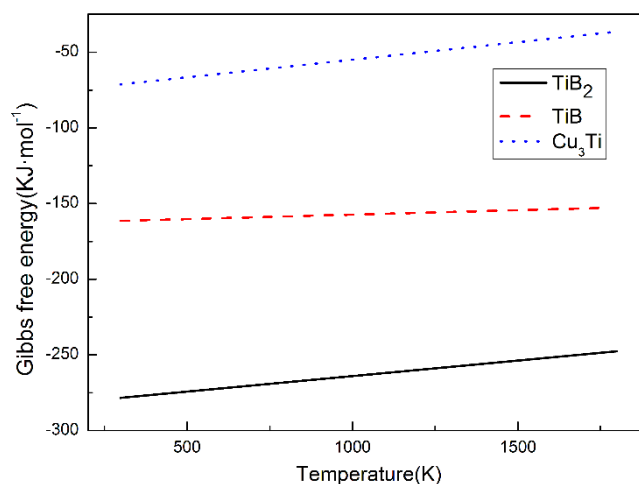

Figure 2. Variation of standard Gibbs free energy of possible reactions vs temperature in Cu-Ti-B melt.

The probable reactions that may exist in melt are as follows:

$$Ti+2[B]Cu \rightarrow TiB_2 \qquad (1)$$
$$Ti+[B]Cu \rightarrow TiB \qquad (2)$$
$$3Cu+Ti \rightarrow Cu_3Ti \qquad (3)$$

By thermodynamics data of elements and compounds, the corresponding equations of Gibbs free energy change with temperature as follows:

$$\Delta G_1^o = -284500 + 20.5T \qquad (4)$$

$$\Delta G_2^o = -163200 + 5.9T \qquad (5)$$

$$\Delta G_3^o = -78200 + 23.2T \qquad (6)$$

The thermodynamic calculation results are shown in figure 2. Due to the lowest Gibbs free energy of TiB_2 among the possible products of TiB, TiB_2, Cu_3Ti, the formation of TiB_2 in molten copper is more susceptible than that of other components. Therefore, only in situ formation of TiB_2 occurred in the copper melt. As shown in figure 3, only TiB_2 phase is determined in the composite.
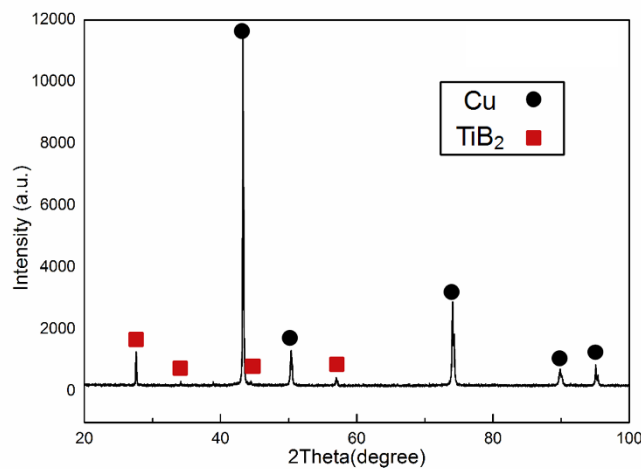

Figure 3. XRD pattern of Cu–1 wt.% TiB_2 composite.

3.2. Microstructure and TiB_2 distribution of Cu/TiB_2 composites
Figure 4 shows the microstructure the SEM micrographs of the distribution of TiB_2 particles in the Cu-1wt.% TiB_2 composites prepared with different field currents ranging 0, 20 A and 40 A.

It is well known that the distribution, size and spacing of reinforced particles in a copper matrix composite are the predominant factors influencing its mechanical properties. The aggregation of coarse TiB_2 particle can be clearly observed in the copper matrix figure 4(a). However, when a 20 A field current was applied, the distribution of TiB_2 in the copper matrix has been greatly improved in the mass. This implies that the RMF can be beneficial to the particle distribution of metal matrix composites. The improved distribution of TiB_2 is attributed to the electromagnetic stirring, which is produced inside the melt by RMF. It should be noted that some aggregates are still observed in this composite in certain local regions, as shown in figures 4(b). With field currents reach to 40 A, an increase in the field current leads to an improvement in the distribution of TiB_2. When a 40 A field current was applied, thanks to the stirring effect of RMF, the initial TiB_2 aggregates in the untreated composite were modified into a more homogeneously distribution throughout the copper matrix (figure 4(c)).

3.3. Mechanical properties
The effect of RMF on the tensile strength, elongation and hardness are shown in figure 5 and table 1. As shown in the figure, the implication of RMF on composite makes an obvious improvement of tensile strength and hardness. The strength increases from 178 MPa, 102HV to 200 MPa, 120.5HV respectively. However, the elongation of Cu-1 wt.% TiB_2 composites decreased a little after the

application of RMF. As is known that the particles are always the crack initiation source in particle reinforced metal matrix composites. Therefore, the presence of TiB_2 particles will limit the ductility of the in situ composites [13]. Without RMF, the aggregation of TiB_2 particles results in more grains with fewer reinforcing particles, which may contribute a lot to the elongation.
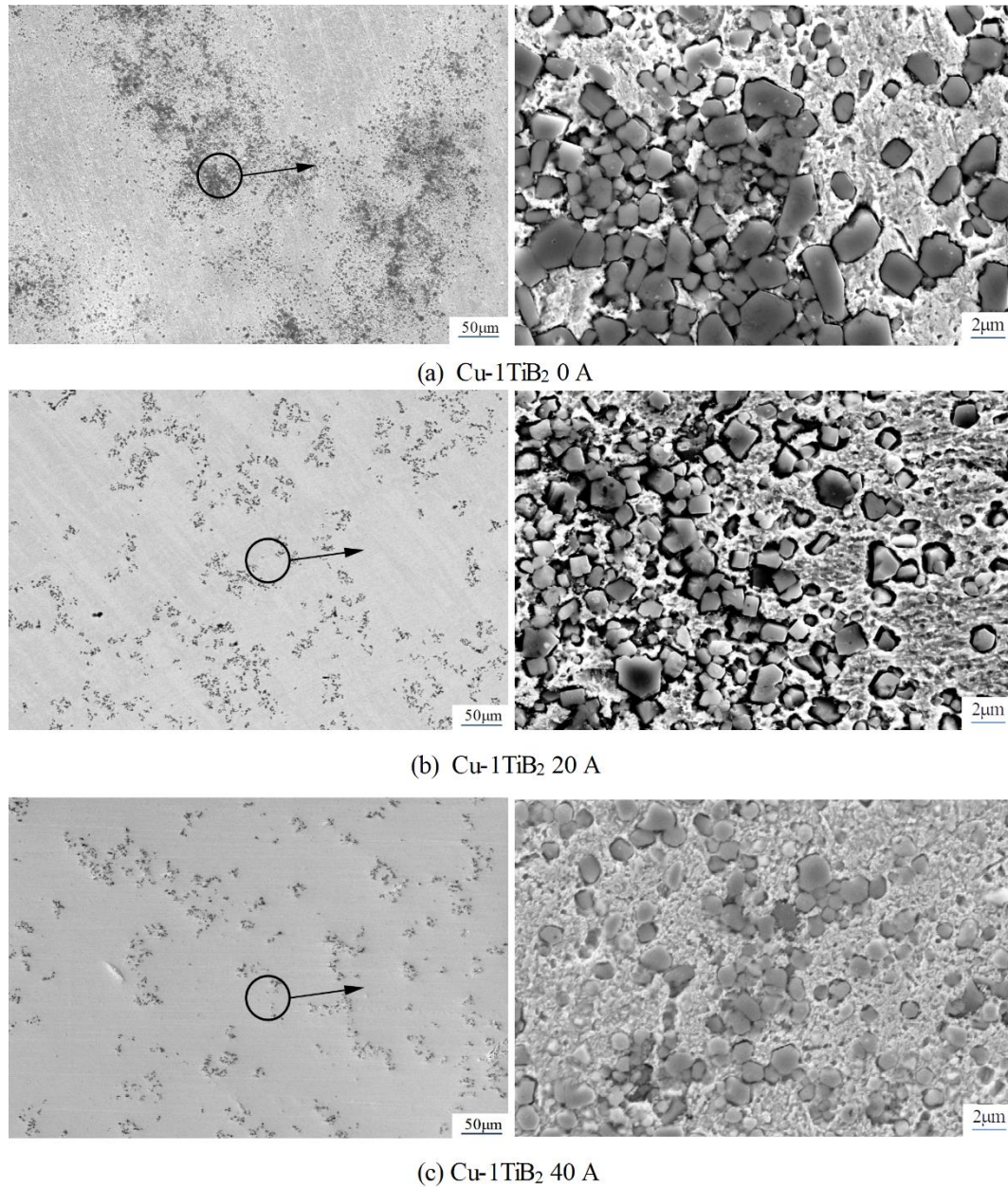

(a) Cu-1TiB$_2$ 0 A

(b) Cu-1TiB$_2$ 20 A

(c) Cu-1TiB$_2$ 40 A

Figure 4. SEM micrographs of Cu-1wt.% TiB_2 composite shows distribution of TiB_2 particles in the matrix under different field currents: (a) without, (b) 20 A, (c) 40 A.

Table 1. Hardness of Cu-1TiB$_2$ under different RMF.

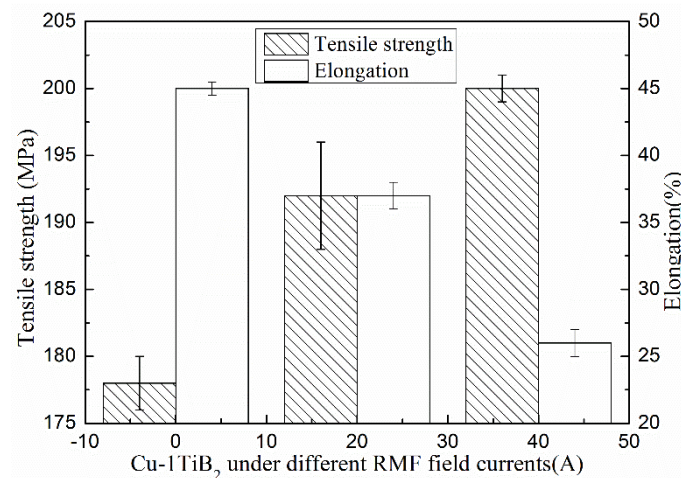| ω% TiB$_2$/Cu	Field Current (A)	Hardness (HV)
	0	102.0±6.4
1	20	108.5±4.3
	40	120.5±3.5

Figure 5. Tensile strength and elongation of Cu-1wt.%TiB$_2$ composites samples under different field currents.

The improvement of mechanical properties of Cu-TiB$_2$ is attribute to the more dispersive distribution of TiB$_2$ in copper matrix. The electromagnetic body force generated by RMF drives forced convection in composite melt. Besides the rotating, a secondary flow in the meridional plane engenders by a radial pressure gradient. The driven flow makes a strong agitation in composite, resulting in a homogenous dispersion of TiB$_2$ particles in the copper matrix, so the well properties Cu-TiB$_2$ composites were acquired.

4. Conclusions

In situ Cu-1TiB$_2$ composites were synthesised under different RMF. The RMF, imposed during the solidification of Cu-TiB$_2$ composite, facilitates a more homogenous distribution of TiB$_2$ particles in the copper matrix and significantly increased tensile strength and hardness. That because the driven flow generated by RMF separate the aggregation TiB$_2$ particles. The homogenous dispersion of TiB$_2$ particles in the copper matrix resulting in well mechanical properties of Cu-TiB$_2$ composites.

Acknowledgements

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