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EFFECT OF SILICON CARBIDE VOLUME FRACTION UPON INTERFACE REACTION IN SiC_p/Al COMPOSITES①

Fan Tongxiang, Shi Zhongliang, Zhang Di and Wu Renjie
State Key Lab of Metal Matrix Composites,
Shanghai Jiao Tong University, Shanghai 200030, P. R. China

ABSTRACT The interfacial reaction behavior during the remelting of SiC_p/Al composite with different volume reinforcement has been studied by liquid metal X-ray diffraction (LMXRD) and differential scanning calorimeter(DSC). Experiment results show that the interface reaction during remelting is associated closely with the reaction during fabrication; chemical reaction is strengthened during fabrication due to high reinforcement volume fraction, but it is not susceptible to the reinforcement fraction and the remelting number during multiple remelting. The mechanism of reaction dynamics which controls the evolution of chemical reaction during remelting is also presented.

Key words SiC_p/Al composite remelting recycling interface reaction reinforcement volume

1 INTRODUCTION

Discontinuously reinforced metal matrix composites are very attractive in aerospace and automotive industry for their properties, such as high modulus, high specific stiffness, high temperature strength, low coefficient of thermal expansion, good wear resistance and good workability and isotropy. The research on the recycling behavior has been hot-spot in the composite materials under the effect of recycling resource and environment protection[1-3], which is also a main problem in the large scale commercialization of the advanced material. The interfacial reaction between the reinforcement and the matrix plays an important role in the microstructure and the properties of resultant composite materials. As a result, the knowledge of the interfacial reaction behavior during the remelt recycling in discontinuously reinforced metal matrix composites is very important in order to bring to light the nature and the controlling methods of the interfacial reaction, which helps to reveal the recycling and reclamation mechanisms and laws. In this field, some research work have been carried out in recent years abroad[1,2], but not profound and entirely. In this paper, the interfacial reaction behavior during the remelting of SiC_p/Al composite with different volume reinforcements has been investigated by using liquid metal X-ray diffraction (LMXRD) and differential scanning calorimeter (DSC) in order to reveal the effect of reinforcement volume fraction upon the interfacial reaction in SiC_p/Al composite during fabrication and remelting.

2 EXPERIMENTAL PROCEDURES

2.1 Material preparation

SiC_p/pure Al composites used in this study were fabricated using vacuum-high pressure infiltration processing. The dominant phase of SiC_p used as the reinforcement was α-SiC(6H) and the average size SiC_p was about 7 μm. Two type of SiC_p/Al composite(10% and 40%SiC) were used to vary the reaction during fabrication and remelting. DSC samples were cut into the disc of d 4 mm × 4 mm in size, and LMXRD samples were cut into the cuboid of 22 mm × 12 mm × 9 mm in size.

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2.2 DSC experiment

For the DSC work, the composite samples were remelted in a high purity alumna pan in Netsch DSC404 instrument. Three runs were carried out at a heating rate 20 ℃/min from ambient to 1 050 ℃, subsequently cooling to ambient temperature at 20 ℃/min, and in dynamic high purity argon atmosphere (80 mL/min). High purity corundum was used as a reference, and all DSC thermograms were normalized to the actual amount of metal (in percentage of mass) in each composites.

2.3 LMXRD experiment

LMXRD was carried out by using θ- θ type liquid metal X-ray diffraction meter in Ukraine academy of science. High temperature X-ray diffractions were carried out in high purity helium atmosphere of 1.3×10^5 Pa before the chamber was cleaned in a vacuum of 2×10^{-6} Pa. Specimen was placed in an alumna crucible of 30 mm × 25 mm × 8 mm in size, using Ta sheet as heating element. Surface of the specimen was fit to one horizontal position using laser calibrator. Some other parameters used in this experiment included: scanning voltage was 40 kV, current was 30 mA, exposition time is 30s, measured angle was from 5° ~ 90°. Compared with solid X-ray diffraction, the liquid metal X-ray diffraction has distinct features, such as the exactness in low angle scanning, the detection of reaction phase with small size and low amount, the controlled scanning steps and the ability to imply the information of solid and liquid phase in molten composite melt at elevated temperature at the same time.

3 RESULTS

In order to real the interfacial reaction feature and the reaction products, low angle scanning with the step of 0.1 ℃ were applied in 5 ~ 20 ℃ in the liquid metal X-ray diffractions. Fig. 1 and Fig. 2 showed the liquid metal X-ray diffraction profiles at ambient temperature for 10 % and 40 % (in volume fraction) SiC_p/Al composite, respectively; the interfacial reaction behavior during fabrication can be implied by monitoring the variation of the diffraction intensity of Al_4C_3 and Si. Fig.1(b) and Fig.2(b) showed the enlarged profiles in the first main peak of Fig.1(a) and Fig.2(a), respectively. The diffraction intensities of Al_4C_3 and Si in Fig. 1 are much more lower than those of Al_4C_3 and Si in Fig.2 in the same peak position.

Fig.3 and Fig.4 showed the liquid metal X-ray diffraction profiles at 775 ℃ during the remelting of 10 % and 40 %(in volume fraction)

Fig.1 Liquid X-ray diffraction profile at ambient temperature for 10 % (Volume fraction) SiC/Al composite

(a) —Liquid X-ray diffraction profile; (b) —Enlarged profile of A marked in the first main peak of Fig.1(a)

Fig.2　Liquid X-ray diffraction profile at ambient temperature for 40 % (volume fraction) SiC/Al composite

(a) —Liquid X-ray diffraction profile; (b) —Enlarged profile of B marked in the first main peak of Fig.2(a)

Fig.3　Liquid X-ray diffraction profile at 775 ℃ during remelting of 10 % (volume fraction) SiC/Al composite

SiC_p/Al composites, respectively, indicating the interfacial reaction feature of the co-existing reinforcing particles and molten aluminum. Compared with those at ambient temperature, the diffraction intensities of Al_4C_3 and Si at 775 ℃ are almost the same.

The DSC heating trace for the multiple remelting of 10 % and 40 % (Volume fraction) SiC_p/Al composites are shown in Fig.5 and Fig.6, respectively. As shown in Fig.5 and Fig.6, the eutectic heat (218.05 J/g) in the first heating trace of 40 % SiC_p/Al composite is more greater than that (174.05 J/g) of 10 % SiC_p/Al composite, the results suggested that the chemical reaction is more serious in 40 %SiC_p/Al composite compared with 10 % SiC_p/Al composite, producing more silicon content and resulting in more greater eutectic heat. On the other hand, the heating trace is progressively fitting with

Fig.4　Liquid X-ray diffraction profile at 775 ℃ during remelting of 40 % (volume fraction) SiC/Al composite

Fig.5 DSC trace for remelting of 10 %SiC_p/ Al composite

Fig.6 DSC trace for remelting of 40 % SiC_p/ Al composite

remelting number increasing, and the variation of the eutectic heat is also progressively increasing with the remelting number increasing, but not greatly.

4 DISCUSSION

4.1 Interfacial reaction during fabrications

The experimental results in Fig.1 and Fig. 2 under the LMXRD and in the first DSC heating trace in Fig.5 and Fig.6 showed that the reinforcers volume fraction plays an important role in interfacial reaction between SiC and Aluminum. The SiC_p/ Al composite used in this experiment is produced under the same circumstance: vacuum-high pressure infiltration processing, and infiltration temperature is 1 023 K, holding temperature and pressure for 5 min. But the diffraction intensities of reaction products Al_4C_3 and Si in Fig.1(a) are much lower than those of in Fig.2(a), respectively. It is suggested that the chemical reaction in the interface is improved due to the high reinforcer volume fraction during fabrication and that the reaction is suitable for common assumption.

Several studies have shown that SiC will react with molten aluminum to form Al_4C_3 and Si according to Eqn.(1):

$$4Al + 3SiC \rightarrow Al_4C_3 + 3Si \qquad (1)$$

Limited only in SiC/ pure Al composite, some efforts[4-9] as shown in Fig.7 have been made to predict Si contents to avoid the interfacial reaction. The results vary significantly depending on authors, but they all reveal that the reactivity between SiC and pure aluminum is active greatly, high Si content is needed to keep balance of thermodynamics. During the fabrication of composite at a given temperature, high reinforcer fraction provides higher contacting

Fig.7 Variation in equilibrium Si content in SiC_p/ pure Al composite plotted as a function of fabrication temperature surface between SiC_p and molten aluminum melt than low reinforcers fraction, resulting in more serious chemical reaction.

4.2 Reaction characteristics in SiC/ Al composite above liquidus during remelting

According to Eqn.(1), SiC can react with molten aluminum, producing Al_4C_3 and silicon. Simensen[10] has established that carbon solubility in molten aluminum, found experimentally by Oden and McCune[11], is less than 2.34×10^{-6}, and the system Al-C-Si can be approximated with the well known binary subsystem Al-Si below 1100 ℃.

The element silicon produced during the interfacial reaction is allocated in the matrix between α-phase primary crystals and eutectic phase (second α-crystals and β-crystals). Thus the total mass of produced silicon is the addition of silicon mass contained in the two phases. If the system is in thermodynamic equilibrium, the chemical composition of the phase is a definite value, according to Ref.[12], the extent of interfacial reaction can be expressed as follow:

$$\theta = \left[1 - s + \left(\frac{r_\beta}{r_\alpha} - 1\right)\left(\frac{m_e}{m_c}\right) \right] \Big/ \left\{ \frac{s}{M_{SiC}} \left[\left(\frac{1}{r_\alpha} - 1\right) M_{Si} + \frac{4}{3} M_{Al} \right] \right\} \quad (2)$$

where r_α and r_β are the silicon fraction contained in the two respective phases respectively. m_e and m_c are the mass of eutectic phase contained in the matrix and the composite mass respectively. M_{SiC} and M_{Si} are the atomic and molecular, mass s is the mass fraction of reinforcement (SiC) in the matrix and θ is the reaction degree, namely the reaction conversion of SiC. Replacing the values of numeral constants, we have, for the composite with a SiC content of 10% and 40%(Volume fraction), as examined material.

$$\theta_1 = 0.179 + 1.33\left(\frac{m_e}{m_c}\right) \quad (10\% \text{ SiC/ Al}) \quad (3)$$

$$\theta_2 = 0.030 + 0.35\left(\frac{m_e}{m_c}\right) \quad (40\% \text{ SiC/ Al}) \quad (4)$$

At the eutectic point of the binary system Al-Si(577 ℃), the β-crystals react with eutectic α-crystals to form liquid alloy. The determination of eutectic phase melting-heat by caloremetry allows to the evaluation of the eutectic amount and the contained silicon. The reaction conversion can be determined by the evaluation of the eutectic amount in the matrix. For unit mass of composite, the amount of eutectic phase contained in the matrix can be estimated calorimetrically by calculation of its melting heat ΔH_f in the DSC heating trace. The eutectic phase mass[12] is

$$m_e = \frac{\Delta H_r}{\Delta \overline{H_f}}$$

where $\Delta \overline{H_f}$ is the specific melting heat. The calorimetric tests provide the thermal flux per gram of composite, ΔH_S, meanwhile, the specific melting heat of the eutectic Al-Si has been evaluated calorimetrically on Al-Si eutectic alloy (12.6% Si, mass fraction) and the resulting value is $\Delta \overline{H_f} = 439.8$ J/g, therefore, Eqns.(3) and (4) become

$$\theta_1 = 0.179 + 1.33\left(\frac{\Delta H_S}{\Delta \overline{H_f}}\right) \quad (5)$$

$$\theta_2 = 0.030 + 0.35\left(\frac{\Delta H_S}{\Delta \overline{H_f}}\right) \quad (6)$$

This method assumes the presence of the eutectic phase in the matrix and a silicon content lower than the eutectic composition. Viala *et al*[9] found the silicon content in the SiC/ Al melt is lower than the eutectic composition, which is a resultant dependent of the matrix composition and particle size and the fabrication conditions when below 1000 ℃. Meanwhile, the solidification features of hypereutectic Al-Si doesn't exist on the DSC heating trace in the present work (as shown in Fig.5 and Fig.6). As a result, Fig.8 shows the calculation results for the interfacial reaction extent θ during multiple-remelting of SiC_p/ Al composites, according to above Eqns. (5), (6) and the experimental results, ΔH, in Fig.5 and Fig.6.

It seems that the chemical reaction should be very serious in SiC_p/ Al composite under multiple remelting due to the active reactivity between SiC and Al at high temperatures. However, in fact the results showed that even SiC particles in 10 % SiC/ Al composite are not exhausted after three multiple remelting, which is agreed with the LMXRD results in Fig. 3 and Fig. 4 when the SiC_p and Al melt are being held at 775 ℃. The experiment about the reaction conversion of SiC particles suggested that during the multiple-remelting the interfacial reaction in SiC/ Al composite isn't sensitive to the reinforcement fraction and the remelting number.

Fig. 8 Interface reaction degree versus remelting number in SiC_p/ Al composite

The reason for this phenomenon lies in that the interfacial chemical reaction is controlled by the dynamics during remelting. The reaction between SiC and Al_4C_3 is believed to be dissolution-diffusion-precipitation steps[13, 14]. The dissolution kinetics of SiC_p in Al has been suggested to be the rate-determining step in SiC_p/ Al interfacial reaction during fabrication of composites. With the SiC_p dissolution, Al_4C_3 grows and Si will diffuse into the melt around Al_4C_3. With time increasing, a layer of Al_4C_3 may form around SiC particles. The Al_4C_3 layer may act as a diffusion barrier for the diffusion of Si, C and Al. On the other hand, the silicon produced from the reaction between SiC_p/ Al formed a solute rich band with much more Si content above liquidus, which puts the local composition of the composite melt in the two-phase Al-SiC field from Al-SiC-Al_4C_3 three-phase field[15]. So further reaction can be avoided and the interfacial reaction in SiC_p/ Al composite during multiple remelting isn't sensitive to SiC fraction and remelting number.

5 CONCULUSIONS

(1) During the fabrication of SiC_p/ Al composites, the chemical reaction is more serious due to a high reinforcer fraction.

(2) During multiple-remelting of SiC_p/ Al composite, the chemical reaction feature is different from that during fabrication, and the interfacial reaction isn't sensitive to the remelting number and reinforcement fraction;

(3) During the remelting of SiC_p/ Al composite, the interfacial reaction feature depends on the kinetics factors.

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