Infiltration kinetics of pressureless infiltration in SiC_p/ Al composites [①]

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Abstract: The pressureless infiltration kinetics was investigated by plotting the infiltration distance as function of the infiltration time. The effects of key process parameters such as time, temperature, Mg content on the pressureless infiltration of silicon carbide particle compacts were studied and quantified. The preform with high volume fraction SiC was obtained by mixing SiC particles with bimodal size distribution, whose diameters are 5 and 50 μm, respectively. The results show that an incubation period exists before infiltration, the influence of temperature on the incubation time exceeds that of Mg content, infiltration rate increases with the increasing temperature and Mg content, infiltration rate decreases as Mg consumes. A model of macroscopical infiltration and microscopical infiltration of liquid alloy in porous SiC preform was proposed.

Key words: SiC_p composites; metal matrix composites; pressureless infiltration; kinetics; electronic packaging

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1 INTRODUCTION

Aluminum infiltrated silicon carbide composite (SiC_p/ Al) exhibits excellent properties such as high thermal conductivity, low coefficient of thermal expansion(CTE), high modulus and low density, therefore it has extensive potential applications in electronic packaging and thermal management[1]. Electronic packaging materials are required to possess compatible CTEs with the IC, thus the composites with high volume fraction of SiC particles are preferred[2]. Methods to fabricate high volume fraction composites include pressure infiltration[3, 4], liquid-exchange[5] and pressureless infiltration[6, 7]. During pressureless infiltration, porous preform can be infiltrated by liquid metal without the aid of an externally applied force. The method is low-cost and simple alternative, thus it obtains close attention in recent years. Sound composites with high density and excellent properties can be produced by the pressureless infiltration. Recent investigation on pressureless infiltration focus on SiC particle reinforcement with single size[6−9], however, pressureless infiltration with bimodal size particles distribution is little touched upon. It is difficult to infiltrate the SiC preform with bimodal size particles distribution because of the inconstant porosity. To obtain good infiltration, it is very important to understand the infiltration kinetics of pressureless infiltration in SiC_p/ Al composites. In this paper, incubation period and infiltration rate during pressureless infiltration are investigated.

2 EXPERIMENTAL

Al-Si alloy was used as the matrix material, magnesium was added to help infiltration at the levels of 3%, 6%, 10% and 12%, while SiC particles with bimodal size at the average size of 50 μm and 5 μm were used as the reinforcement, the particle volume fractions(v_p) of preform were 70%. The rectangle preform (60 mm × 30 mm × 10 mm) was produced by mixing, forming, and sintering. The sintering of SiC particles was carried out in an electric furnace with open air at a pre-set temperature of 1 200 ℃ for 2 h. The infiltration experiment is shown in Fig. 1. After preheated, preform was pushed into the Al-Mg-Si liquid alloy at 800, 900, 950, 1 000 and 1 100 ℃, respectively. The system was run at a very slight overpressure to ambient under N_2 atmosphere. Infiltration rates were figured out by infiltration distance (height) as a function of infiltration time.

3 RESULTS AND DISCUSSION

Fig. 2 shows the distribution of coarse and fine SiC particles and the distribution of pores in a composite produced by the infiltration parameters as temperature 1 000 ℃, 70% v_p, 10% Mg in alloy and 2 h. The value of v_p is easily changed from 56% to
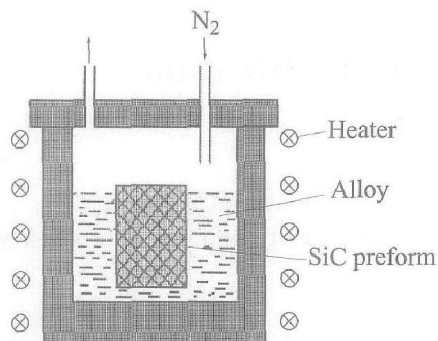
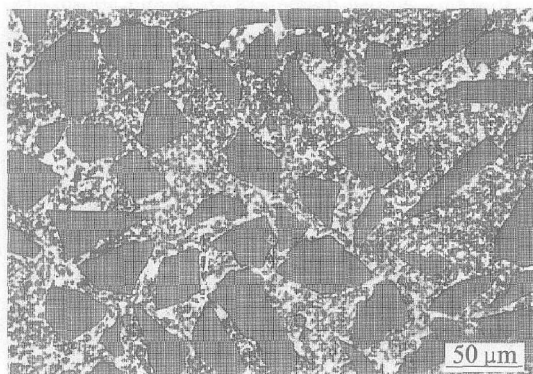**Fig. 1**   Schematic diagram of infiltration

Fig. 2   Distribution of SiC particles in composite

75% through varying the proportion of coarse to fine particle.

3. 1   Incubation period

When the preform was contacted with the alloy, a period that was called incubation period before infiltration was found. For a given temperature, the incubation time is the minimum time required to initiate infiltration. In the experiment, the incubation time is primarily controlled by infiltration temperature and Mg content.

Figs. 3 and 4 show the effects of temperature and Mg content on the incubation time. Fig. 3 shows that incubation time decreases from 45 min to 6 min as temperature increases from 800 ℃ to 1 100 ℃ at 10% Mg. When the temperature increases to 900 ℃, the incubation time decreases to 20min, indicating that a sharp decline of incubation time appears at the temperature between 800 and 900 ℃. When the temperature varies from 1 000 to 1 100 ℃, the decrease of incubation time is only from 8 to 6 min, which shows that there is not significant effect on incubation time when the temperature is over 1 000 ℃. It is shown in Fig. 4 that the incubation time decreases from 18 to 6 min at 1 000 ℃, as Mg content increases from 3% to 12%. It is easily found that the effect of temperature on incubation time exceeds that of Mg content.

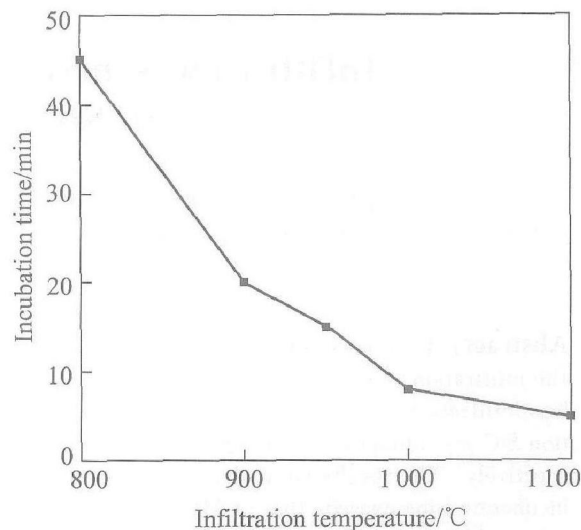Eqn. (1) shows the chemical reaction on the surface of preoxied SiC particles during incubation

Fig. 3   Effect of infiltration temperature on incubation time
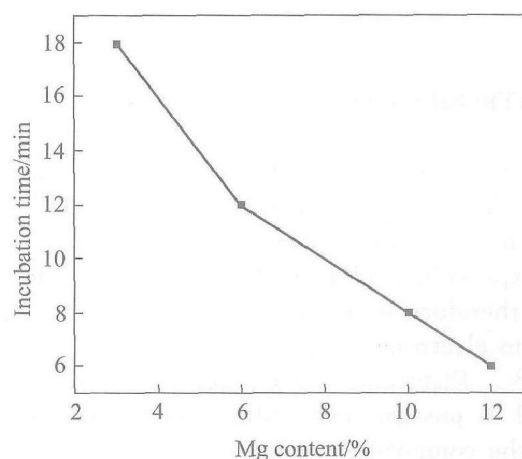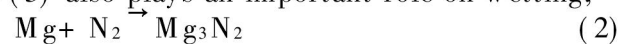

Fig. 4   Effect of Mg content on incubation time

period, the reaction promotes the wetting ability between SiC and aluminum alloy[10, 11] and thus induce the infiltration:

$$SiO_2 + Mg + Al \xrightarrow{} MgAl_2O_4 + Si \qquad (1)$$

Many researchers[8] indicate that Eqns. (2) and (3) also plays an important role on wetting,

$$Mg + N_2 \xrightarrow{} Mg_3N_2 \qquad (2)$$

$$Al + Mg_3N_2 \xrightarrow{} AlN + Mg \qquad (3)$$

The increase of Mg content promotes Eqn. (1) along positive direction reaction, and the increase of temperatures decreases the Gibbs free energy. Furthermore, the increases of both Mg content and temperatures decrease the alloy viscosity and liquid-vapor surface tension[6, 12], and consequently decrease the incubation time.

3. 2   Infiltration rate

The infiltration in the porous preform is not regular and stable. Fig. 5 gives a model of the typical infiltration process. The gray region, dark gray region and white region represent the infiltrated area, the congregation of fine particles and the por-

ous preforms, respectively. The process is also shown in Fig. 6. At first, the large pores among coarse particles in the preform are infiltrated, and this process is called macroscopical infiltration. And then the small pores among fine particles are gradually infiltrated, and this process is called microscopical infiltration.
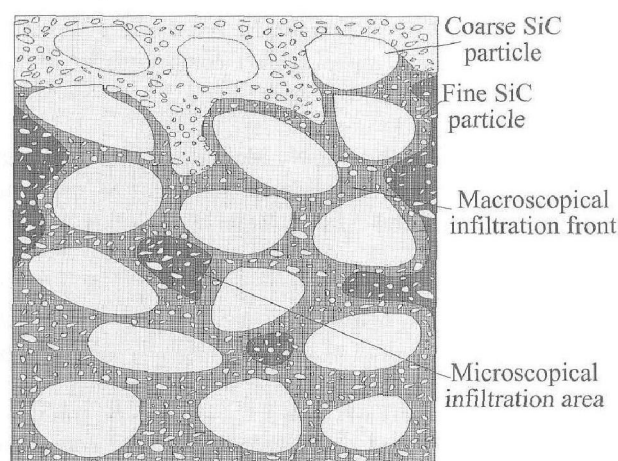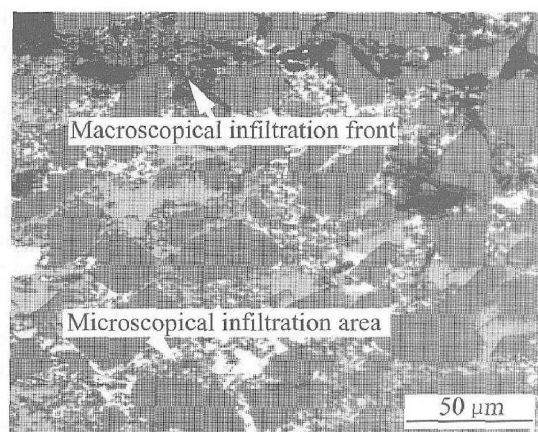

Fig. 5   Infiltration model

Fig. 6   Optical micrograph of infiltration front

The above infiltration processes can be explained according to the equation proposed by Semlak and Rhines[13], as shown in Eqn. (4):

$$L = \frac{2}{\pi}\left[\left[\frac{R_c\,\gamma_{lv}\cos\theta}{2\eta}\right]t\right]^{1/2} \qquad (4)$$

where R_c is the radius of the pore that will be infiltrated by the liquid phase, γ_{lv} is the liquid-vapor surface tension, θ is the contact angle, and η is the viscosity of liquid alloy. The value of R_c among coarse particles is 10 times than that among fine particle, thus infiltration rate of liquid alloy in large pores is about three times than that in small pores. Besides, the gas among coarse particles can be gradually expelled by liquid alloy, but the gas among fine particles must be consumed by surrounding liquid alloys that have enveloped the fine particles.

The effects of Mg content and temperature on

the macroscopical infiltration are shown in Figs. 7 and 8. The infiltration rate increases as Mg content and temperature increase. When infiltration temperature is 1 000 ℃ and Mg content is over 6% for 2 h, the preform can be infiltrated completely. When Mg content is over 10%, the infiltration rate is quickened. It is known that the completion of macroscopical infiltration does not mean the infiltration period is over, because it requires longer time to carry through microscopical infiltration. When Mg content is over 10% and temperature is over 1 000 ℃ for 2 h, the infiltration is accomplished.
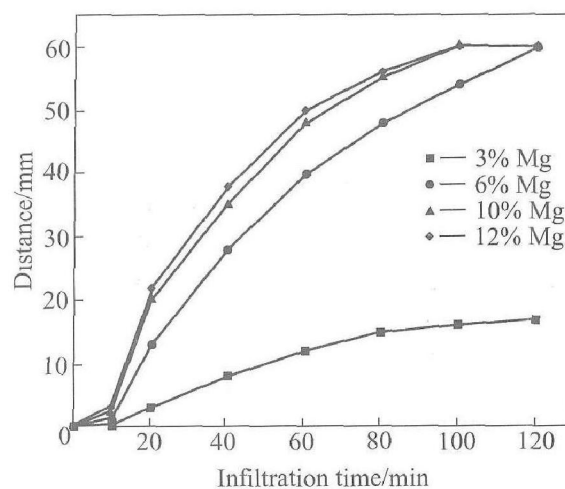

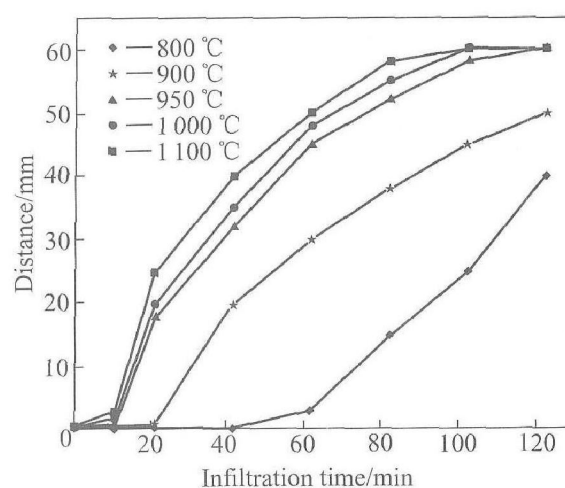**Fig. 7**   Effect of Mg content on infiltration rate

Fig. 8   Effect of temperature on infiltration rate

According to Eqns. (1) and (4), the increase of temperature can increase the chemical reactions at interface between the liquid alloy and SiC preform, and thus promotes the infiltration rate. Mg is a powerful surfactant and scavenges any oxygen that may be present at the SiC/Al interface[13], therefore decreases the fine pores among the fine particles. The infiltration rate is influenced by Mg

content due to its change of chemical composition during infiltration. At 1 000 ℃, Mg content is decreased from 10% to 5% after infiltration by 2 h, this decrease is due to its low vapor pressure. Part Mg vapor is condensed on the furnace wall as Mg, MgO or Mg$_2$N$_3$ and part is taken out by the flowing nitrogen[14]. The infiltration rate decreases after 60 min resulted from Mg loss after long time infiltration.

4 CONCLUSIONS

1) There exists an incubation period which is determined by the chemical reactions between molten alloys and silicon oxide at SiC particle surfaces. When the chemical composition of silicon is fixed, the period will decrease with the increase of infiltration temperature and the chemical composition of magnesium.

2) The infiltration rate of molten alloys into porous SiC is related to the loss of magnesium and the temperature. When the atmosphere is inert, the rate will decrease severely by the loss of magnesium and lower temperature(800~1 100 ℃).

3) A model of macroscopical infiltration and microscopical infiltration of liquid alloy in porous SiC preform was proposed.

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