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Microstructure and thermal conductivity of submicron Si_3N_4 reinforced 2024Al composite

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Abstract: An 2024Al matrix composite reinforced with 36%(volume fraction) β-Si_3N_4 particles was fabricated by pressure infiltration method, and its microstructure and the effect of annealing treatment on thermo-conductivity were discussed. Si_3N_4 particles distribute uniformly without any particle clustering and no apparent particle porosity or significant casting defects are observed in the composites. The combination of particles and matrix is well. The raw Si_3N_4 particles are regular cylindrical polyhedron with flat surface and change to serrated surface in composite due to reactions during fabrication. Thermal conductivity of as-cast Si_3N_{4p}/2024 composite is 90.125 W/(m·K) at room temperature, and increases to 94.997 W/(m·K) after annealing treatment. The calculated results of thermal conductivity of the Si_3N_{4p}/Al composite by Maxwell model, H-S model and PG model are lower than experimental results while that by ROM model is higher.

Key words: composite; interface; thermal conductivity; calculation; submicron particles

1 Introduction

Silicon nitride (Si_3N_4) has a good thermal and chemical stability, high mechanical strength and hardness, and good wear, creep, and corrosion resistance. It attracts research community[1−3]. The Si_3N_4 particles reinforced aluminium matrix composites (Si_3N_{4p}/Al) present several advantages that are high specific strength, low thermal expansion coefficient, high thermal conductivity, high intensity and good dimensional stability, which make them very attractive for applications in aerospace, automotive, electronics, machinery manufacture industry [4−6].

However, the major obstacle for fabricating high performance Si_3N_{4p}/Al composite is the poor wettability of Si_3N_4 particles to liquid aluminium[7]. Fortunately, pressure infiltration method is an effective technique to fabricate poor-wetting system via forcing infiltration of molten metal alloy under high pressure, and it has the advantages of simple fabrication technology, high density and no restrictions on alloy[8]. In this work, 2024Al matrix composite reinforced with 36%(volume fraction) β-Si_3N_4 particles (about 0.2 μm) was fabricated by pressure infiltration method, the microstructure characteristics of Si_3N_{4p}/2024 composite and the interactions between Si_3N_4 and aluminium were studied, the effect of annealing treatment on thermal conductivity was discussed.

2 Experimental

The β-Si_3N_4 particles with an average particle size of 0.2 μm, as shown in Fig.1, were used to reinforce 2024Al alloy by pressure infiltration method, and the chemical compositions of the alloy are Cu 4.79%, Mg 1.49%, Mn 0.611%, Fe 0.245%, Si 0.168%, Zn 0.068%, Cr 0.049%, Ti 0.046%, Ni 0.013% (mass fraction) and Al balance.

The microstructure analysis was conducted by Philips CM-12 and JEOL 200CX transmission electron microscope (TEM) with an accelerated voltage of 100−120 kV and 200 kV, respectively. The thermal conductivity and specific heat capacity of composite and matrix with diameter of 12.7 mm and length of 3 mm were examined on a laser thermal conduction analyzer (JK2, Germany) from 25 to 500 ℃ with a rate of 5 ℃/min.

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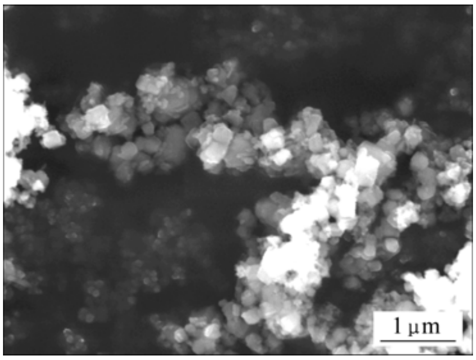

Fig.1 SEM image of Si_3N_4 particles

3 Results and discussion

It is well established that a dense microstructure is beneficial to thermal conductivity. The optical microstructure of the as-cast Si_3N_{4p}/2024 composite is shown in Fig.2. It can been seen from Fig.2 that Si_3N_4 particles distribute uniformly without any particle clustering and no apparent porosity or significant casting defects are observed in the composites.

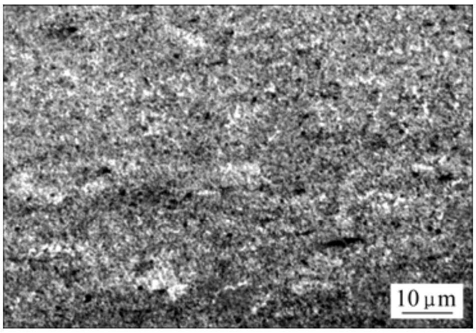

Fig.2 Optical microstructure of as-cast Si_3N_{4p}/2024 composite

TEM images of Si_3N_4 particles is shown in Fig.3. It can been seen from Fig.3 that the Si_3N_4 particles in matrix alloy distribute uniformly. The distance between particles is very small due to high volume fraction, and the combination of particles and matrix is good and no separation between Al and Si_3N_4 particles is observed. Generally, high density of dislocations is always found in aluminium composites reinforced with micron particles because of large thermal mismatch stress generated by coefficient of thermal expansion (CTE) difference between the ceramic particles and matrix [9−10]. However, it is difficult to observe the dislocation in Al matrix reinforced with submicron Si_3N_4 particles although the CTE difference of them is also extreme. It accords to other researches in submicron particles reinforced composite[11−12].

It should be noted that the raw Si_3N_4 particles (see Fig.1) are regular cylindrical polyhedron with flat surface.

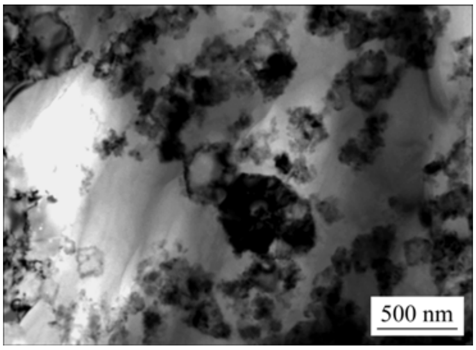

Fig.3 TEM image of Si_3N_4 particles

However, its surface changes to be serrated after composite fabrication (see Fig.4) and all reactions are as follows[13]:

$$Si_3N_4(s)+O_2(g)\longrightarrow 3SiO_2(s)+2N_2(g) \quad (1)$$

$$3SiO_2(s)+4Al(l)\longrightarrow 2Al_2O_3(s)+3Si(l) \quad (2)$$

$$SiO_2(s)+2Mg(l)\longrightarrow 2MgO(s)+Si(l) \quad (3)$$

$$2SiO_2(s)+Mg(l)+2Al(l)\longrightarrow MgAl_2O_4(s)+2Si(l) \quad (4)$$

$$4Al(l)+Si_3N_4(s)\longrightarrow 4AlN(s)+3Si(l) \quad (5)$$

$$2Mg(l)+Si(l)\longrightarrow Mg_2Si(s) \quad (6)$$

The above reactions are all possible in thermodynamics and the thermodynamic stability or

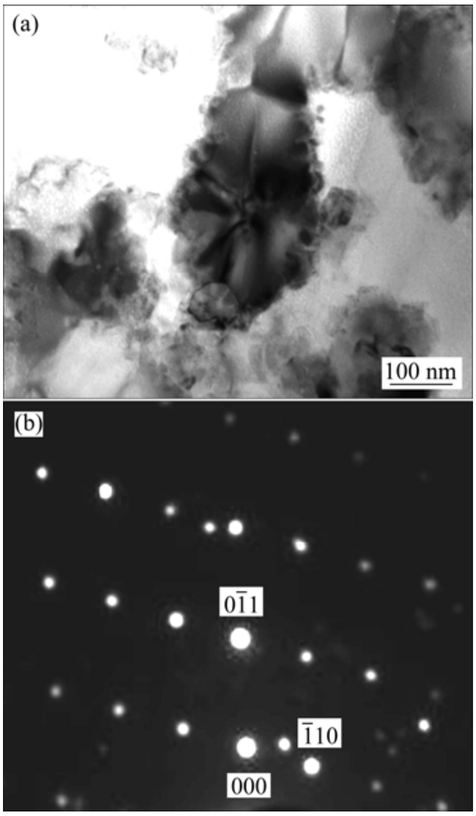

Fig.4 TEM image of Si_3N_4 particle and interface (a) and electron diffraction pattern of Si_3N_4 particle (b)

kinetics needs to be in consideration for confirming the actual reaction. Unfortunately, phonons movement will be impeded by the interface and the interfacial reactant, which eventually performs negative effect on thermal conduction.

Thermal conductivity λ is calculated by following equation:

$$\lambda=\alpha\cdot C_p\cdot\rho \tag{7}$$

where α, ρ and c_p are referred to thermal diffusivity, density and specific heat capacity, respectively. The density of Si_3N_{4p}/2024 composite is 2.9 g/cm^3, and test results of as-cast and as-annealing are listed in Table 1.

Table 1 Thermal conduction testing results of Si_3N_{4p}/2024 composite

Composite	Specific heat capacity/ ($J\cdot g^{-1}\cdot K^{-1}$)	Thermal diffusivity/ ($mm^2\cdot s^{-1}$)	Thermal conductivity/ ($W\cdot m^{-1}\cdot K^{-1}$)
As-cast	1.145	27.142	90.125
As-annealed	1.039	31.528	94.997

There are two ways for heat transfer in Si_3N_{4p}/2024 composite, free electron in Al matrix and phonon in Si_3N_4 particles. Both movements would be scattered by interface. Therefore, heat conduction in Si_3N_{4p}/2024 composite depended on Al matrix, Si_3N_4 particles and their interface. The annealing treatment would release residual thermal stress in Al matrix generated in fabrication, which is beneficial to heat conduction, and then improve the thermal conductivity of Si_3N_{4p}/2024 composite.

One significant advantage of composites is their designable. The accurate prediction of composite performance before fabrication or control of component to meet performance requirement, which would shorten the design and fabrication cycle of composites, is attracting all researchers on composite. However, accurate prediction must be based on the accurate theoretical foundation. The main theoretical prediction models for the thermal conductivity of composite include Maxwell model, H-S model and PG model.

1) Maxwell considers the effect of matrix conduction, particle conduction and particles fraction while particle is simplified as spherical particle, and the following equation can be obtained:

$$\kappa_{\mathrm{com}}=\frac{\kappa_{\mathrm{m}}\left[1+2\dfrac{\kappa_{\mathrm{m}}}{\kappa_{\mathrm{p}}}-2\varphi_{\mathrm{p}}\left(\dfrac{\kappa_{\mathrm{m}}}{\kappa_{\mathrm{p}}}-1\right)\right]}{1+2\dfrac{\kappa_{\mathrm{m}}}{\kappa_{\mathrm{p}}}+2\varphi_{\mathrm{p}}\left(\dfrac{\kappa_{\mathrm{m}}}{\kappa_{\mathrm{p}}}-1\right)} \tag{8}$$

where κ_{com}, κ_{m}, κ_{p} are referred to thermal conductivities of composite, matrix and particle, respectively; φ_{p} is volume fraction of particle.

2) Based on ESHELBY's equivalent inclusion theory[14], HATTA and TAYA[15] consider the effect of particle shape and obtained the general expression of thermal conductivity, which can be expressed as follows for particle reinforced composite:

$$\kappa_{\mathrm{com}}=\kappa_{\mathrm{m}}\times\left[1+\frac{\varphi_{\mathrm{d}}}{\dfrac{1}{3}(1-\varphi_{\mathrm{d}})+\dfrac{\kappa_{\mathrm{m}}}{\kappa_{\mathrm{d}}-\kappa_{\mathrm{m}}}}\right] \tag{9}$$

3) KLEMENS[16] derived the relationship among thermal conductivity of composites, thermal conductivity of matrix, thermal conductivity and volume fraction particle of as follows:

$$\kappa_{\mathrm{com}}=\kappa_{\mathrm{m}}(1-\varphi_{\mathrm{p}})+K_{\mathrm{p}}\varphi_{\mathrm{p}}-\frac{1}{3}(1-\varphi_{\mathrm{p}})\varphi_{\mathrm{p}}\frac{(\kappa_{\mathrm{m}}-\kappa_{\mathrm{p}})^2}{\kappa_{\mathrm{m}}(1-\varphi_{\mathrm{p}})+\kappa_{\mathrm{p}}\varphi_{\mathrm{p}}} \tag{10}$$

The models mentioned above were adopted to calculate thermal conductivity of Si_3N_{4p}/2024 composite and the theoretical values and experimental value are listed in Table 2.

Table 2 Theoretical and experimental thermal conductivities of as-annealed Si_3N_{4p}/2024 ($W\cdot m^{-1}\cdot K^{-1}$)

Maxwell model	H-S model	PG model	ROM model	Experimental
83.051	83.051	91.140	100.400	94.997

It is obvious that the calculated values are different from values by models, and the values calculated by Maxwell model, H-S model and PG model are lower than experimental results while the value by ROM model is higher.

4 Conclusions

1) Si_3N_4 particles distribute uniformly without any particle clustering and no apparent porosity or significant casting defects are observed in the composites. The distance between particles is very small due to its high volume fraction and dispersive distribution, and the combination of particles and matrix is good.

2) The raw Si_3N_4 particles are regular cylindrical polyhedron with flat surface and its surface changes to be serrated due to reactions during composite fabric.

3) Thermal conductivity of casting Si_3N_{4p}/2024 composite is 90.125 W/(m·K) at room temperature, and after annealing treatment, it will improve to 94.997 W/

(m·K). The calculated values by thermal conductivity of Maxwell model, H-S model and PG model are lower than experimental result while value by ROM model is higher.

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