

Fabrication and properties of Si/Al interpenetrating phase composites for electronic packaging

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Abstract: In order to improve the thermal properties of MMCs for electronic packaging, the concept of fabrication MMCs with particular interpenetrating phases(IPCs) was proposed. Based on the diffusion theory of reinforcement element in matrix alloys of some particular PMMCs, a novel fabrication method to produce IPCs was proposed. The Si/Al composites (65%Si, volume fraction) with interpenetrating phases were fabricated successfully by squeeze casting and hot press sintering technology. Microstructure observations indicate that the reinforcements Si are of three-dimensional continuous network and the composites are compact without obvious defects. The average linear thermal expansion coefficient (CTE) between 20 °C and 100 °C of the Si/Al IPCs is $8.27 \times 10^{-6}/\text{K}$, and the thermal conductivity(TC) is 124.03 W/(m·K), and the composites can meet the demands of electronic packaging. ROM model and Turner model can be used to predict the CTEs of IPCs, and the experimental CTEs are between their theoretical and calculated values.

Key words: fabrication method; interpenetrating phase composites; Si/Al alloy; electronic packaging

1 Introduction

With the rapid development of electronic technology, materials with high properties for electronic packaging are needed urgently[1–3]. Many particles reinforced metal matrix composites(PRMMCs) for electronic packaging with low coefficient of thermal expansion(CTE), high thermal conductivity(TC) and high strength, such as SiC_p/Al [4], AlN_p/Al [5], Si_p/Al [6], can meet the demands of electronic packaging ($\alpha < 7 \times 10^{-6} - 9 \times 10^{-6}/\text{K}$ and $\lambda > 100 \text{ W}/(\text{m}\cdot\text{K})$)[1–3]. But the TC of composites is declined by the thermal resistance of interfaces existing in composites[7–9]. Therefore, it will improve the thermal conductivity of composites by decreasing the interfacial area in composites[10–12]. The method to decrease the interfacial area of PRMMCs is to make the particle reinforcements be connected[10], and then the composites with interpenetrating phase become interpenetrating phase composites (IPC). The theoretical study results about thermal expansion of IPCs show that the CTEs of IPCs are large than those of PRMMCs with

the same volume fraction of reinforcement[11]. Therefore, the concept that interpenetrating phase composites (IPC) used for electronic packaging of MMCs has been proposed, but there are just few studies about this field[10].

Nowadays, the studies about IPCs are focused mainly on fabrication process, mechanical properties and theory models about some properties, and most are about ceramic matrix composites and polymer matrix composites[11, 13–18]. The most common method of fabricating IPCs is infiltrating liquid metal into a porous ceramic[13–15] or polymer performs[19–21]. The way, however, is essentially restricted by the manufacturing of performs, whose open porosities are often very low[17–18], and liquid metal cannot be infiltrated sufficiently into preforms[10, 22–23]. In the present work, a novel method was proposed to fabricate Si/Al IPCs by squeeze casting and hot press sintering technology[10], which utilizes the diffusion of reinforcement element in metal matrix. Their microstructure and thermal properties were investigated primarily.

2 Experimental

2.1 Fabrication of IPCs

The reinforcements used in this work were angular-shaped Si particles with nominal diameter of 10 μm , and the volume fraction of the reinforcements was 65%. The matrix alloy was an Al-Si-Cu-Mg alloy (4023Al), whose nominal compositions are listed in Table 1. The properties of Si particles and 4023Al alloy are listed in Table 2.

The fabrication process of Si/Al interpenetrating phase composites is illustrated in Fig.1.

Table 1 Chemical compositions of 4023Al alloy (mass fraction, %)

Si	Cu	Mg	Fe	Ni	Al
11.99	0.5–1.3	0.8–1.3	1.0	0.5–1.3	Bal.

Table 2 Properties of Si particles and 4023Al alloy

Material	Density/ ($\text{g}\cdot\text{cm}^{-3}$)	α / (10^{-6}K^{-1})	λ / ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)	E / GPa
Si	2.33	4.1	148	112.4
4023Al	2.68	20.8	213	69

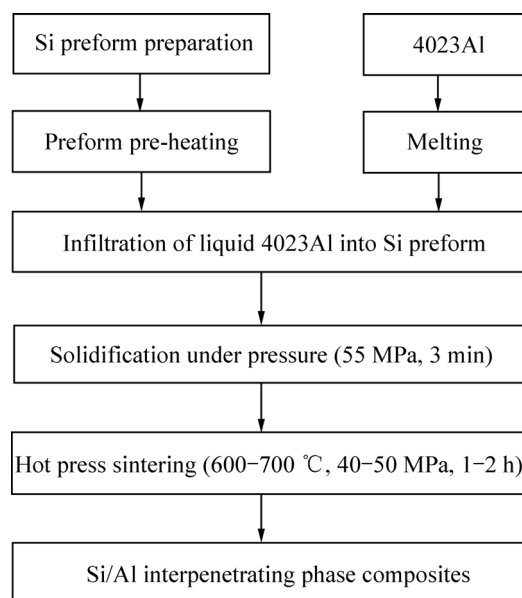


Fig. 1 Fabrication process of Si/Al IPCs

Firstly, Si particles were filled and pressed into a steel mold to produce an Si preform according to the given volume fraction, and then the preform was pre-heated at 500–600 $^{\circ}\text{C}$ in a die. Meanwhile, the 4023Al alloys was melt, degassed, cleaned in a graphite crucible and heated to 750–800 $^{\circ}\text{C}$. Then, the molten 4023Al alloys were poured into the steel die and a

vertical pressure up to 55 MPa was applied immediately. The pressure was maintained for 3–5 min until the solidification was completed. Finally, the composites were treated under the conditions of 600–700 $^{\circ}\text{C}$, 40–50 MPa, for 1–2 h in a hot press sintering furnace. The furnace was filled with N_2 gas to avoid oxidation.

2.2 Experimental procedures

The microstructure of composites was observed on an OLMPUSPMG3 optical microscope. The bulk density of Si/Al IPCs was measured by Archimedes' method and compared with the theoretical density to obtain relative density. The CTE was obtained by NETZSCH DIL402C thermal analyzer, with scanning from 25 to 495 $^{\circ}\text{C}$ at a rate of 5 $^{\circ}\text{C}/\text{min}$. The specimen size was d 6 mm $\times$ 25 mm, and two ends of it were ground with precision grinder. TC was measured by the laser flash method with thermal diffusivity NETZSCH LAF427 made in Germany. The tested temperature was 25 $^{\circ}\text{C}$. The specimen size was d 12.7 mm $\times$ 3 mm, and two sides were polished.

3 Results and discussion

3.1 Microstructure

The OM microstructures of Si/Al IPCs made by squeeze casting and hot press sintering technology are shown in Fig.2. The reinforcements of composites, no matter on the horizontal section or vertical section of composites, are composed of a particular three-dimensional continuous network structure. Therefore, the original angular-shaped Si particles have connected and become a three-dimensional continuous network structure. So Si/Al IPCs could be fabricated by squeeze casting and hot press sintering technology.

From the Al-Si binary eutectic phase diagram[10], we can know that the Al-Si eutectic temperature is 577 $^{\circ}\text{C}$ and Al-Si eutectic point is 12.2%Si(molar fraction). The solubility of Si in Al under eutectic temperature is 1.56%(molar fraction), but that of Al in Si is almost zero. There is no compound in Al-Si system and the wettability of it is good[6]. According to Gibbs-Thomas theory, the concentration of Si in Al around smaller Si particles is higher than that of Si in Al around larger Si particles, so the diffusion happens. Therefore, during the process of hot press sintering, the material transfer goes on by the Si diffusion, and the original angular-shaped Si particles change slowly into a three-dimensional continue network structure.

3.2 Density

The measured density of composites is 2.35 g/cm^3 , and the relative density is 95.41%. Therefore, Si/Al IPCs fabricated by this method are compact. During the process of hot press sintering, the temperature (600–

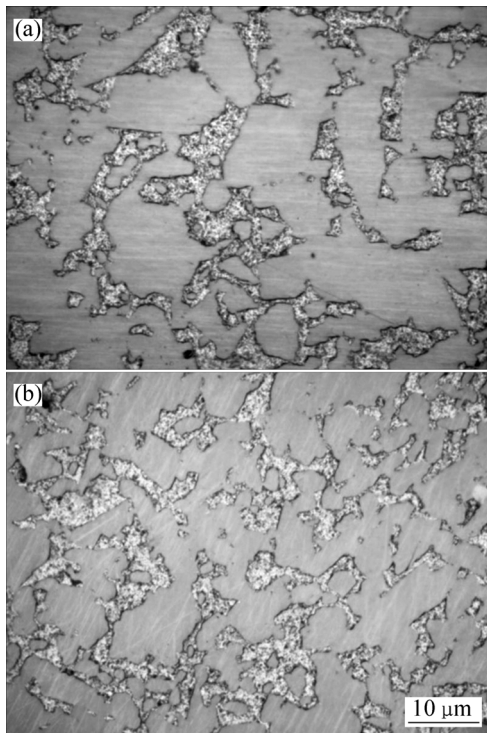


Fig.2 OM microstructures of Si/Al IPCs: (a) Horizontal section; (b) Vertical section

700 °C) is higher than the eutectic temperature(577 °C), so the matrix alloys is melted and expanded (because the volume of liquid state of the same material is larger than that of solid state). After the matrix alloys solidify, there are some small pores left in Si/Al IPCs, as shown in Fig.2. So the relative density of Si/Al IPCs(65%, volume fraction) is lower than that of Si_p/Al(65%, volume fraction) fabricated by squeezing casting technology[6].

3.3 Thermal expansion

The measured CTE of Si/Al IPCs is $8.27 \times 10^{-6}/\text{K}$ between 20 °C and 100 °C, which can greatly meet the demands of electronic packaging. The CTE of composites is determined commonly by that of reinforcements and matrix alloys. As the temperature is increased, both of reinforcements and matrix alloys will expand, but the expansion ability of matrix alloys is larger than that of reinforcements, so the expansion of matrix alloys will be restricted. Because of the three-dimensional continuous network structure of IPCs, the expansion of matrix alloys will be more rigidly restricted, and the CTE of composites with a three-dimensional continuous network reinforcement will be declined. Besides, as the temperature is increased, part of matrix alloy will expand into pores, thus limiting the increase of its bulk volume. The result of SHEN's theory study[11] also shows that the CTE of IPCs is much lower than that of PRMMCs with the same reinforcement volume

fraction.

As shown in Fig.3, the measured CTE of Si/Al IPCs is compared with the theoretical prediction values of ROM model and TURNER[24] model.

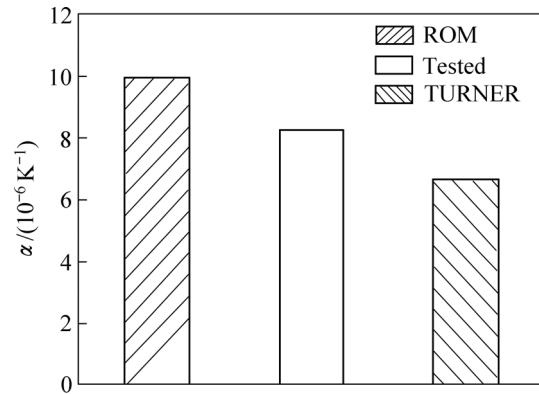


Fig.3 Comparison of CTEs of Si/Al IPCs between experimental value and theoretical prediction values

As shown in Fig.3, the experimental CTE is between the calculated values of ROM and TURNER models. ROM model neglects the restrict effect of the matrix on the particle deformation, while in real materials the deformation restrain effect is much higher. Therefore ROM model can not completely describe the real internal stress conditions in composites, and the results are always larger than the tested results. TURNER model only considers the conditions in material under isopressure, while the internal stress in real material is far more complicated; furthermore, TURNER model also neglects the internal stress generated in the cooling process from high preparing temperature. So TURNER model cannot sufficiently describe the real internal stress conditions in composites either, and its theoretical values are always much lower than the experimental values. Therefore, the CTE of IPCs is in the scope of the predicted values of two models.

3.4 Thermal conductivity

The measured thermal diffusivity is 66.81 m²/s, specific heat is 0.79 kJ/(m·K), and the TC of Si/Al IPCs is 124.03 W/(m·K). During the process of heat transfer, there is an effect of thermal resistance of interfaces in composites, and the effect of it will be increased with the interfacial area, thus the thermal conductivity will be declined and the decrease degree of it will depend upon the interfacial area. Therefore, if the interfacial area is declined, the thermal conductivity of composites will be increased. LIN's et al[25] study about IPCs shows that the interfacial area of IPCs in unit volume is lower than that of PRMMCs. So the TC of IPCs will be improved.

4 Conclusions

1) The concept of interpenetrating phase composites (IPCs) used for electronic packaging is proposed, which is the development direction of high property materials for electronic packaging.

2) Particles of some particular PRMMCs are connected through the diffusion of particle reinforcement element in matrix alloys. Based on this, a novel fabrication method for interpenetrating phase composites (IPCs) is proposed, and Si/Al IPCs (Si 65%) are produced by squeeze casting and hot press sintering technology.

3) OM microstructure of the composites shows that the reinforcement of composites, no matter on the horizontal section or vertical section of composites, is of a particular three-dimensional continue network structure. There are some small pores in the composites, so relative density of the composites is only 95.41%.

4) The average linear CTE of Si/Al IPCs is $8.27 \times 10^{-6}/\text{K}$ between 20 °C and 100 °C, and the TC is 124.03 W/(m·K), which can greatly meet the demands of electronic packaging.

5) ROM model and TURNER model can be used to predict the CTEs of IPCs. The CTE of the Si/Al IPCs is in the scope of the theoretical prediction values of them.

References

- [1] ZWEBEN C. Advances in composite materials for thermal management in electronic packaging[J]. JOM, 1998, 50(6): 47–51.
- [2] JOHNSTON C, YOUNG R. Advanced thermal management materials[J]. International Newsletter on Microsystems and MEMs, 2000, 2(1): 14–15.
- [3] PREMKUMAR M K, HUNT W H, SAWTELL R R. Aluminum composite materials for multichip modules[J]. JOM, 1992, 44 (7): 24–28.
- [4] LEE H S, JEON K Y, KIM H Y, HONG S H. Fabrication process and thermal properties of SiC_p/Al metal matrix composites for electronic packaging applications[J]. J Mater Sci, 2000, 35(6): 6231–6236.
- [5] COUTURIER R, DUCRET D, MERLE P, DISSON J P, JOUBERT P. Elaboration and characterization of a metal matrix composite: Al/AlN[J]. J Eur Ceram Soc, 1997, 17: 1861–1866.
- [6] CHEN C W. Effects of Si_p size and volume fraction on properties of Al/Si_p composites[J]. Materials Letters, 2002, (52): 334–341.
- [7] SWARTZ E T, POHL R O. Thermal boundary resistance[J]. Reviews of Modern Physics, 1989, 61(3): 605–667.
- [8] HASSELMAN D P H, DONALDSON K Y, GIGER A L. Effect of reinforcement particle size on the thermal conductivity of particulate-silicon carbide-reinforced Al matrix composite[J]. J Am Ceram, 1992, 75(11): 3137–3140.
- [9] NAN C W, BIRRINGER R, DAVAI D R C, GLEITER H. Effective thermal conductivity of particulate composites with interfacial thermal resistance[J]. J Appl Phys, 1997, 81(5): 6692–6699.
- [10] WANG Xiao-feng. Study on fabrication and properties of three-dimensional network structure Si-Al composites[D]. Harbin: Harbin Institute of Technology, 2006.
- [11] SHEN Y L. Thermal expansion of metal-ceramic composites: A three-dimensional analysis[J]. Mater Sci Eng A, 1998, 252: 269–275.
- [12] HAN Jie-cai, HONG Chang-qing, ZHANG Xing-hong, WANG Bao-lin. Thermal shock resistance of TiB₂-Cu interpenetrating phase composites[J]. Compos Sci Technol, 2005, 65: 1711–1718.
- [13] CLARKE D R. Interpenetrating phase composites[J]. J Am Ceram Soc, 1992, 75: 739–759.
- [14] PILLIAR R M, HONG J. Method of manufacture of porous inorganic structures and infiltration with organic polymers. US2004043051[P]. 2004.
- [15] SOKOL G E, POWELL B R. Metal matrix composite and preform therefor[P]. US 5679041, 1997.
- [16] BRESLIN M C, RINGNALDA J, SEEGER J, et al. Alumina/aluminum Co-continuous ceramic composite (C⁴) materials produced by solid/liquid displacement reactions: Processing kinetics and microstructures[J]. Ceram Eng Sci Proc, 1994, 15(4): 104–112.
- [17] HORVITZ D, GOTMAN I, GUTMANAS E Y, CLAUSSEN N. In situ processing of dense Al₂O₃-Ti aluminide interpenetrating phase composites[J]. J Eur Ceram Soc, 2002, 22: 947–954.
- [18] MATTERN A, HUCHLER B, STAUDENECKER D, OBERACKER R, NAGEL A, HOFFMANN M J. Preparation of interpenetrating ceramic-metal composites [J]. J Eur Ceram Soc, 2004, 24: 3399–3408.
- [19] LIU Yu, GONG Xiao-lu. Compressive behavior and energy absorption of metal porous polymer composite with interpenetrating network structure[J]. Trans Nonferrous Met Soc China, 2006, 16: 439–443.
- [20] LIU Yang, HONG J, WANG Jian, PILLIAR R M, SANTERRE J P. Influence of anionic monomer content on the biodegradation and toxicity of polyvinyl-urethane carbonate-ceramic interpenetrating phase composites[J]. Biomaterials, 2005, 26: 5951–5959.
- [21] WINTHER-JENSEN B, WEST K. Interpenetrating networks of two conducting polymers[J]. Synthetic Metals, 2005, 148: 105–109.
- [22] KIM J S, KWON Y S, LOMOVSKY O I, KORCHAGIN M A, MALI V I, DUDINA D V. A synthetic route for metal-ceramic interpenetrating phase composites[J]. Materials Letters, 2006, 60: 3723–3726.
- [23] KONOPKA K, SZAFRAN M. Fabrication of Al₂O₃-Al composites by infiltration method and their characteristic[J]. J Mater Processing Technology, 2006, 175: 266–270.
- [24] TURNER P S. Thermal expansion stresses in reinforced plastics[J]. Journal of Research of the National Bureau of Standards, 1946, 37: 239.
- [25] LIN Jun-pin, ZHANG Yong, CHEN Guo-liang. Microstructures and properties of skeleton reinforced composite[J]. Journal of Materials Engineering, 2006, 6: 20–21. (in Chinese)

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