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Microstructure evolution of Si_3N_4/Si_3N_4 joint brazed with Ag–Cu–Ti + SiCp composite filler

J. Zhang *, Y.M. He, Y. Sun, C.F. Liu

School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, PR China

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Abstract

The Si_3N_4 ceramic was brazed by Ag–Cu–Ti + SiCp composite filler (p = particle) prepared by mechanical mixing. Effects of the content of Ti and SiC particles on microstructure of the joint were investigated. A reliable Si_3N_4/Si_3N_4 joint was achieved by using Ag–Cu–Ti + SiCp composite filler at 1173 K for 10 min. A continuous and compact reaction layer, with a suitable thickness, forms at the Si_3N_4/braze interface. The SiC particles react with Ti in the brazing layers, forming Ti_3SiC_2 thin layers around the SiC particles themselves and Ti_5Si_3 small particles in the Ag[Cu] and Cu[Ag] based solid solution. The higher content of SiC particles in the filler ($\geq$10 vol%) depresses interfacial bonding strength between the Si_3N_4 substrate and composite brazing layer due to the thinner reaction layer and the bad fluidity of the filler. The $Ti_3SiC_2 \rightarrow TiC + Ti_5Si_3$ reaction occurs when Ti concentration around SiC particles in the filler increases.

Keywords: Si_3N_4 ceramic; Brazing; Ag–Cu–Ti + SiCp composite filler; Microstructure

1. Introduction

Silicon nitride is the most attractive nitride ceramic for structural applications. It possesses high hardness, low density, excellent low and high temperature properties and good corrosion resistance. Like most ceramics, silicon nitride is difficult to produce complex or large workpieces, which restricts its wide use. To join small and simple ceramic pieces to form complex-shape and large-size ceramic components becomes necessary [1].

So far, the research on the bonding technologies of ceramics has been well documented, such as brazing, diffusion bonding, partial transient liquid phase bonding [2]. Brazing has become the effective method for joining ceramic due to its convenience and cost-effectiveness [3]. Active metal brazing is a well-established technique for joining ceramics to both to themselves and to metals. The active element consisted in the brazing filler, such as Ti or Zr, is characterized as having sufficient thermodynamic driving force to destabilize the ionic or covalent bonding of the ceramic by reacting with one or more of the ceramic elements to form a reaction layer [4].

Ag–Cu–Ti has been widely adopted as the brazing alloy for joining Si_3N_4 ceramic [5]. This kind of brazing alloy shows good wettability to Si_3N_4 ceramic. However, large residual stress can be caused in the ceramic near the joint interface due to the high thermal expansion coefficient (CTE) mismatches between Si_3N_4 ceramic and brazing alloy [6,7]. In recent year, by means of the addition of a low CTE material (particles or fibers) into the brazing alloy, the CTE mismatch between ceramic substrates and brazing alloy was lowered and the joint strength was increased [8–15]. It was reported [8,9] that the addition of 12 vol% short carbon fibers to 50Ag–46Cu–3Ti–1Sn (at.%) brazing alloy resulted in up to 30% improvement in the shear/tensile joint strength of the stainless steel and alumina joints. Blugan et al. [10] used the SiC-particle-reinforced active brazing alloy to braze silicon nitride ceramic composite to steel successfully and the joint strength was improved with the optimum SiC particles content in the joint at room temperature or elevated temperatures. However, the reaction between SiC particles and active Ti was not clearly clarified. In this investigation, Silicon carbide particles were added to Ag–Cu–Ti brazing alloy for brazing Si_3N_4 ceramic. The bonding mechanism was discussed and the reaction between Ti and SiC

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* Corresponding author. Tel.: +86 451 86414234; fax: +86 451 86414234.
E-mail address: hitzhangjie@hit.edu.cn (J. Zhang).

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particles was clarified firstly. Simultaneously, the effects of the content of Ti and SiC particles on microstructure of the joints were also investigated.

2. Materials and experimental procedures

The Si_3N_4 ceramic used in this investigation was sintered by a hot-pressure process. The bending strength of Si_3N_4 at room temperature is over 700 MPa. The raw Si_3N_4 was sawed by diamond cutting machine into the brazing samples with the size of 3 mm × 4 mm × 2 mm. The brazed surface (3 mm × 4 mm) of the ceramic sample was coarsely ground with SiC abrasive papers and then polished using 1 μm diamond paste.

The active brazing alloy with chemical content of Ag 55.85 at.%, Cu 36.86 at.%, Ti 7.28 at.% was in powder form with an average size of 50 μm. SiC particles with an average diameter of 10 μm were added to Ag–Cu–Ti brazing alloy. The volume fractions of the SiC particles in the composite filler were 0, 5%, 10% and 15% respectively. Additional Ti particles with an average size of 25 μm were employed to change the Ti content in the brazing alloy. The overall atomic fractions of the Ti particles in the composite filler were 7.28%, 10.74%, 14.08% and 17.32% respectively. SiC particles were dispersed with ethanol in an ultrasonic device. Then, Ag–Cu–Ti powders and Ti particles together with a small amount of cellulose nitrate and octylacetate were added to the SiCp/ethanol solution for making a composite brazing paste. Before joining, the Si_3N_4 ceramics were cleaned ultrasonically in acetone and dried by air blowing. The composite brazing paste was placed between two

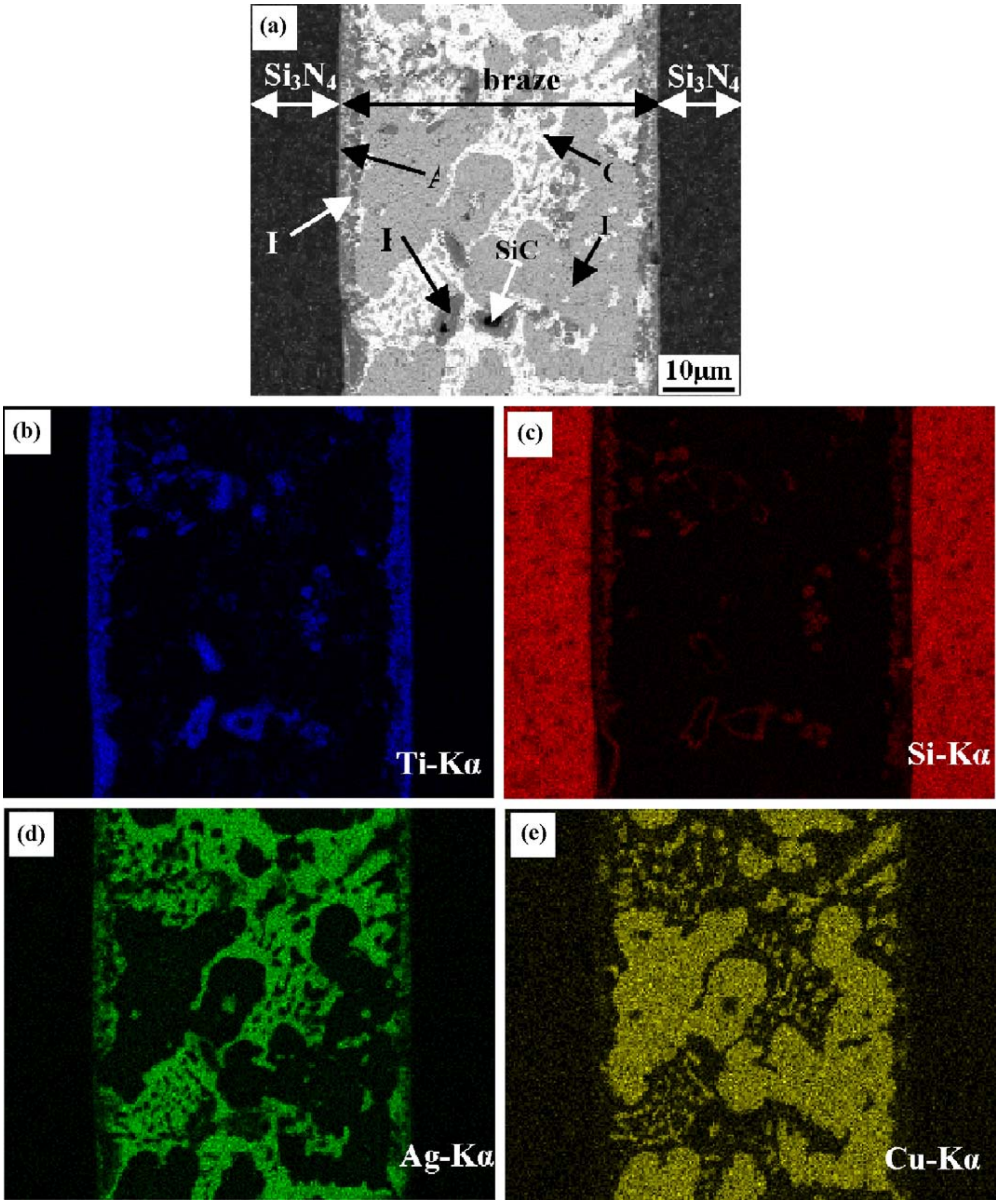

Fig. 1. Morphology and elemental analysis of the Si_3N_4/Si_3N_4 joint brazed using $(Ag_{60.1}Cu_{39.9})_{92.72}Ti_{7.28\ (at.\%)}$ + 5 vol% SiCp composite filler at 1173 K for 10 min.

Table 1
Compositions of the points B, C, D and E shown in Fig. 1.

Point	Composition (at.%)					Phase
	Ag	Cu	Ti	Si	C	
B	1.14	12.62	53.89	32.35	–	Ti_5Si_3
C	81.60	18.40	–	–	–	Ag[Cu]
D	3.18	96.82	–	–	–	Cu[Ag]
E	1.31	10.25	27.38	13.83	47.23	Ti_3SiC_2

Si_3N_4 samples and then the assembly was brazed in a graphite jig.

During the brazing process, a pressure of 0.016 MPa was exerted on the brazing sample to ensure each part contact closely. A vacuum was kept at $(1.3–1.7) \times 10^{-3}$ Pa during brazing. At the beginning of the brazing process, the brazing sample was heated up to 573 K at a rate of 20 K/min and held steady for 10 min to make the organic glue volatilize and keep the brazing surfaces clean. Then the temperature was continuously increased to 1173 K at a rate of 10 K/min and held constant for 10 min. At last, the brazing sample was cooled down at a rate of 5 K/min to 473 K and then cooled freely in the furnace.

To characterize the microstructure, the cross-sections of the brazed joints were cut, polished with 1 μm diamond paste and then examined by means of a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). The different phases were identified using an X-ray diffraction (XRD) method.

3. Results and discussion

3.1. Microstructure of the Si_3N_4/Si_3N_4 joints

Fig. 1 shows the microstructure and the corresponding element area distribution of a Si_3N_4/Si_3N_4 joint brazed with Ag–Cu–Ti + 5vol% SiCp at 1173 K for 10 min. The brazing layer exhibits sound bonding without any defects to the Si_3N_4 ceramic and the SiC particles. The joint consists of three regions: (1) continuous reaction layer A next to ceramic; (2) discontinuous reaction layer B adjacent the continuous reaction layer; and (3) solid solution in the middle of the joint where some reaction phases exist.

Fig. 1(b) shows a continuous Ti layer close to the Si_3N_4 ceramic, indicating that Ti, as an active element, has diffused toward the Si_3N_4 ceramic. According to the free energy of formation of TiN and Si_3N_4 [16], Ti has reacted with Si_3N_4 to form TiN and Si. As indicated in Fig. 1(c), the continuous

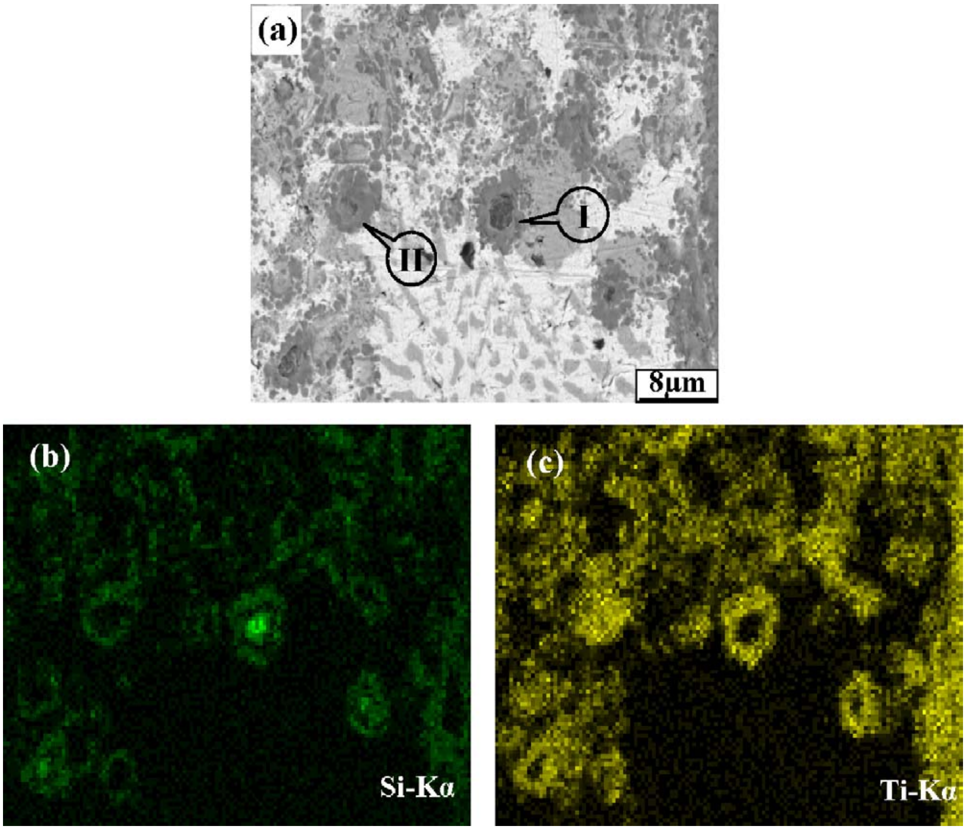

Fig. 2. Cross-sectional view (a) of a Si_3N_4/Si_3N_4 joint brazed with $(Ag_{60.1}Cu_{39.9})_{92.72}Ti_{7.28\ (at.\%)}$ + 5 vol% SiCp and the Si Kα (b) and Ti Kα (c) map showing the Ti and Si distributions in (a).

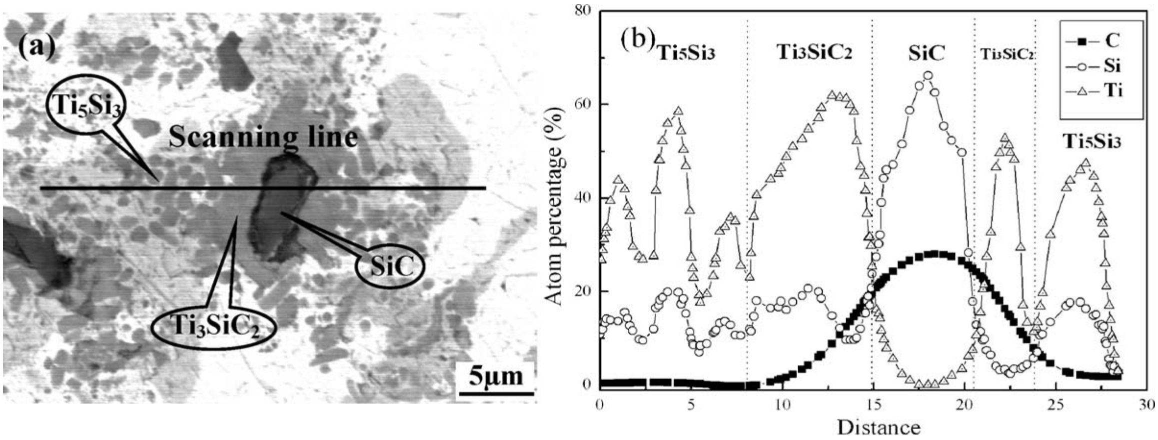

Fig. 3. Morphology of the joint (a) and elemental distributions (b) of C, Si, and Ti elements along the black line in (a).

reaction layer did not contain Si, implying that the Si released from the reaction has diffused towards the liquid brazing alloy. Together with the higher concentration of Ti near the TiN reaction layer, a discontinuous reaction layer which had few distances from the continuous TiN layer, about 2–3 μm thick, was formed as demonstrated in Fig. 1(b and c). According to composition analysis provided in Table 1, the discontinuous reaction layer which was made up of 32.35 at.% Si, 53.89 at.% Ti and a definite amount of Ag and Cu should be Ti_5Si_3 phase. The central part of the joint was composed of bright Ag based solid solution (marked by C), grey Cu based solid solution (marked by D) and SiC particles together with reaction phases of Ti and SiC particles around SiC particles. When Ag–Cu–Ti brazing alloy melts, Ti will cluster beside the SiC particles and react with them. So it is necessary to investigate the reaction between SiC particles and active Ti element.

Different reaction phases between Ti and SiC particles are shown in Fig. 2: one is marked by arrow I, which will be elucidated in Fig. 3; the other is indicated by arrow II. The central part of area II is rich in Ti and lack of Si, as demonstrated in Fig. 2(b and c). According to chemical composition analysis, this area which is made up of 43.92 at.% C, 49.69 at.% Ti can be determined to be TiC. There is discontinuous Ti and Si distributing surrounding TiC, as displayed in Fig. 2(b and c). Associating with the X-ray diffraction results presented in Fig. 4, these disperse phases are Ti_5Si_3.

Fig. 3 presents magnified back-scattered micrograph of SiC particle in the seam (marked by arrow I in Fig. 2) and corresponding line scans. The dark part in the center of the scanning line, which is rich in C and Si, can be determined to be unreacted SiC. There is a continuous reaction layer around the remnant SiC particle. Combining with the Ti–Si–C ternary phase diagram [17] and the elemental distributions results (Fig. 3(b)), the continuous reaction layer which is made up of 51.66 at.% C, 27 at.% Si, 18.96 at.% Ti and a small quantity of Ag and Cu is namely Ti_3SiC_2 (the content of C could not be detected precisely because it is light element). Furthermore, some disperse reaction phases beside Ti_3SiC_2 can also be found in Fig. 3(a), where the concentration of Ti and Si fluctuated. Together with the XRD results shown in Fig. 4, these disperse phases are also Ti_5Si_3.

3.2. Effect of SiC particles content on microstructure of the brazed joints

Fig. 5 shows scanning electron micrographs of the cross-sections of Si_3N_4/Si_3N_4 joints for different content of SiC particles ((a) 0 vol%, (b) 5 vol%, (c) 10 vol% and (d) 15 vol%) in the brazing alloy treated at 1173 K for 10 min. Different kinds of reaction phases in the seam are highlighted in Fig. 5. As can be seen from the pictures, the black SiC particles spread fairly randomly throughout the brazing alloy of the performed joint. Meanwhile, Ag and Cu based solid solution became more uniform and fine when SiC particles content in the filler was up to 15 vol%. In terms of crystallographic theory, new phase in the melt usually prefers to precipitate around impurity. Thus, the undissolved SiC particles during brazing cycle can act as nucleation site for Ag and Cu based solid solution. The higher amount of SiC particles in the filler is, the more nucleation

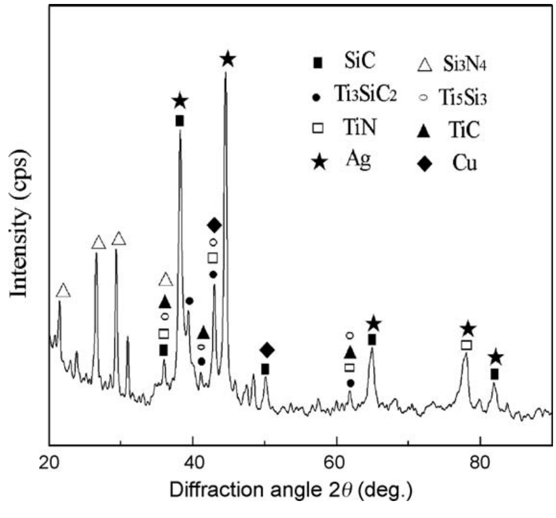

Fig. 4. XRD pattern of the brazing layer in the joint brazed with $(Ag_{60.1}Cu_{39.9})_{92.72}Ti_{7.28\ (at.\%)}$ + 5 vol% SiCp composite filler at 1173 K for 10 min.

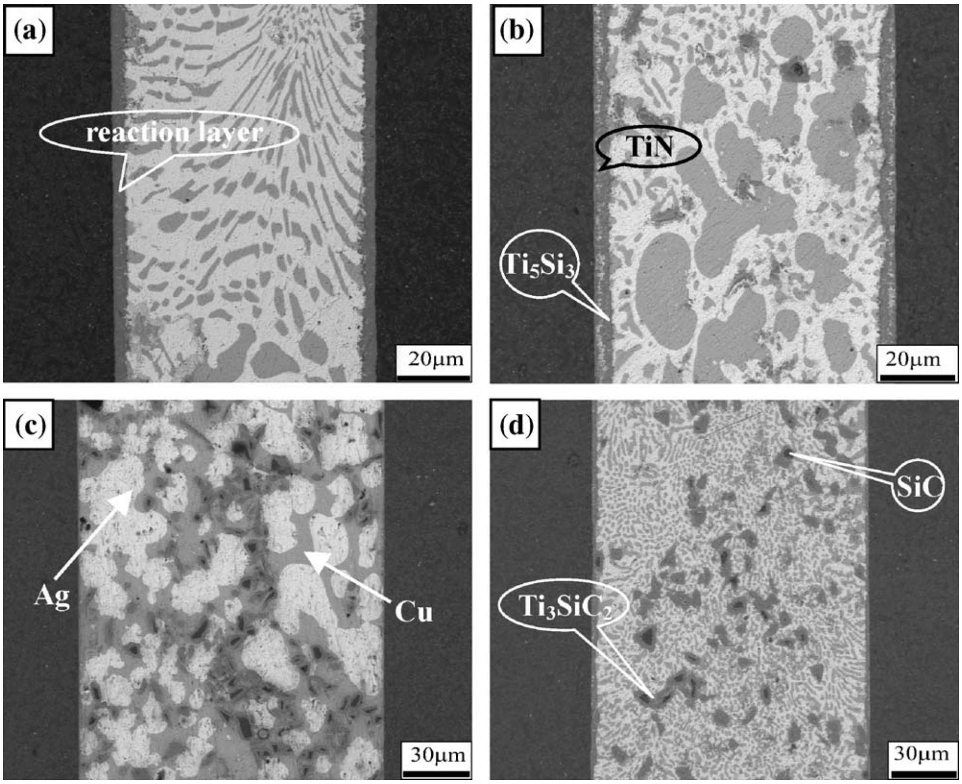

Fig. 5. SEM images of the cross-sections of the Si_3N_4/Si_3N_4 joints brazed using composite filler with increasing SiC particles contents: (a) 0 vol%, (b) 5 vol%, (c) 10 vol% and (d) 15 vol% at 1173 K for 10 min.

center will be supplied for Ag and Cu based solid solution, being beneficial for the uniformity and refinement of those two solid solutions.

Another remarkable phenomenon is that the thickness of Ti_3SiC_2 reaction layer around SiC particles in the seam became thinner with rising SiC particles content in the filler as compared with Fig. 5(b–d). All the surfaces of SiC particles contained in the filler will participate to the reaction. It can be inferred that the higher content of the SiC particles in the brazing layer is, the lower the concentration of Ti element around single SiC particle will be. Thus, excessive SiC particles will necessarily bring about insufficient reaction between the Ti element and the SiC particles, inducing a thin reaction layer around SiC particles.

Comparing with the interfacial structure brazed with Ag–Cu–Ti brazing alloy as shown in Fig. 5(a), the structure of Si_3N_4/solder interfacial reaction layer brazed with the Ag–Cu–Ti + SiCp composite filler did not appear variably, whereas Ti_5Si_3 reaction layer adjacent TiN could not be detected clearly when the SiC particles content in the joint was up to 10 vol% or higher. Furthermore, the total thickness of reaction layer at the interface was found to decrease with increasing SiC particles content in the filler, as confirmed in Table 2. Especially, continuous reaction layer could not be formed at local area while the SiC particles content in the joint was up to 10 vol% or higher. This could be attributed to insufficient Ti concentration at the interface due to trapping of Ti by the excessive SiC particles present in the joint. Actually, the addition of SiC particles resulted in a large increase in the total surface area available to react with Ti, depressing the amount of Ti diffusing towards Si_3N_4 ceramics.

3.3. Effect of Ti content on microstructure of the brazed joints

According to the Ag–Cu binary phase diagram [18], the Ag–Cu eutectic temperature is 1053 K (the eutectic composition of Ag–Cu is $Ag_{60.1}Cu_{39.9}$ (at.%)). During the heating process, Ag–Cu brazing alloy melts at 1053 K. Then, Ti begins to be dissolved in the liquid. When the temperature reaches 1123 K, the entire Ag–Cu–Ti is in liquid state. Ti, as an active element, will diffuse towards the Si_3N_4 ceramics and reacts with them to form TiN at the Si_3N_4/braze alloy interface by the following

Table 2
Effect of SiC particles content in the composite filler on the reaction layer thickness.

		SiCp content (vol%)			
		0	5	10	15
Average reaction layer thickness (μm)	Ti_5Si_3 + TiN	4.7 ± 0.5	4.25 ± 0.5	1.5 ± 0.5	2.25 ± 0.5

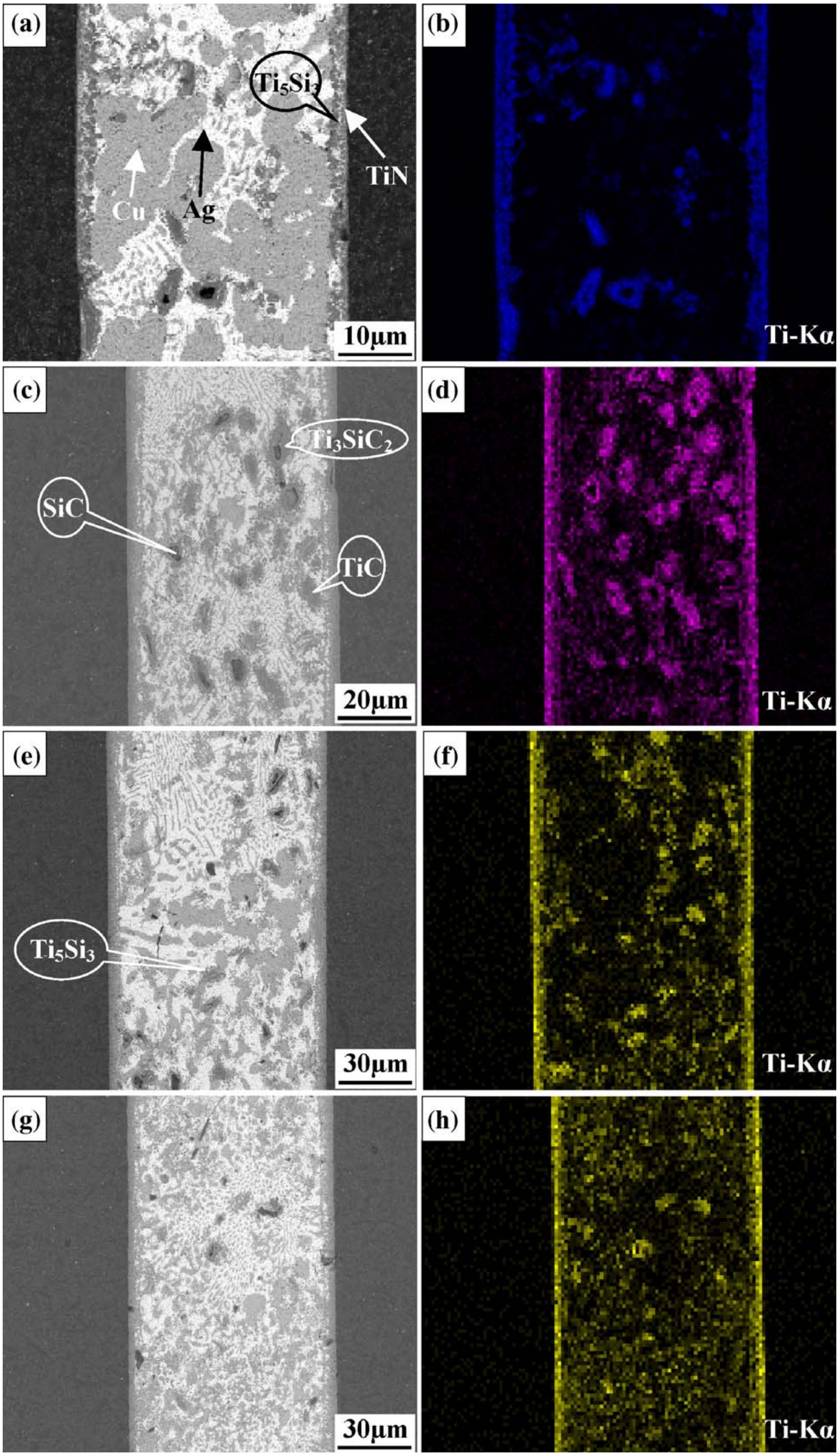

Fig. 6. SEM micrographs and Ti Kα maps of Si_3N_4/Si_3N_4 joints with increasing Ti contents: (a and e), 7.28 at.%, (b and f) 10.74 at.%, (c and g) 14.08 at.% and (d and h) 17.32 at.% in the brazing material.

Table 3
Effect of Ti content in the composite filler on the reaction layer thickness.

		Ti content (at.%)			
		7.28	10.74	14.08	17.32
Average reaction layer thicknes (μm)	TiN	0.5 ± 0.5	2.8 ± 0.5	2.8 ± 0.5	2.8 ± 0.5
	Ti_5Si_3 + TiN	3.7 ± 0.5	4.9 ± 0.5	5.6 ± 0.5	6.8 ± 0.5

reaction [19–21]:

$$Si_3N_4(s) + 4Ti(l) = 4TiN(s) + 3Si(s) \quad (1)$$

$$\Delta G_f^0(Si_3N_4)(kJ/mol) = -361.9 + 0.1575T$$

$$\Delta G_f^0(TiN)(kJ/mol) = -672.6 + 0.1865T$$

$$\Delta G_f^0(kJ/mol) = -1356 + 0.199T$$

It can be found that the free energy of formation for Si_3N_4 and TiN are both negative at the brazing temperature ranging from 1053 to 1173 K. Moreover, the free energy for TiN is lower than that for Si_3N_4, indicating that TiN is more stable at the brazing temperature. Simultaneously, the free energy of the reaction (1) is also negative during the brazing process; so, definite TiN reaction layer is formed. At the same time, Si is released from the decomposition of the Si_3N_4 ceramic and diffuses into the molten braze. Ti_5Si_3 reaction phase occurs according to the following reaction [19–21]:

$$5Ti(s) + 3Si(s) = Ti_5Si_3(s) \quad (2)$$

$$\Delta G_f^0(kJ/mol) = -194.14 + 0.0167T$$

Thermodynamic calculations from the free energy of formation of Ti_5Si_3 confirm its stability. Consequently, the Si_3N_4 ceramic/solder interface is consisted of an inner TiN layer near the ceramic and an outer Ti_5Si_3 layer adjacent the Ag–Cu–Ti + SiCp composite filler.

SiC particles contained in the composite filler are in solid state during the brazing cycle, when Ti is dissolved in the liquid, it also diffuses towards SiC particles in the molten braze and reacts with them, so it is necessary to clarify the reaction between Ti and SiC particles. According to relevant literatures [22–27], when Ti contacts SiC, Ti_3SiC_2 and Ti_5Si_3 are firstly formed by the following reactions:

$$14Ti + 6SiC = 3Ti_3SiC_2 + Ti_5Si_3 \quad (3)$$

Some researchers [28,29] reported Ti_5Si_3C may occur during reaction between Ti and SiC. What is more, the free energy of formation for Ti_5Si_3C (−1140.78 kJ/mol) is far more negative than Ti_3SiC_2 (−735.78 kJ/mol) at the brazing temperature. Actually, Ti_5Si_3C was not detected in the joint according to X-ray diffraction results (shown in Fig. 4).

When Ti content is higher around SiC particles in the composite filler or other elements, such as Zr, are introduced to enhance the activity of Ti. Ti_3SiC_2 starts to decompose according to:

$$Ti_3SiC_2 \rightarrow TiC_{1-x} + Ti_5Si_3 \quad (4)$$

In this condition, Ti_3SiC_2 decomposes continuously and breaks up the equilibrium of Eq. (3), accelerating the reaction of SiC particles with Ti until SiC particles deplete. TiC_{1-x} were determined to be TiC [30–34].

Considering the composite filler containing SiC particles, which will react with Ti element during brazing and then decrease amount of Ti diffusing towards Si_3N_4 ceramic, thus, a thin Si_3N_4/solder interfacial reaction layer will generally be formed. Too thin interfacial reaction layer is adverse to the joint quality. So, a definite amount of Ti will be supplied to the filler for compensating Ti consumed by SiC particles. Fig. 6 provides the typical morphology and the corresponding Ti distribution in the observed area for the Si_3N_4/Si_3N_4 joints brazed with $(Ag_{60.1}Cu_{39.9})_{100-x}Ti_{x\ (at.\%)}$ + 5 vol% SiCp composite filler at 1173 K for 10 min. Integrated joints could be achieved with diverse Ti content in the composite filler.

As shown in Fig. 6, when Ti content in the filler is 7.28 at.% or 10.74 at.%, Ti mainly distributes near Si_3N_4 ceramic and around single SiC particle (which can be seen from the Ti map of Fig. 6(e or f)), indicating Ti_3SiC_2 surrounding SiC particles can not be decomposed completely due to insufficient Ti. As Ti content increases to 14.08 at.% or higher, Ti distribution is more uniform in the seam as displayed in Fig. 6(g or h). Combining with the above analysis of reaction between Ti and SiC, it was attributed to the presence of TiC and Ti_5Si_3 deriving from the decomposition of the Ti_3SiC_2 surrounding the SiC particles and dispersed in the seam homogeneously.

Table 3 demonstrates the effect of Ti content in the composite filler on the reaction layer thickness. The total reaction layer (Ti_5Si_3 + TiN) thickness increased with Ti content rising. However, the thickness of TiN reaction layer close to Si_3N_4 ceramic presents diverse trend. It increases from 0.5 to 2.8 μm as Ti content varies from 7.28 to 10.74 at.% and remains stable subsequently. When Ti content in the filler rose, more Ti would diffuse towards Si_3N_4 ceramic, favoring the thicker TiN reaction layer formed. However, when definite TiN reaction layer was formed, it became a barrier-layer of further reaction between Ti and Si_3N_4. In other words, further reaction between Ti and Si_3N_4 must get across TiN reaction layer which has been formed. Therefore, the growth rate of the TiN reaction layer was controlled by the diffusion rate of Ti atom in the TiN layer, resulting in the thickness of TiN reaction layer varied less as Ti content increased continuously. Practically, the higher Ti content in the filler is, the more it concentrates near Si_3N_4 ceramic. The high concentration of Ti beside Si_3N_4 which can not get across previous formed TiN layer, would react with Si that released from Si_3N_4, due to the previous reaction, leading to the increase of the total thickness of the reaction layer.

4. Conclusions

The Si_3N_4 ceramic was successfully brazed to itself using Ag–Cu–Ti + SiCp composite filler at 1173 K for 10 min. The following conclusions can be drawn.

The joint is comprised by three regions: a continuous TiN reaction layer close to Si_3N_4 ceramic, a discontinuous Ti_5Si_3 reaction layer connecting the continuous reaction layer, Ag and Cu based solid solution and SiC particles in the middle of the joint together with the reaction phases of Ti and SiC particles around SiC particles.

The reaction between Ti and SiC particles carries out step by step. At the beginning, Ti reacts with SiC to form Ti_3SiC_2 and Ti_5Si_3, then Ti_3SiC_2 starts to decompose into TiC and Ti_5Si_3 when Ti concentration around SiC particles increases in the composite filler.

With increasing SiC particles content in the filler continuously, the microstructure of the joints revealed that Ag and Cu based solution became more uniform and fine and the degree of reaction between both the Si_3N_4 ceramic substrate and the SiC particles in the seam and Ti element declined.

With increasing Ti content in the composite filler continuously, SiC particles reacted completely after brazing cycle, the ultimate reaction products, such as TiC, Ti_5Si_3, dispersed in the brazing seam homogeneously.

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