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Brazing Si_3N_4 ceramic using a Cu–Pd–Ti filler alloy for high-temperature applications

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Si_3N_4 ceramic is an attractive material for engineering applications because of its low density, higher thermal resistance, and excellent wear resistance without lubricants. It has been proposed for manufacturing the rotor of jet turbines. However, it is difficult to manufacture the Si_3N_4 ceramic workpieces with large dimensions and complicated shapes due to its poor workability and low ductility. Therefore, the engineering application of Si_3N_4 ceramic greatly depends on the development of bonding techniques for the ceramic. In recent 20 years, many studies have been focused on joining techniques for Si_3N_4 ceramic.

Active metal brazing has been widely investigated and used for joining the Si_3N_4 ceramic because it is a simple process to obtain high-strength ceramic joints in different shape and size components [1–3]. For active brazing of Si_3N_4 ceramic, filler alloys containing some active metals such as Ti, Zr, Hf, etc., are usually used, and a high-strength ceramic joint can be obtained by the reaction between the active metals and the Si_3N_4 ceramic. Cu–Ag-based alloys containing various amount of Ti have been widely adopted as the filler alloy for the brazing of Si_3N_4 ceramic [4, 5]. These filler alloys have a good wettability to almost all the ceramics. However, the ceramic joints using these filler alloys have low thermal and oxidation resistance, and cannot be used at temperatures above 700 K. Therefore the Cu–Ag–Ti filler alloys are not suitable for brazing the Si_3N_4 ceramic for use in high-temperature environment.

To improve the thermal and oxidation resistance of the Si_3N_4 ceramic joint, it is necessary to develop new filler alloys with higher melting point and oxidation-resistance. It was reported that the Si_3N_4/Si_3N_4 joints with high thermal and oxidation resistance have been obtained using active filler alloys, which were developed by adding metals such as Au, Pd, Pt, V, Co, etc., into the filler alloys [6–10]. In this investigation, a Cu–Pd–Ti alloy was used as the filler alloy for brazing an Si_3N_4 ceramic. The effect of brazing parameters on mechanical properties of the Si_3N_4/Si_3N_4 joint was measured and analyzed.

The Si_3N_4 ceramic used in this investigation was made by a pressure sintering process. The content of MgO and Al_2O_3 in the ceramic was less than 5 wt%. The raw materials of the filler alloy were Cu, Ti, and Pd foils with the thickness of 10, 20, and 10 μm, respectively. By adjusting the amount of the three kinds of foils, an alloy with a composition of 76.5Cu–8.5Pd–15Ti (at.%) was formed during brazing.

The Si_3N_4 ceramic sample with a size of $\Phi 6 \times 4$ mm was ground to a surface finish of $R_a = 30\ \mu$m, and then was cleaned together with the metal foils in a supersonic device. The cleaned metal foils were placed between two pieces of Si_3N_4 ceramic samples and a small weight was put on the upper ceramic sample to give a pressure of 2×10^{-3} MPa. The brazing of the Si_3N_4 ceramic was carried out in a vacuum of 1.33×10^{-3} Pa at 1373 or 1473 K for times up to 5.4 ks. The microstructure of the Si_3N_4/Si_3N_4 joint was observed by scanning electron microscopy (SEM) and electron probe microanalysis (EPMA), and the shear strength of the joint was measured at room temperature.

Fig. 1 shows the morphology and elemental analysis results of the Si_3N_4/Si_3N_4 joint brazed at 1373 K. It can be seen that a continuous TiN reaction layer with an average thickness of 5 μm exists between the Si_3N_4 ceramic and the filler alloy as shown in Fig. 1d and f. The matrix in the central of the joint is Cu base solution containing 5 at.% Pd. There are two phases in the Cu–Pd solution as shown in Fig. 1a. It was determined by the elemental analysis and X-ray diffraction results that the white blocky one is PdTiSi phase as indicated from the results shown in Fig. 1c, d, and e, and the small black one is TiN phase as indicated from the results shown in Fig. 1d and f.

Fig. 2a shows the morphology of the joint brazed at 1473 K. Compared with Fig. 1a obtained at lower brazing temperature (1373 K), a new phase appears in the Cu–Pd solution when the brazing temperature is higher (1473 K). Elemental analysis results indicated that the new phase is Pd_2Si. From the results of Fig. 1 and Fig. 2a, the microstructure of the Si_3N_4/Si_3N_4 joint brazed using the 76.5Cu–8.5Pd–15Ti filler alloy with the brazing conditions used here is schematically shown in Fig. 2b. It shows that the microstructure of the joint is "Si_3N_4 ceramic/TiN reaction layer/filler alloy." The base of the filler alloy is Cu–Pd solid solution, in which there are TiN, Pd_2Si, and PdTiSi phases.

Fig. 3 shows the effect of brazing temperature on the shear strength of the Si_3N_4/Si_3N_4 joint using a filler

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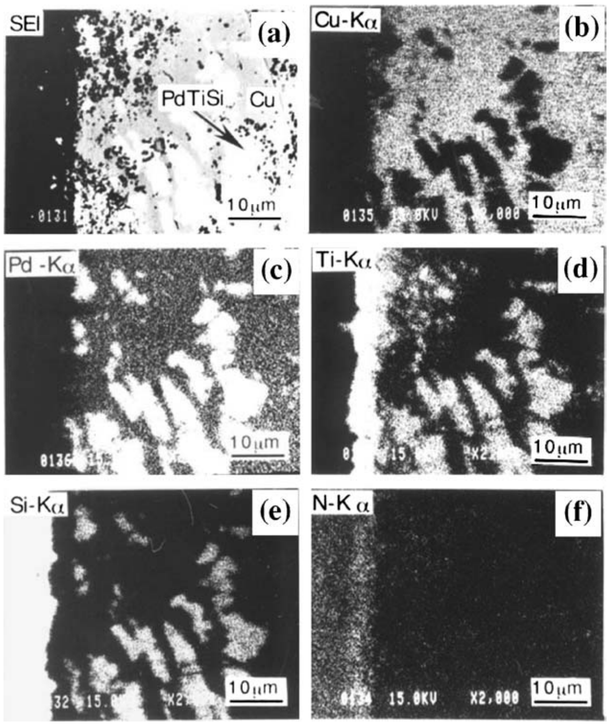

Figure 1 Morphology and elemental analysis results of the Si_3N_4/Si_3N_4 joint brazed using 76.5Cu–8.5Pd–15Ti filler alloy at 1373 K for 1.8 ks.

alloy of 76.5Cu–8.5Pd–15Ti and brazed for a constant time of 1.8 ks. With increasing brazing temperature from 1371 to 1473 K, the shear strength of the joint decreases from 160 to 70 MPa. Tilimann *et al.* [7] have pointed out that the decomposition of the Si_3N_4 ceramic at 1473 K leads to the appearance of voids in the Si_3N_4 ceramic, resulting in a decrease of the joint properties. It has been also found that if Si_3N_4 ceramic is coated by a layer of AgCuTi alloy in a vacuum, the decomposition of the Si_3N_4 ceramic can be avoided during high-temperature brazing, and the wettability between the filler alloy and the Si_3N_4 ceramic can be improved [7].

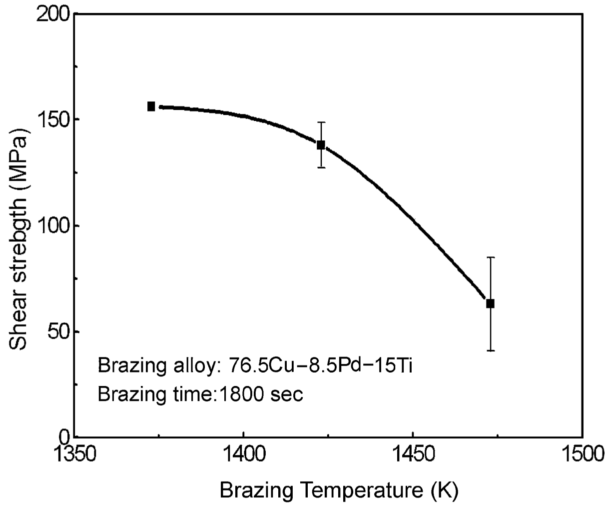

Figure 3 Effect of brazing temperature on the shear strength of the Si_3N_4/Si_3N_4 joint brazed using 76.5Cu–8.5Pd–15Ti for 1.8 ks.

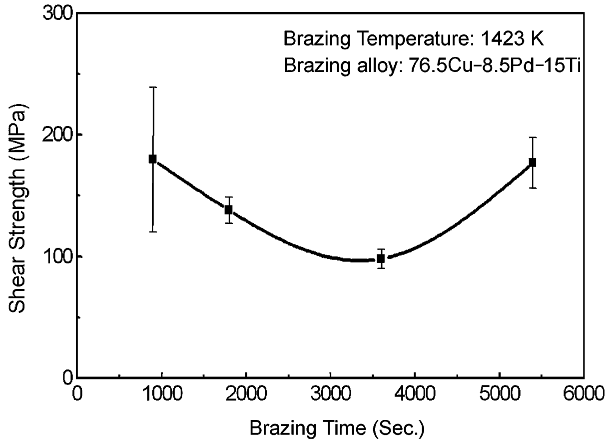

Figure 4 Effect of brazing time on the shear strength of the Si_3N_4/Si_3N_4 joint brazed using 76.5Cu–8.5Pd–15Ti at 1423 K.

Fig. 4 shows the effect of brazing time on the shear strength of the Si_3N_4/Si_3N_4 joint at 1423 K using a filler alloy of 76.5Cu–8.5Pd–15Ti. When the brazing time is 0.9 ks, the average shear strength of the joint is 180 MPa. With increasing brazing time the shear strength of the joint decreases, and when the brazing time is 3.6 ks, it has the lowest value of 100 MPa. With further increasing brazing time, the shear strength of the joint increases, and when the brazing time is 5.4 ks, it reaches 198 MPa. At the beginning of the brazing process, the Si released from the interfacial reaction between Ti and Si_3N_4 ceramic diffuses into the filler alloy and reacts with Ti and Pd forming the PdTiSi phase, which contributes to the bonding strength of the Si_3N_4/Si_3N_4 joint. With increasing brazing time, more and more Ti in the filler alloy reacts with the Si_3N_4 ceramic, leading to a decrease of Ti content in the filler alloy and in turn to the amount of the PdTiSi phase. This is the main reason of the decreasing shear strength with

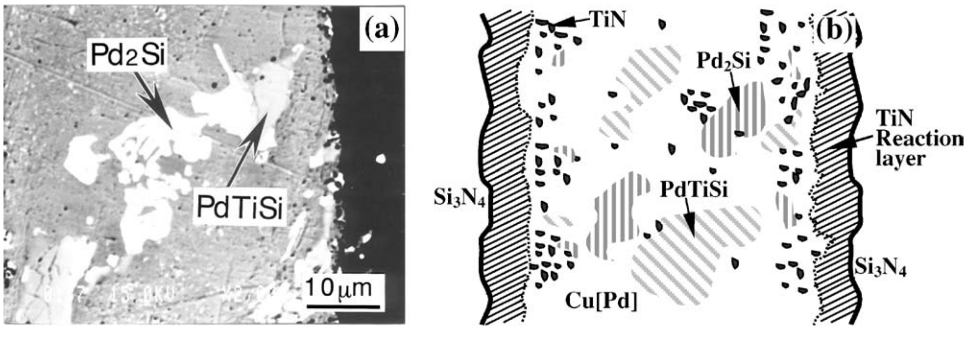

Figure 2 Morphology and schematic drawing of the Si_3N_4/Si_3N_4 joint brazed using 76.5Cu–8.5Pd–15Ti filler alloy at 1473 K for 1.8 ks: (a) morphology and (b) schematic drawing.

increasing brazing time from 0.9 to 3.6 ks as shown in Fig. 4. With further increasing brazing time, because of the decreasing amount of Ti in the filler alloy, Si will react with Pd to form Pd_2Si phase in the filler alloy. With increasing brazing time, both amount and size of the Pd_2Si phase increase, leading to an improvement in the shear strength of the joint. The existence of both PdTiSi and Pd_2Si phases in the filler alloy contributes to the shear strength of the joint. Therefore, it is concluded that the larger amount of PdTiSi or Pd_2Si phase in the filler alloy accounts for the higher shear strength of the Si_3N_4/Si_3N_4 joint when the brazing time is either short (0.9 ks) or long (5.4 ks).

An Si_3N_4 ceramic was brazed using a filler alloy of 76.5Cu–8.5Pd–15Ti at 1373 or 1473 K and under a pressure of 2×10^{-3} MPa for up to 5.4 ks. The results indicate that for a given brazing time of 1.8 ks the shear strength of the joint decreases with increasing brazing temperature because of the decomposition of the Si_3N_4 ceramic at higher temperature. When the brazing temperature is 1423 K, the shear strength of the joint decreases from 180 to 100 MPa with increasing brazing time of 0.9 to 3.6 ks, and then with further increasing brazing time, the shear strength of the joint increases. The shear strength of the Si_3N_4/Si_3N_4 joint reaches a maximum value of 198 MPa when the brazing temperature and holding time is 1423 K and 5.4 ks, respectively.

Acknowledgments

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