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Interfacial characterization of silicon nitride/silicon nitride joints brazed using Cu-base active metal interlayers

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Abstract

Silicon nitride/silicon nitride joints were vacuum brazed at 1317 K for 5 min and 30 min using ductile Cu-base active metal interlayers. The joints were characterized using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), electron back scattered diffraction (EBSD), and transmission electron microscopy (TEM). An inhomogeneous Ti-rich reaction layer (~2–3 μm thick) formed in 5 min at the Si_3N_4/braze interface. The inhomogeneity disappeared after brazing for 30 min and was replaced with a compact and featureless reaction zone. TEM studies revealed fine grains in the reaction layer, and larger grains in the inner part of the joint interfaces. The joints were crack-free and presented features associated with plastic deformation, which indicated accommodation of strain associated with CTE mismatch. Electron Backscatter diffraction (EBSD) revealed a highly textured braze alloy interlayer and its crystallographic orientation was determined. The formation of additional phases at the joint interface during brazing is discussed.

Keywords: A. Joining; B. Electron microscopy; B. Interfaces; D. Si_3N_4; Active metal interlayer

1. Introduction

Silicon nitride ceramics possess excellent high-temperature strength and creep resistance. They are projected to significantly raise engine efficiency and performance when used as turbine components in the next-generation turbo-shaft engines without the extensive cooling that is needed for metallic parts. In the last few decades, considerable amount of research has been done on joining of silicon nitride to itself and to metals. One key aspect of Si_3N_4 utilization in such applications is its response to joining via brazing which is the most widely used method to join ceramics [1–5].

As with brazing of most ceramics, promoting wetting through reactions at the interface between Si_3N_4 and braze improves the adhesion. A wide variety of braze compositions containing active metals have been evaluated including Ni–Cr, Fe–Cr, Ni–Cr–Si, Ni–Cr–B, Au–Pd–Ti, Pd–Ni–Ti and Cu–Pt–Ti/Nb alloys. Besides chromium and titanium as active metals, vanadium (e.g., in Ni–Au–V fillers) has been used to braze Si_3N_4 ceramics and Pd/CuTi interlayers have been used for partial liquid phase bonding.

Among the active metal interlayers (braze filler metals) that have been used to join Si_3N_4 ceramics [2,6–10], Ag–Cu eutectic alloys containing Ti have been most widely used. Contact angle measurements [11,12] reveal that Ti-containing Cu and Ag-base brazes rapidly wet Si_3N_4 ceramics, and the most significant gains are achieved at small (2–10%) amounts of Ti at which braze ductility is not impaired but there is sufficient Ti activity for reaction and bond formation with Si_3N_4. Besides Ti, active elements such as Hf, Zr, Nb and Ta also have been used in braze alloys to join Si_3N_4 to itself and to metals [13–16].

The Ag–Cu–Ti fillers ($T_L < 1200$ K) are known to produce the highest levels of joint integrity in Si_3N_4 ceramics and are some of the most widely used brazes to join Si_3N_4. However, these alloys have relatively low temperature (<800 K) capability, and low thermal and oxidation resistances [9]. There is thus considerable interest in evaluating higher

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temperature brazes with better oxidation resistance. In this study, an active braze alloy based on Cu–Al–Si–Ti system, Cu-ABA, with liquidus temperature ($T_L \sim 1297$ K) and oxidation resistance superior to Ag–Cu–Ti brazes was used to join Si_3N_4. In an earlier work [17], we had compared the oxidation kinetics of Cu-ABA with two other Ti-active brazes, Ticusil and Ticuni, using thermo-gravimetric analysis (TGA) at 1023–1123 K. We had found that Cu-ABA had the smallest weight gain and most sluggish oxidation kinetics; it exhibited a marginal saturation weight gain (<0.05 mg cm^{-2}) in 200 min. Cu-ABA has large Cu content (92.8%) which protects it against oxidation (the Pilling–Bedworth ratios of Cu_2O and CuO are <2). The alloying elements Si and Al in Cu-ABA also form protective scales; the PB ratios of Si and Al are 2.27 and 1.28, respectively (Si is known to exhibit an anomalous behavior in that even though its PB ratio >2, it forms a protective oxide scale). Thus, Si_3N_4 joints made using Cu-ABA are expected to provide higher use temperatures and better oxidation resistance than the widely used Ag–Cu–Ti brazes. The Si_3N_4/Si_3N_4 joints made using Cu-ABA were examined for microstructure and composition using scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDS), electron backscattered diffraction (EBSD) and transmission electron microscopy (TEM).

2. Materials and methods

Kyocera Si_3N_4 (SN-281) was used in the joining work. The material contains about 9–10 wt% Lu_2O_3 and has an acicular, interlocking grain structure and fine distribution of intergranular amorphous phases [18]. A commercial Cu–Si–Al–Ti braze alloy, Cu-ABA, with a nominal composition of 92.75Cu–3Si–2Al–2.25Ti, and the solidus and liquidus temperatures of $T_S \sim 1231$ K and $T_L \sim 1297$ K, respectively, was obtained as a foil (thickness $\sim$50 μm) from Morgan Advanced Ceramics, Hayward, CA. The Cu-ABA braze is ductile (42% elongation) and the elastic modulus, yield strength and tensile strength of the braze are 96 GPa, 278 MPa, and 520 MPa, respectively, and its coefficient of thermal expansion (CTE) is 19.5×10^{-6}/K.

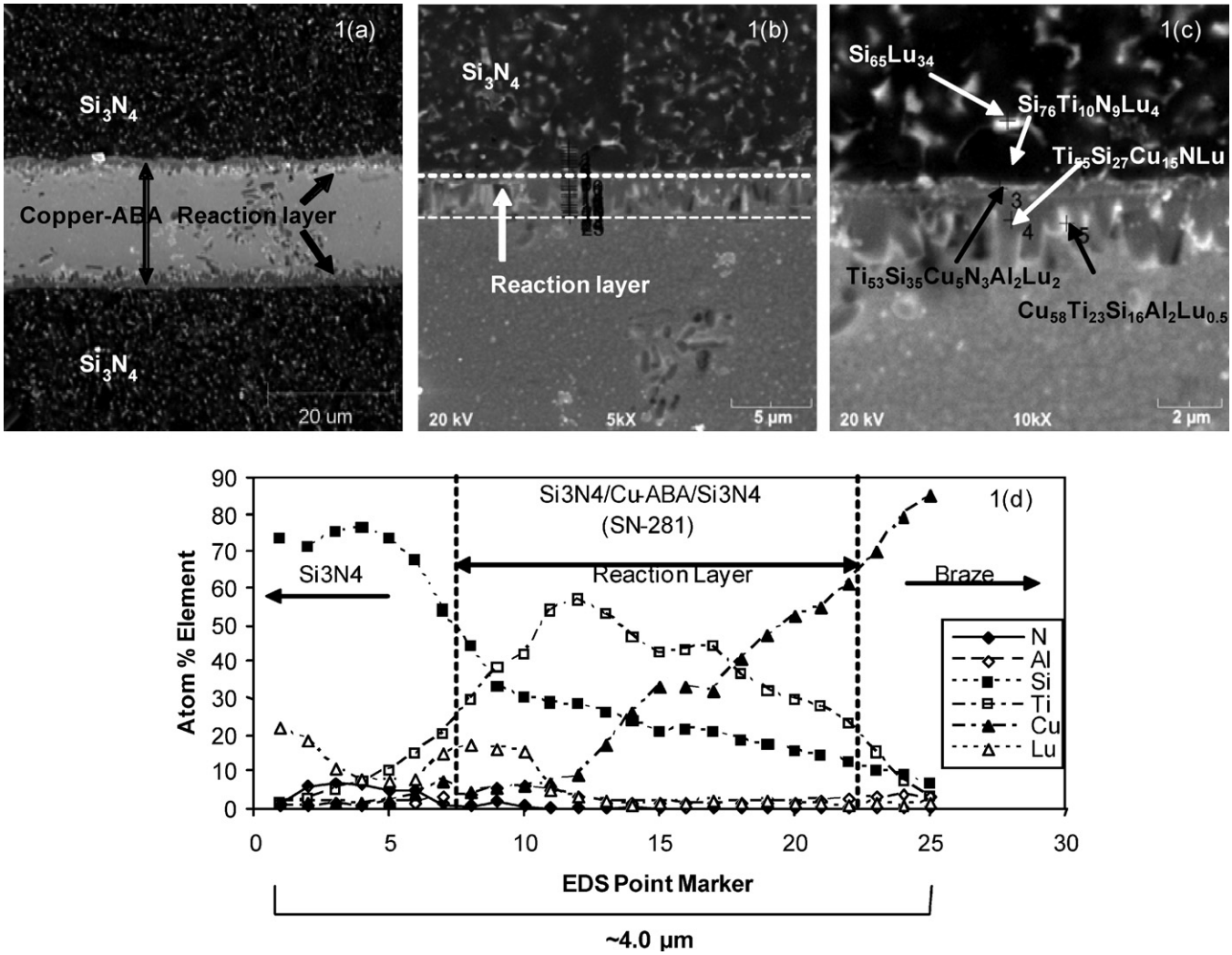

Fig. 1. (a)–(c) Joint microstructure and (d) elemental distribution in self-joined Kyocera SN-281 Si_3N_4 material brazed at 1317 K for 5 min. The EDS data in (d) correspond to the point markers shown in (b).