



Microstructural evolution and characterisation of interfacial phases in Al₂O₃/Ag–Cu–Ti/Al₂O₃ braze joints



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ABSTRACT

Alumina ceramics with different levels of purity have been joined to themselves using an active braze alloy (ABA) Ag–35.3Cu–1.8Ti wt.% and brazing cycles that peak at temperatures between 815 °C and 875 °C for 2 to 300 min. The microstructures of the joints have been studied using scanning electron microscopy, transmission electron microscopy, and energy dispersive X-ray spectroscopy. A limited number of joints prepared with the ABA Ag–26.7Cu–4.5Ti wt.% have also been studied. In terms of characterising the interfacial phases, efforts were made to understand the interfacial reactions, and to determine the influence of various brazing parameters, such as the peak temperature (T_p) and time at T_p (τ), on the microstructure. In addition, the extent to which impurities in the alumina affect the interfacial microstructure has been determined.

Ti₃Cu₃O has been identified as the main product of the reactions at the ABA/alumina interfaces. At the shortest joining time used, this phase was observed in the form of a micron-size continuous layer in contact with the ABA, alongside a nanometre-size layer on the alumina that was mostly composed of γ -TiO grains. Occasionally, single grains of Ti₃O₂ were observed in the thin layer on alumina. In the joints prepared with Ag–35.3Cu–1.8Ti wt.%, the interfacial structure evolved considerably with joining time, eventually leading to a high degree of inhomogeneity across the length of the joint at the highest T_p . The level of purity of alumina was not found to affect the overall interfacial microstructure, which is attributed to the formation of various solid solutions. It is suggested that Ti₃Cu₃O forms initially on the alumina. Diffusion of Ti occurs subsequently to form titanium oxide at the Ti₃Cu₃O/alumina interface.

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1. Introduction

Alumina ceramics are often joined to metals to form assemblies with a diverse range of applications, such as vacuum feedthrough technology for high voltage and pressure environments, semiconductor housing and accelerator, plasma and laser technologies [1]. Applications for alumina–alumina joints are more limited. These joints are usually made when ceramic components with complex geometries are required [2].

The various methods for fabricating ceramic to ceramic and ceramic to metal joints for several engineering ceramics have been recently reviewed [3]. Of these, brazing is a relatively simple and versatile technique to join similar and dissimilar materials (notwithstanding any issues relating to joint design). Currently, there are two commonly used brazing techniques in industry to

join alumina ceramics to metals and to themselves. These are active metal brazing (AMB) and sintered metal powder processing (SMPP); the most commonly known variant of SMPP being the moly–manganese (Mo–Mn) process. The Mo–Mn process was originally developed for zirconium silicate, magnesium silicate and oxide ceramics such as alumina containing a glassy secondary phase [4]. It is a multi-step process, which is achieved by modifying the bonding surfaces of the ceramic to render them more wettable by a conventional braze alloy, such as the Ag–28Cu wt.% eutectic alloy. Hey [5] has discussed several practical aspects of this process relating to the preparation of the ceramic, the composition and application of the primary metallising layer, and subsequent firing, plating and further heat treatment steps. The Mo–Mn process is lengthy and considerably more complex than AMB. During AMB, joining is achieved in a single step using an alloy that has been chemically modified with the addition of an element to cause precipitation of intermetallic compounds at the braze alloy/alumina interface. These interfacial phases are wetted by

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the braze alloy, and so their formation can be considered as an in-situ metallisation process. The element added is commonly known as an active element and the resultant braze alloy is called an active braze alloy (ABA).

The majority of alumina brazing studies have used ABAs based on the Ag–Cu system activated by small amounts of Ti, typically ranging from 1 to 5 wt.%. Other group IV and V elements such as Zr, Hf and V can also be added to Ag, Cu and Ag–Cu alloys [6–9], but the resultant binary and ternary ABAs have received far less attention than the Ag–Cu–Ti system. By comparison, X-ray diffraction (XRD) and electron microscopy investigations to characterise the reaction products at Ag–Cu–Ti thin foil/Al₂O₃ interfaces have been numerous. A large selection of reports on the interfacial phases that have formed using various joining conditions in the Al₂O₃/Ag–Cu–Ti/Al₂O₃ system are summarised in Table 1; the studies are arranged so that the concentration of Ti in the Ag–Cu–Ti alloy is increasing, and, for the same concentration of Ti, the peak joining temperature is also increasing.

A common characteristic in the majority of these studies is the formation of a micrometre-size reaction bilayer at the interface, which is typically reported as being composed of a Ti_xO_y compound in the form of a thin continuous layer on alumina, along with a thicker M₆O layer, where M is a mixture of Ti and Cu, in contact with the braze alloy. However, there are several points of disagreement on the stoichiometry of Ti_xO_y and M₆O that form. For example, Hahn et al. [10] suggested TiO_{1.04} and Ti₄Cu₂O form by heat treating 96 wt.% α-Al₂O₃ with Ag–33.5Cu–1.5Ti wt.% at 830 °C for 10 min, while Stephens et al. [11] found evidence for γ-TiO and Ti₃Cu₃O when a similar ABA (Ag–34.1Cu–1.7Ti wt.%) was held at 845 °C for 6 min on sapphire. In a more recent study, Lin et al. [12] found evidence for Ti₃O₂ (designated Ti₂O) and Ti₃Cu₃O at a 99.9 wt.% α-Al₂O₃/Ag–26.7Cu–4.5Ti wt.% interface, which was held at 915 °C for 20 min. In addition, various binary compounds such as Cu₂O [13], AlTi and CuTi₂ [14] have been reported.

The inconsistencies in the reported data have provided motivation for the current work to establish conclusively, using

Table 1
Reaction products observed at Ag–Cu–Ti thin foil/Al₂O₃ interfaces, which have been prepared in vacuum or with inert gases.

Al ₂ O ₃ /Ag–Cu–Ti interface (wt.%)	Joining conditions		Interfacial reaction product(s)		Method ^a	Refs.
	T _p (°C)	τ (min)	Phase	Thickness (μm)		
>96.0%Al ₂ O ₃ /Ag–33.5Cu–1.5Ti	830	10	TiO _{1.04} Ti ₄ Cu ₂ O	– –	TEM EDS WDS EPMA	[10]
99.0%Al ₂ O ₃ /Ag–28.1Cu–1.5Ti	950–1100	1–10	δ-TiO (only at 950 °C) α-TiO Ti ₃ Cu ₃ O	– – –	XRD EDS AES	[16]
Al ₂ O ₃ /Ag–35.1Cu–1.6Ti (0001) Sapphire/Ag–34.1Cu–1.7Ti	850 845	5 6	Ti _x O _y γ-TiO Ti ₂ O ^b Ti ₃ Cu ₃ O	1.5–2.3 0.02–0.2 – ~2.2	OM TEM EDS EPMA AES	[17] [11]
99.5%Al ₂ O ₃ /Ag–36.1Cu–1.8Ti Al ₂ O ₃ /Ag–44.8Cu–1.8Ti	900 950	15 15	Ti ₃ (Cu + Al) ₃ O γ-TiO Cu ₂ O Ti ₂ O ₃	2–3 – – –	EDS–SEM TEM EDS–SEM	[18] [13]
Sapphire/Ag–35.0Cu–2.0Ti	825	10	(Ti + Cu + Al) ₆ O	>1	TEM EDS EELS	[19]
Al ₂ O ₃ /Ag–36.0Cu–6.0Sn–2.0Ti	900	20	γ-TiO Ti ₃ (Cu + Al + Sn) ₃ O	0.1–0.2 ~3.0	TEM EDS	[20]
99.5%Al ₂ O ₃ /Ag–26.8Cu–2.9Ti	800–1200	15	γ-TiO Ti ₃ Cu ₃ O	– –	TEM EPMA	[21]
Al ₂ O ₃ /Ag–27.2Cu–3.0Ti Al ₂ O ₃ /Ag–38.8Cu–3.0Ti Al ₂ O ₃ /Ag–48.5Cu–3.0Ti Al ₂ O ₃ /Ag–67.9Cu–3.0Ti	800–1200	15	γ-TiO Ti ₃ Cu ₃ O	1.5–2.4 3.2–5.5	XRD EDS EPMA	[22]
99.9%Al ₂ O ₃ /Ag–48.1Cu–3.8Ti	920	20	TiO _{1±x} (x = 0.1) Ti ₄ Cu ₂ O	~1.2 ~2.4	XRD EDS AES	[23]
Al ₂ O ₃ /Ag–17.9Cu–4.2Ti 99.9%Al ₂ O ₃ /Ag–26.7Cu–4.5Ti	900 915	30 20	Ti ₃ Cu ₃ O and Ti ₄ Cu ₂ O Ti ₃ O ₂ (designated Ti ₂ O) Ti ₃ Cu ₃ O	3.0–5.0 0.6–1.2 5.4–6.3	EDS TEM EDS	[24] [12]
99.9% Sapphire/Ag–28.4Cu–4.9Ti	900	5	γ-TiO Ti ₃ Cu ₃ O ^c	0.01–0.05 1.0–2.0	TEM EDS EELS	[25]
99.9%Al ₂ O ₃ /Ag–38.0Cu–5.0Ti	800–850 900–1050	0–60 ^d	AlTi Ti ₄ Cu ₂ O TiO and Ti ₂ O CuTi ₂	e	XRD	[14]
99.8%Al ₂ O ₃ /Ag–27.0Cu–5.0Ti	980	5–90	(Ti + Al) ₄ Cu ₂ O	–	XRD	[6]

^a TEM includes techniques such as selected area diffraction and convergent-beam electron diffraction; XRD includes the technique of glancing angle-XRD, optical microscopy (OM), energy-dispersive X-ray spectroscopy (EDS), wavelength-dispersive X-ray spectroscopy (WDS), electron probe microanalysis (EPMA), Auger electron spectroscopy (AES), scanning electron microscopy (SEM) and electron energy loss spectroscopy (EELS).

^b Isolated Ti₂O particles were observed between the γ-TiO and Ti₃Cu₃O layers.

^c Most frequently observed microstructure. Isolated particles of Ti₂O were observed in the γ-TiO layer, along with isolated areas of Ti₃Cu₃O without TiO and occasionally a reaction phase without Ti, that is a Cu–Al–O phase.

^d 30 min for 800–1050 °C and 0–60 min for 850 °C were used.

^e The total reaction layer thickness was found to vary significantly with T_p and τ.

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