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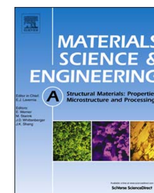

Low-temperature metal-to-alumina direct bonding process utilizing redox reaction between silver oxide and organic agent

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ABSTRACT

In the present study, low temperature bonding of gold-to-alumina was investigated by the in situ generation of silver via a redox reaction between silver oxide microparticles and diethylene glycol. The shear strength of gold-to-alumina joints with a silver layer bonded at 400 °C was comparable to that of conventional methods that require a high bonding temperature. Microstructural observations revealed that the adhesion of silver nanoparticles, which are formed by redox reaction, on an alumina surface improves the strength leading to the fracture of alumina. High-resolution transmission electron microscopy and energy dispersive spectroscopy analysis showed the direct bonding between silver and alumina at the interface without diffusion or formation of a reaction layer. Additionally, x-ray photoelectron spectroscopy analysis at the interface showed that the bonding of silver and alumina could be attributed to the silver-oxygen bond, which revealed that silver ions generated during the redox reaction could contribute to the bond formation. Further, molecular dynamics simulation confirmed that the presence of silver ions promotes the formation of an initial silver layer, which plays a role in bonding silver nanoparticles to an alumina surface. As mentioned above, it was found that a bonding method utilizing the reduction of silver oxide by reducing agent involves a significant, unique process that realizes metal-to-alumina bonding at a low temperature.

1. Introduction

Successful ceramic-metal bonding is of considerable practical interest for the development of electronic devices. There are marked differences in physical, chemical, and mechanical properties between ceramics and metals, which complicate the bonding process. Therefore, it is generally significant to impart wettability or reactivity to the ceramics to form the appropriate metal/ceramics interface.

Various bonding methods such as brazing and solid-state diffusion bonding have been investigated for producing a reliable joint between metals and ceramics [1–10]; there are many reports on alumina as a representative ceramic in particular. Brazing of ceramics involves surface metallization before the brazing [1,2], the addition of active metal to braze alloy, e.g. titanium to silver-copper alloys [3,4], and transient liquid-phase bonding [5–7]; the wetting and adhesion are promoted owing to obtaining the low contact angles. These processes are mainly carried out at a bonding temperature around the melting point of braze alloy, which may induce thermal stress [1,4] under vacuum. Additionally, the formation of intermetallic compounds is likely depending on the chemical reaction between the ceramic surface and active elements [7]. Solid state bonding utilizing the diffusion of inserted metal has been also investigated for metal-to-ceramics bonding [8–10]. The bonding temperature is usually equivalent to the melting point of the inserted metal in order to promote diffusion into ceramics; diffusion into Al_2O_3 results from the bulk diffusion through the grain boundary or dislocations [11,12]. As stated above, conventional bonding methods require a multistep, high-temperature process that restricts applications requiring a low temperature. Thus, the development of an effective method for reducing the bonding temperature in metal-ceramic bonding is necessary.

It is well known that nanoparticles below 10 nm show an apparent melting-point depression [13–15] caused by the high surface to volume ratio [16], making them more reactive. Hence, many researchers have reported that nanoparticles can be applied to the bonding of metals by sintering process [17–19]. While typically the preparation of nanoparticles generally requires advanced fabrication and control techniques because of the cohesive property [20], Hirose et al. reported the in situ generation of silver nanoparticles during the bonding process via

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the combination of silver oxide microparticles and organic agents (silver oxide paste) [21]. Organic agents can act as a reducing agent for the silver oxide based on the difference in the Gibbs free energy of oxide formation [22]. While various metals such as gold, copper, and nickel are directly bonded to the sintered silver layer [23], aluminum is bonded to the silver layer though the natural aluminum oxide film [23]. Although a comprehensive understanding of the bonding morphology at silver/alumina interface, e.g. either formation of compound or direct bonding, the result should provide the possibility of low-temperature bonding between silver and alumina bulk substrate by utilizing in situ generation of silver during the redox reaction.

It is important to elucidate the bonding morphology between silver and alumina in order to obtain a sound joint. The adsorption behavior of silver atoms or clusters on the alumina surface may possibly result in their bonding [24], in addition to diffusion or the formation of a reaction layer in conventional bonding methods. Certainly, it is known that the bonding of silver atoms can bond to alumina surface owing to either physisorption for aluminum termination [25–27] or ionic bond for oxygen termination [28,29]. Hence, it is important to basically elucidate the mechanism for silver-to-alumina bonding in order to ascertain the process to be applied to various ceramics.

In the present study, we demonstrate the low temperature bonding of gold and alumina substrate, without any conventional surface treatment, through the silver layer formed in situ by a redox reaction between silver oxide and a reducing agent. The influence of crystal orientation of alumina substrate, bonding temperature, and pressure on the joint strength is evaluated in order to confirm the possibility of low temperature bonding. The morphology of silver and alumina bonding is investigated by microstructural observations of the interface. Further, the bonding mechanism is discussed based on the analysis of the interfacial bonding state and the bonding process of silver to alumina.

2. Experimental procedure

Silver oxide (Ag_2O) particles with 2–3 μm diameters were used as the bonding material. The particles were milled for 10 min using an agate mortar and mixed with diethylene glycol as the organic agent to a concentration of 180 μl/g. Then, the mixture was processed to form a paste for bonding (hereafter referred to as the silver oxide paste). The thermal characteristics of the silver oxide paste were measured by performing differential thermal analysis (DTA) and thermogravimetric analysis (TGA) (Rigaku, TG8120) at 10 °C/min heating rate in air.

Au, in cylindrical shape with 5 mm diameter and 2 mm thickness, were fabricated by Ni/Au plating with a thickness of 3 μm and 1 μm, respectively, on a Cu disc. Pure polycrystalline Al_2O_3 was obtained from Kojundo Chemical Laboratory Co., LTD. in cylindrical shape, with dimensions of 10 mm diameter and 5 mm thickness. Monocrystalline Al_2O_3 with orientations $(11\bar{2}0)$ and (0001) was obtained from Techno Chemics, Inc. in plate shape, with dimensions of 10 mm square and 0.5 mm thickness. The surface of polycrystalline Al_2O_3 was polished with emery paper (up to 4000 grade) in order to decrease the initial roughness. Both Au and Al_2O_3 samples were cleaned by acetone in an ultrasonic cleaning bath. An Au substrate was used as the upper bonded material and a polycrystalline Al_2O_3 substrate was used as the lower bonded material, as shown in Fig. 1. For comparison, monocrystalline Al_2O_3 substrates were also used to evaluate the crystal orientation dependence on bondability between Al_2O_3 and Ag.

Silver oxide paste was applied to the surface of the Al_2O_3 substrate with a thickness of 50 μm and then preheated at 100 °C for 11 min in order to remove the excessive agent; excessive agent is likely to inhibit the sintering of Ag. The Au substrate was placed on the silver oxide paste layer after preheating. The samples were heated to the bonding temperature of 300–500 °C at a rate of 60 °C/min in an infrared heating furnace and held for 5 min with a pressure of 5 MPa. The samples were cooled by forced air after bonding.

In order to evaluate the influence of bonding temperature and applied pressure to the bondability, the shear strengths of Au-to-Al_2O_3 joints bonded at 300–500 °C with a pressure of 5–15 MPa were measured with a strain rate of 30 l/min as shown in Fig. 1(b). The shear strength was evaluated as the average value of three joints, and the error bars were determined as the maximum and minimum values. The cross-sectioned joints were prepared by using a cross section polisher (CP; JEOL SM-09010). The cross section and fractured surface of the joint was observed using a scanning electron microscope (SEM; Hitachi S-3000H). The interfacial microstructure between the sintered Ag layer and the Al_2O_3 substrate was investigated using transmission electron microscopy (TEM; JEOL JEM-2100F) combined with energy dispersive X-ray spectroscopy (EDS). Thin foil samples for TEM observations were prepared with a focused ion beam (FIB; Hitachi FB-2000A) processing method. The relationships between Ag, Al, and O atoms at the Ag/Al_2O_3 interface were determined using X-ray photoelectron spectroscopy (XPS; SHIMADZU ESCA-850M). XPS spectra were obtained from the sintered Ag layer formed on the Al_2O_3 substrate after argon ion sputtering from the top.

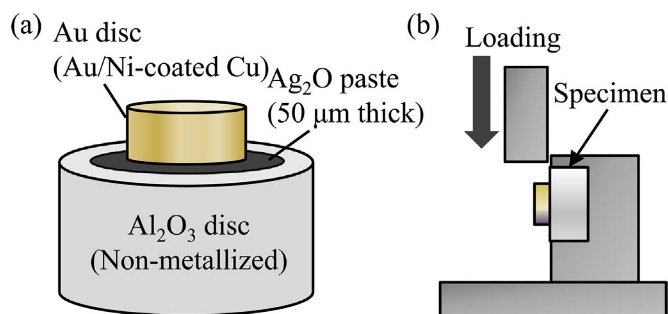

Fig. 1. Schematic illustration of shear test: (a) construction of a shear test sample; (b) shear test method.

Molecular dynamics simulation was performed by using the conventional software SCIGRESS ME 2.0 (Fujitsu Ltd.). For potential functions, GEAM04 potential, CMAS potential, and ME3Organic potential, which are included in the MD software package, were applied. The simulated temperature was set to 400 °C. The orientation of the Al_2O_3 substrate was $(11\bar{2}0)$ and the velocity of atoms in the substrate was set as 0.

3. Results

3.1. Thermal analysis of silver oxide paste

The DTA and TGA results are shown in Fig. 2, where blue and red lines correspond to DTA and TGA profiles, respectively. An exothermal DTA peak accompanied by significant weight loss in TGA was confirmed at 110 °C. The position of this peak indicates the formation of Ag through the redox reaction between the Ag_2O and diethylene glycol. The formation of excess amounts of Ag nanoparticles initially during the preheating process could result in their cohesion and lead to inhibition of sintering. Hence, the preheating temperature was lower than 110 °C.

3.2. Influence of crystal orientation of Al_2O_3 substrate

Fig. 3 shows the SEM images of the cross section of the Au-to-Al_2O_3 joints using monocrystalline and polycrystalline Al_2O_3 substrate bonded at 400 °C with a pressure of 5 MPa. Both substrates bonded via the sintered Ag layer: Ag particles adhered not only to the Au, but also to the Al_2O_3 substrates regardless of the difference in crystallinity.

Fig. 4 shows the shear strength of the Au-to-Al_2O_3 joints. The shear strength of the Pb-5Sn solder, the conventional industrial standard for die-bonding is shown with a broken line for comparison (~ 18 MPa). The strength of the joints was 18–24 MPa, which is higher than that of

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