

Joining of Metal to Ceramic Plate Using Super-Spread Wetting

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Abstract: Ceramic-metal composites with novel performance are desirable materials; however, differences in their properties result in difficulties in joining. In this study, the joining of metal to ceramic is investigated. We recently succeeded in causing super-spread wetting on the surface fine crevice structures of metal surfaces produced by both laser irradiation and reduction-sintering of oxide powders. In this work, joining copper onto an Al_2O_3 plate was achieved by taking advantage of super-spread wetting. Fe_2O_3 powder was first sintered under reducing conditions to produce a microstructure which can cause super-spread wetting of liquid metal on an Al_2O_3 plate. A powder-based surface fine crevice structure of metallic iron with high porosity was well-formed due to the bonding of the reduced metallic iron particles. This structure was joined on an Al_2O_3 plate with no cracking by the formation of an FeAl_2O_4 layer buffering the mismatch gap between the thermal expansion coefficients of iron and Al_2O_3 . We successfully achieved metalizing of the Al_2O_3 surface with copper without interfacial cracks using super-spread wetting of liquid copper through the sintered metallic iron layer on the Al_2O_3 plate. Then, laser irradiation was conducted on the surface of the copper-metalized Al_2O_3 plate. A laser-irradiated surface fine crevice structure was successfully created on the copper-metalized Al_2O_3 plate. Moreover, it was confirmed that the super-spread wetting of liquid tin occurred on the laser-irradiated surface fine crevice structure, finally accomplishing the joining of a copper block and the copper-metalized Al_2O_3 .

Keywords: metal-ceramic joining; super-spread wetting; wetting; surface fine crevice structure; laser irradiation; reduction sintering

1. Introduction

Ceramic-metal joining is required in a wide range of engineering applications on account of the beneficial properties of this material combination. Brazing, a bonding technique in which a continuous interface between two solid materials is formed via the spreading of a filler liquid metal into a joint, has been widely employed for ceramic-metal joining [1–3]. However, two major problems exist for the joining of a ceramic and a metal by brazing: namely, the poor wettability of ceramics by most metals and the large differences in physical properties between ceramics and metals [4–7]. Because a molten metal generally cannot wet ceramic materials, ceramic-metal joining by brazing starts with metallization of the ceramic to create a more wettable surface [4,5]. The thermal expansion coefficient of a ceramic is generally much lower than that of a metal. When a material is cooled from a metalizing or brazing process, the thermal expansion mismatch generates thermal stress at the

ceramic-metal interface, eventually resulting in cracks [6,7]. Therefore, achieving a successful joint in a ceramic-metal join by brazing is still a challenge.

Alumina (Al_2O_3) is a popular ceramic material because of its various properties, including high strength, high resistance to wear and corrosion, and high electrical insulation [8–10], so joining alumina ceramic to a metal has become an important topic. A combination of metal- Al_2O_3 , for example, Cu- Al_2O_3 joining, may provide not only the abovementioned characteristics of Al_2O_3 , but also the advantages of copper, which include high conductivity and machinability. It has been reported that the molten copper-solid Al_2O_3 couple shows contact angles of 158° – 170° at 1100 – 1300°C under oxygen-free conditions [11,12], which indicates that these materials are non-wetting. Likewise, most metals exhibit poor wetting ability for ceramics; such low wettability of Al_2O_3 by liquid copper is one of the major difficulties in achieving a good Cu- Al_2O_3 join.

Our research group discovered a phenomenon known as “super-spread wetting”, in which a liquid metal penetrates into the fine surface structure of a solid metal that has high porosity, similar to that of a sponge structure, by capillary action, and completely spreads on that surface [13–21]. Our research group [13–15] originally found that super-spread wetting occurs on the surface fine porous structure that is produced by atmospheric oxidation-reduction treatment of a metal surface. In further studies, we found two kinds of surface fine crevice structures that can promote region-selective super-spread wetting of liquid metals: namely, (1) a laser-irradiated surface fine crevice structure, created by laser irradiation [16–20] and (2) a powder-based surface fine crevice structure, created by reduction sintering of an oxide powder [21] on copper and iron surfaces. Super-spread wetting of these two surface fine crevice structures showed that similar and dissimilar metal-to-metal joining, such as Cu-Cu [16,17,21] and Cu-Fe [20] joining, can be achieved. Super-spread wetting is believed to be applicable to solving limitations of ceramic-to-metal joining caused by low wettability.

In this study, a ceramic-metal joining technique using super-spread wetting is proposed to address the challenge of joining an Al_2O_3 ceramic plate and copper block as a representative ceramic-metal combination. The technique comprises two steps: metallization of the ceramic surface and joining between the metallized ceramic and metal part. In the metallization step, we first carried out sintering of Fe_2O_3 powder under an atmospheric-reducing condition in which Fe_2O_3 is reduced to FeO and metallic Fe. This provides the method for producing a powder-based surface fine crevice structure that can cause super-spread wetting on the surface of the Al_2O_3 plate. We then metallized the Al_2O_3 plate with copper using super-spread wetting of liquid copper on the surface structure of the sintered metallic iron layer formed on the surface of the Al_2O_3 plate. In a joining step, laser irradiation was used to form a surface fine crevice structure on the metallized Al_2O_3 plate. Finally, metal copper-to-metallized Al_2O_3 plate joining by super-spread wetting on the surface fine crevice structure was attempted.

2. Experimental Method

2.1. Metallizing

Using the characteristics of super-spread wetting, we first investigated copper metallizing on an Al_2O_3 plate by forming a metallic iron surface fine crevice structure and penetrating copper into that structure. Figure 1a shows a schematic diagram of the experimental procedure. A $15\text{ mm} \times 15\text{ mm} \times 2.5\text{ mm}$ plate of Al_2O_3 (99.5% purity), Fe_2O_3 powders ($<3\text{ }\mu\text{m}$, 95% purity), and a copper block (99.99% purity) were used. A slurry was first prepared by mixing 2.2 g of Fe_2O_3 powder with 4 mL of ethanol. The Fe_2O_3 slurry was applied onto the Al_2O_3 plate and the ethanol evaporated. The sample was heated to sinter the Fe_2O_3 powders plated on the surface of the Al_2O_3 plate. The temperature profile for sintering is shown in Figure 1b. The atmosphere in the furnace was maintained during heating using an Ar (99.99% purity) gas flow rate of 6 mL/min. When the temperature reached 1373 K, a mixture of CO_2 gas (99.99% purity, 60 mL/min flow rate) and H_2 gas (99.995% purity, 15 mL/min flow rate) was introduced into the furnace and the Ar flow was stopped. The atmosphere in the furnace was maintained at an oxygen partial pressure about 10^{-12} atm during the sintering process to reduce Fe_2O_3 to FeO and metallic Fe. After this gas-exchange process, the temperature was kept constant for 1 h to

complete the sintering. The temperature was then decreased to 298 K in a mixed gas atmosphere of Ar gas (60 mL/min) and H₂ gas (15 mL/min). The obtained sintered sample was subsequently subjected to metalizing with copper using super-spread wetting. A mass of 0.2 g Cu was placed on the sintered metallic iron layer on the Al₂O₃ plate, as shown in Figure 1a. The melting point of copper is 1356 K [22], so the sample was heated to 1373 K to completely melt the copper using the heating process shown in Figure 1b. A reducing atmosphere with a mixed gas of Ar gas (60 mL/min) and H₂ gas (15 mL/min) was employed during the heating step of metalizing to prevent oxidation of the sample.

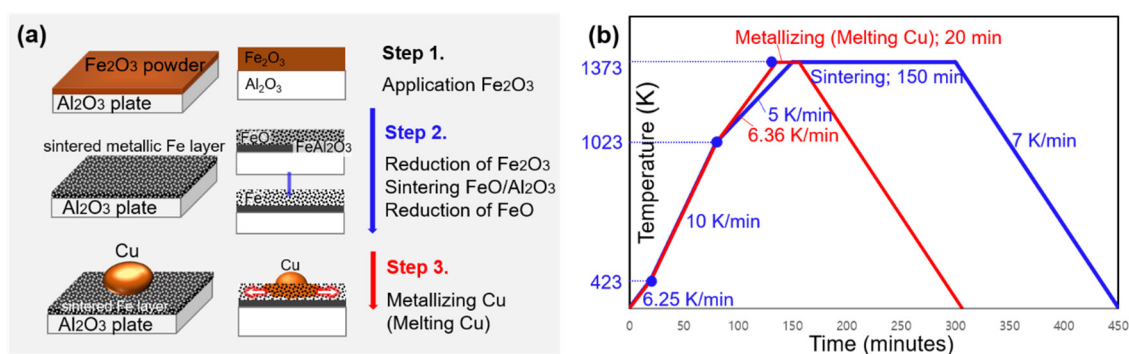


Figure 1. (a) Schematic diagram of experimental procedure; (b) Heating profiles used in Step 2 (blue line) of simultaneous reduction and sintering of applied Fe₂O₃ and in Step 3 (red line) of metalizing copper.

2.2. Joining

We attempted to join a copper block to the copper-metalized Al₂O₃ plate using super-spread wetting into a laser-irradiated surface fine crevice structure. Figure 2 illustrates the sample preparation and setting for the joining experiment. A 5 mm × 5 mm × 2 mm copper block was used as the metal object to be joined to the metalized Al₂O₃ plate and tin was used as liquid material to be penetrated between two materials to join them. They were cleaned in an ethanol bath with ultrasonic treatment. Laser irradiation was conducted to produce a surface fine crevice structure in a 5 mm × 7 mm region on the copper-metalized surface of the Al₂O₃ plate and the entire surface of one side of the copper block. A Q-switched Nd:YAG laser (ML-7062A, Miyachi Corporation, Tokyo, Japan) with a wavelength of 1064 nm was used for the laser-irradiation process. Each surface to be irradiated was positioned at a distance of 110 mm from the scanning lens, so that the laser beam focus was adjusted to a spot diameter of 0.1 mm. The maximum output power of the laser was 50 W at a frequency of 6.0 kHz. Laser irradiation was carried out at a scanning rate of 10 mm/s and a scanning interval of 0.01 mm, using a grid pattern, i.e., scanning in horizontal and vertical directions. Our previous study reported that copper oxides of CuO and Cu₂O are generated in response to laser treatment [16] under the same conditions as this experiment. Therefore, the copper block and copper-metalized Al₂O₃ plate were heated at 773 K for 1 h under reducing conditions with a H₂ gas flow rate of 20 mL/min to reduce these formed oxides. After reduction, the copper block was placed on the copper-metalized Al₂O₃ plate, such that the laser-irradiated surfaces were in contact with each other, as illustrated in Figure 2. A piece of tin, which was used as liquid metal to be infiltrated into the joint of the copper block and metalized Al₂O₃ plate, was placed on the laser-irradiated region of the underlaid copper-metalized Al₂O₃ plate. After transferring this setup into a furnace, residual air was evacuated, a flow of H₂ gas was introduced at a flow rate of 20 mL/min, and the sample was heated to 773 K at a heating rate of 10 K/min. Once the temperature reached 773 K, the sample was cooled down to 298 K at a rate of 10 K/min.

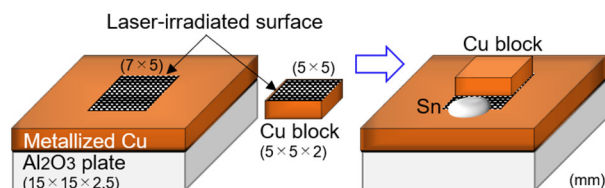


Figure 2. Illustration of the process used for joining experiments.

3. Results and Discussion

3.1. Copper Metallizing of Al_2O_3 Plate by Super-Spread Wetting into Powder-Based Metallic Iron Surface Fine Crevice Structure

Figure 3 shows scanning electron microscope observation of a cross-section of the Al_2O_3 plate after simultaneous sintering of the Fe_2O_3 powder and its reduction to metallic iron on its surface. Using energy-dispersive spectroscopy (EDS, JSM-6500F, JEOL, Tokyo, Japan), the compositions of the dark-gray-colored lower and white upper parts were identified as Al_2O_3 and metallic iron, respectively. Metallic iron particles bonded well with each other on the sintered layer, giving a thickness of around 140 μm . It was assumed that the thickness of the sintered metallic iron layer, i.e., the powder-based surface fine crevice structure of metallic iron, could be controlled by adjusting the amount of Fe_2O_3 slurry. Many pores formed between the bonded iron particles, which were irregularly connected and scattered on the sintered layer. In contrast, from the magnified image (Figure 3b), another very thin layer with thickness of around 3 μm was observed between the powder-based surface fine crevice structure of metallic iron and the Al_2O_3 plate. This was identified as FeAl_2O_4 by EDS analysis.

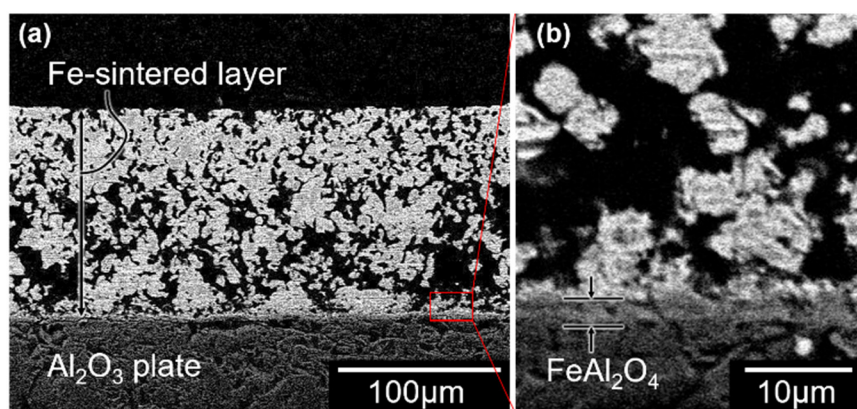


Figure 3. (a) Cross-section of the powder-based surface fine crevice structure of metallic iron created on the Al_2O_3 plate by sintering of Fe_2O_3 powder under reducing conditions; (b) Magnified image of the region of interface between Fe-sintered layer and Al_2O_3 plate.

It is supposed that each layer was produced by the following mechanisms. During the heating process at 1373 K, the oxygen partial pressure was maintained at about 10^{-12} atm. On the basis of the Fe-O phase diagram [23] given in Figure 4, FeO is presumed to be present at the high temperature. On the basis of the FeO- Al_2O_3 phase diagram [24] given in Figure 5, we expect that the FeAl_2O_4 phase appears at the FeO- Al_2O_3 interface at high temperature. Therefore, it is reasonable to assume that Fe_2O_3 was first reduced to FeO at around 1373 K, so that FeO reacted with Al_2O_3 to form FeAl_2O_4 at their interface. The $\text{CO}_2\text{-H}_2$ mixed gas was changed to an Ar- H_2 mixture when switching from holding to cooling, so that the oxygen partial pressure decreased during cooling. Therefore, the remaining FeO that did not react with Al_2O_3 was reduced to metallic iron during cooling in this atmosphere of decreased oxygen partial pressure. The suggested reactions can be described as follows:

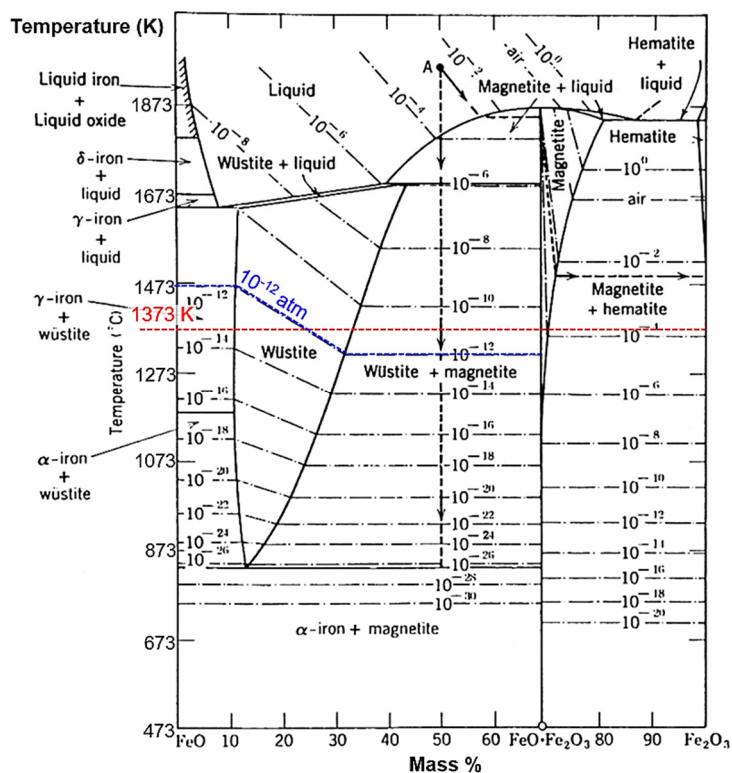
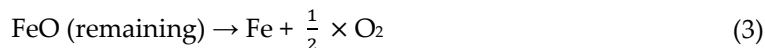


Figure 4. Phase diagram of FeO-Fe₂O₃ system with oxygen pressure isobars. Reproduced with permission from [23], Elsevier, 1999.

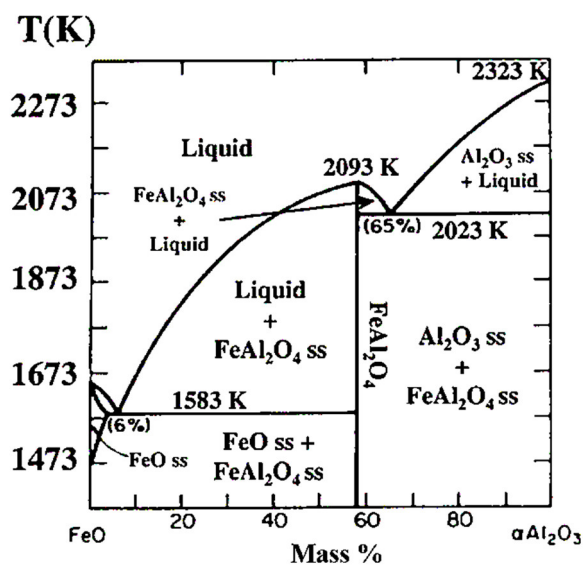


Figure 5. Phase diagram of FeO-Al₂O₃ system. Reproduced with permission from [24], Royal Society of Chemistry, 2002.

These interactions eventually led to bonding between the Al_2O_3 plate and the powder-based surface fine crevice structure of the metallic iron. No cracking was observed at the Al_2O_3 - FeAl_2O_4 -Fe interfaces. Fujimura et al. [25] reported that the thermal expansion coefficient of FeAl_2O_4 is larger than that of Al_2O_3 , but smaller than that of iron. Therefore, it was considered that FeAl_2O_4 acts as a buffer layer, which would reduce the mismatch gap between the thermal expansion coefficients of iron and Al_2O_3 . This system may make a good joint by suppressing crack formation at the interfaces. Consequently, it was confirmed that a powder-based surface fine crevice structure of metallic iron can be created on an Al_2O_3 plate via sintering Fe_2O_3 under reducing conditions.

Figure 6a shows the appearance of the sample after copper metalizing. The initial position of the copper piece was in the central part of the sintered metallic iron layer on the Al_2O_3 plate. However, the copper spread by wetting through the powder-based surface fine crevice structure of metallic iron, therefore, the entire region of the sintered metallic iron layer on the Al_2O_3 plate was covered with copper. From the cross-section of the sample, shown in Figure 6b, it was observed that the copper completely infiltrated the powder-based surface fine crevice structure of metallic iron. This indicates that super-spread wetting of copper occurred through this structure. However, as can be predicted from the Fe-Cu phase diagram [26], the metallic iron structure slightly dissolved into the copper. In addition, it was observed that no obvious voids or cracks formed on any interfaces of Cu-Fe, Fe- FeAl_2O_4 , or FeAl_2O_4 - Al_2O_3 , resulting in good metalizing of copper onto the Al_2O_3 plate. These results indicated that the metallization of the Al_2O_3 plate with copper was achieved by super-spread wetting of liquid copper via the powder-based surface fine crevice structure of metallic iron produced on the Al_2O_3 .

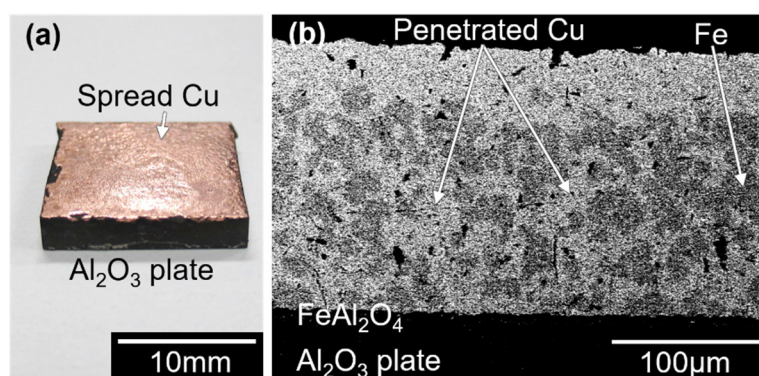


Figure 6. (a) Appearance and (b) cross-section of sample after copper metalizing.

3.2. Joining of Copper Block onto Al_2O_3 Plate with Surface Fine Crevice Structure Created by Laser Irradiation

Figure 7 shows the surface and cross-section of the copper (iron contained or dissolved)-metallized Al_2O_3 plate after laser irradiation. The laser-irradiated surface fine crevice structure of metallic copper consisted of many asperities and gaps formed on the surface of the metal layer of copper and iron on the Al_2O_3 plate. This structure is similar to the surface fine crevice structure created on a pure copper plate by laser irradiation [16,18]. In addition, as shown in Figure 6b, the FeAl_2O_4 layer remained, even after laser irradiation. This might serve to maintain the bonding of the metal layer of copper and iron and Al_2O_3 plate without cracking occurring at their interfaces. The above results confirmed that the laser-irradiated surface fine crevice structure of metallic copper was well produced on the copper-metalized Al_2O_3 plate by laser irradiation.

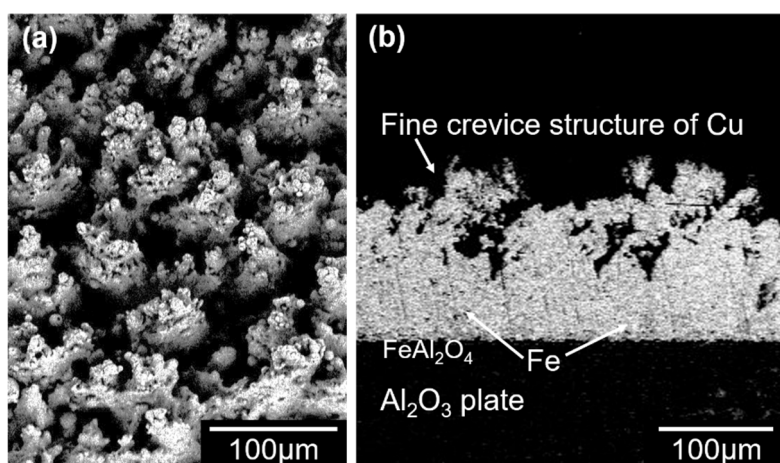


Figure 7. (a) Surface and (b) cross-section of laser-irradiated surface fine crevice structure of metallic copper created on the surface of copper (iron contained or dissolved)-metallized Al_2O_3 plate.

The appearance of the sample after joining is shown in Figure 8a. Tin placed on the laser-irradiated region of the underlaid copper-metalized Al_2O_3 plate disappeared by spreading into the joint interface between the upper copper block and the lower Al_2O_3 plate. Even when the upper copper block was lifted up, it was well joined with the copper-metalized Al_2O_3 plate so the two did not separate from each other.

Figure 8b shows a cross-section of the junction. All space between the upper copper block and lower copper metalized Al_2O_3 plate was completely filled with tin. Moreover, an intermetallic compound (IMC) layer was formed along the laser-irradiated surface fine crevice structure. We conducted composition analysis of some spots of the IMC layer by using EDS to identify the IMCs. The EDS analysis results indicate that the IMC layer formed at Cu block side was composed of Cu_3Sn and Cu_6Sn_5 , while that formed at the metalized Cu side included Cu_3Sn , Cu_6Sn_5 , and FeSn . This means that tin penetrated into the laser-irradiated surface fine crevice structure of both the upper copper block and the lower copper-metalized Al_2O_3 plate by super-spread wetting. In addition, it was presumed that both the upper and lower laser-irradiated surfaces changed into the intermetallic compound layer due to wetting by tin. However, some micro-pores were formed at the interface between the IMC layer and tin, as found in Figure 8b. Thermal expansion coefficients of Cu_3Sn , Cu_6Sn_5 , FeSn , and pure Sn are 19, 16.3, 19, and 23.4 ppm/ $^{\circ}\text{C}$ [27–29], respectively. The thermal expansion coefficient of Sn was significantly large compared to that of Cu_3Sn , Cu_6Sn_5 , and FeSn . Thus, it is supposed that this mismatch of thermal expansion (CTE) between the IMC layer and tin layer caused the formation of voids at the junction. These voids' formation at the joining interface of the joint would significantly degrade the mechanical robustness of the inter-connection [30]. It is considered that this problem can be solved by decreasing the mismatch of CTE on the joining interface, i.e., by determining the appropriate solder materials for application. Therefore, it is proposed that the joining of a copper block onto an Al_2O_3 plate was successfully accomplished by super-spread wetting of tin on the laser-irradiated surface fine crevice structure of metallic copper. Our research groups [16,18] have found that the surface fine crevice structures enable super-spread wetting of liquid Bi on solid Cu and also wetting of liquid Bi, Sn, and In on solid Fe, as well as the wetting of liquid Cu/solid Fe and liquid Sn/solid Fe. This means that the super-spread wetting of liquid metal on the surface fine crevice structure derived from the capillary action can allow one to metalize ceramic or join ceramic to metal with various ceramic-metal combinations. Consequently, we believe that this joining technique will help advance the joining of ceramic-metal systems with difficulty related to the lack of wettability.

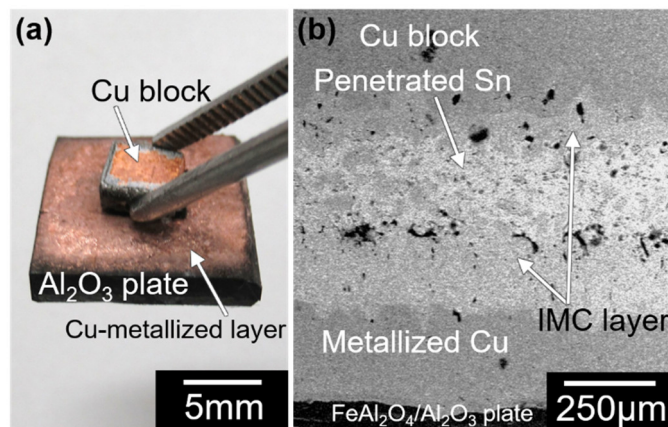


Figure 8. (a) Appearance of sample after joining experiment and (b) cross-section of the junction.

4. Conclusions

In this study, we proposed a method for joining a metal block to a ceramic plate using super-spread wetting. Metallization of Al₂O₃ with copper was first tried using super-spread wetting. An Fe₂O₃ powder sintered under reducing conditions was used to form a powder-based surface fine crevice structure of metallic iron that could lead to super-spread wetting of a liquid metal on an Al₂O₃ plate. We investigated the wetting of liquid copper on the powder-based surface fine crevice structure of metallic iron to metalize the surface of the Al₂O₃ plate. Formation of a laser-irradiated surface fine crevice structure of metallic copper on the copper-metallized Al₂O₃ plate and joining of a copper block to a copper-metallized Al₂O₃ plate using super-spread wetting into their surface fine crevice structures were attempted. The experimental results led to the following findings:

- (1) A powder-based surface fine crevice structure of metallic iron with a high porosity was created by sintering of Fe₂O₃ powder under reducing conditions. The sintered metallic iron layer bonded well to the surface of an Al₂O₃ plate due to the FeAl₂O₄ layer formed at the interface of the sintered metallic iron layer and Al₂O₃ plate during the heating process of the reduction of Fe₂O₃ to FeO.
- (2) Super-spread wetting of liquid copper occurred on the powder-based surface fine crevice structure of metallic iron, which achieved copper metalizing of the Al₂O₃ plate surface.
- (3) A laser-irradiated surface fine crevice structure was produced on the copper-metallized Al₂O₃ plate by laser irradiation. Joining of a copper block onto the copper-metallized Al₂O₃ plate was achieved using super-spread wetting of liquid tin through the structure.

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