



# Effects of brazing temperature on microstructure and mechanical performance of $\text{Al}_2\text{O}_3/\text{AgCuTi}/\text{Fe-Ni-Co}$ brazed joints



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## ARTICLE INFO

### Article history:

Received 6 February 2015

Received in revised form

24 July 2015

Accepted 25 July 2015

Available online 29 July 2015

### Keywords:

$\text{Al}_2\text{O}_3$  ceramic/ Fe–Ni–Co joint

Joint strength

Microstructure

Reaction layer

## ABSTRACT

$\text{Al}_2\text{O}_3/\text{Fe-Ni-Co}$  joints are achieved using Ag–Cu–8Ti filler alloy, and the dependence of the joint microstructure and mechanical performance on the brazing temperature has been studied by means of SEM, EDS, XRD and tensile test. The results show that the brazing seam is composed of TiO,  $\text{Ti}_3\text{Al}$ , Ag (s, s), Cu (s, s), (Cu, Ni) and  $\text{Ni}_4\text{Ti}_3$  phases. A layer of  $\text{Ti}_3\text{Al}$  and TiO products is observed at the  $\text{Al}_2\text{O}_3/\text{AgCuTi}$  interface and the fracture testing indicates that the thickness of the reaction layer plays a critical role in the joint strength. The joint strength firstly increases and then declines with the thickness of the ( $\text{Ti}_3\text{Al} + \text{TiO}$ ) layer increasing, and the formation of the cracks is ascribed to the existence of  $\text{Ti}_3\text{Al}$  phase. The thermokinetic analysis for the interfacial reaction between  $\text{Al}_2\text{O}_3$  and AgCuTi show that the Gibbs free energy equals  $-88.939$  kJ/mol for forming  $\text{Ti}_3\text{Al}$  and TiO phases, and the growth rate of the reaction layer mainly depends on the diffusion rate of Ti across the formed reaction layer. Meanwhile, the quantitative relationship among brazing temperature, holding time and reaction layer thickness has been established.

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## 1. Introduction

Ceramic materials are widely used in many areas of industries due to a combination of high-temperature resistance, corrosion resistance and abrasion resistance. However, the application of ceramic is always limited by its poor toughness and machinability, so in order to make its excellent properties fully utilized in practical applications, ceramic materials are usually joined with metals, which is Fe–Ni–Co alloy in this paper, to manufacture a component. The common methods to join ceramics and metals can be classified into two kinds: one is the method of surface metallization with subsequent Ni plating of ceramic surface before brazing, and the other is active brazing by using active filler containing a small amount of Ti, Zr or Hf elements. The method of surface metallization of ceramic has been a standard industrial process, but the inadequacies of complex process and time-consuming limit its far-ranging applications in engineering. To simplify operation process and shorten the production cycle, a method of directly brazing metal to ceramic with the active filler alloy has been appreciated.

The series of AgCuTi filler alloy are applied to the connections of various ceramics or ceramics to metals, and the effect of reaction process and reaction products between the ceramic and active filler on the joint strength have attracted the most attention in recent years [1–3]. Nanoindentation method is introduced for probing the mechanical properties of reaction phases of  $\text{Si}_3\text{N}_4/\text{AgCuTi}/\text{SiC-Si}_3\text{N}_4$  joint and the digital image correlation (DIC) technique is applied to clarified the strengthening mechanism of the joint [4]. The ZrB<sub>2</sub>–SiC composites (ZS) are also joined using an Ag–Cu/Ti filler alloy [5], and the result shows that the mechanical properties of ZS/ZS joint are improved by interfacial TiB accommodation due to the reduction of the coefficient of CTE mismatch. For  $\text{Al}_2\text{O}_3/\text{Ag-Pd-Ti/Kovar}$  joint [6], the effect of Ti content on the interfacial microstructure and mechanical properties shows that the type of titanium oxide are affected by the thickness of Ti layer, and the joint strength are influenced by the thickness of reaction layer and residual Ti layer. M. Brochu [7] studied the active brazing of  $\text{Si}_3\text{N}_4$  ceramic and FA-129 alloy, and they found a reaction layer formed at  $\text{Si}_3\text{N}_4/\text{Cu}$  interface and discussed the relation between the thickness and brazing time. The  $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$  joint can be achieved using Ag–Cu–Ti + B +  $\text{TiH}_2$  composite filler [8], the results show that the addition of B powders in composite filler increases TiB whiskers content, but decreases the thickness of  $\text{Ti}_3(\text{Cu,Al})_3\text{O}$  layer, while the higher  $\text{TiH}_2$  powders content thickens  $\text{Ti}_3(\text{Cu,Al})_3\text{O}$  layer.

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It is well accepted that during the active brazing process, the active element, such as Ti, in the molten brazing filler is absorbed to the surface of ceramic and forming a layer of reaction product with ceramic at the interface. The morphology and the properties of the reaction layer, which are determined by brazing process, play important roles in the joint strength. However, the relationship of brazing temperature, holding time and thickness of the reaction layer and the discussion of growth process of the reaction layer are rarely mentioned. In order to identify the dependence of seam microstructure and the joint strength on the brazing temperature, holding time and thickness of the reaction layer, AgCuTi filler alloy is applied to braze  $\text{Al}_2\text{O}_3$  and Fe–Ni–Co at different temperatures for various times in the present paper. The study also aims to investigate the reaction between AgCuTi filler with  $\text{Al}_2\text{O}_3$  ceramic, the controlling factors in the growth of the reaction layer, the reasons for the change of joint strength and the mechanisms of the formation of the microcracks during tensile testing.

## 2. Experimental

The ceramic and metal used in this work were 95 wt%  $\text{Al}_2\text{O}_3$  ceramic and Fe–33 wt% Ni–15 wt% Co alloy plates. Fe–Ni–Co alloy plates were chosen as another matrix treated with the surface nickel-plated to ensure a favorable spreadability of the brazing filler on it.  $\text{Al}_2\text{O}_3$  ceramic and Fe–Ni–Co alloy was assembled (shown as Fig. 1) after their surface for brazing were coated by a commercial paste Ag–25.92 wt% Cu–7.89 wt% Ti filler alloy (Lucas-Milhaupt, Inc).

Due to the high Ti content (7.89 wt%) in the used AgCuTi filler alloy, the samples were performed in a horizontal vacuum at a pressure of  $3.0 \times 10^{-3}$  Pa to avoid the oxidation of active Ti during heating. The heating curve was shown in Fig. 2. A heating rate of  $10^\circ\text{C}/\text{min}$  was set, which could effectively reduce the temperature gradient of samples to prevent the generation of internal stress. Heat preservation was set at  $450^\circ\text{C}$  holding for 30 min to make organic adhesive in AgCuTi filler fully decompose before being pumped. All samples were preheated to  $750^\circ\text{C}$  and held for 30 min before they were heated to the brazing temperature to reduce the time delay between the true specimen temperature and program temperature. In order to reduce the temperature gradient of samples and prevent the generation of stress, the sample was cooled at a rate of  $3^\circ\text{C}/\text{min}$  from the brazing temperature to  $720^\circ\text{C}$  and subsequently cooled inside the furnace to room temperature. The tensile strength of brazed joint was determined by RGX-M300 electronic universal testing machine with a speed of 0.5 mm/min.

For the characterization of metallographic structure, the samples were ground to 10 mm in thickness along the direction

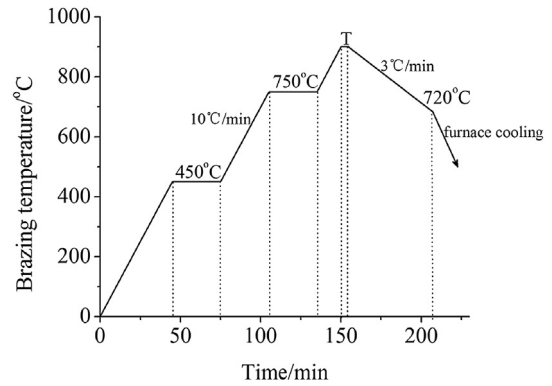


Fig. 2. The programming heating curve during brazing process.

perpendicular to the seam by diamond discs before being etched. Microstructure and reaction phases were examined by means of the HITACHI S-4800 scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDS). The different phases were identified using the X'Pert PRO X-ray diffractometer (XRD).

## 3. Results and discussion

### 3.1. Microstructures and interfaces

The microstructures of the brazing seam detected by SEM are shown in Fig. 3. The results indicate that the joints are mainly composed of three parts: an interfacial reaction layer with a thickness of approximately  $0.8\text{--}1.8\ \mu\text{m}$  adjacent to  $\text{Al}_2\text{O}_3$  ceramic, a brazing seam zone consisting of banded structures, lump phases and concave phases, and an interfacial reaction zone adjacent to Fe–Ni–Co alloy. Fig. 3(a) shows an interfacial reaction layer of about  $0.8\ \mu\text{m}$  in thickness between  $\text{Al}_2\text{O}_3$  ceramic and AgCuTi filler alloy, which indicates the occurrence of metallurgical reactions between  $\text{Al}_2\text{O}_3$  and AgCuTi filler. However, the faying interface between AgCuTi and Fe–Ni–Co is clear, because the dissolution of Fe–Ni–Co towards AgCuTi is largely limited at such a low temperature of  $870^\circ\text{C}$ . In comparison, an interfacial reaction zone with a tortuous joining line adjacent to Fe–Ni–Co alloy is observed in Fig. 3(b), which is benefited from the rise of brazing temperature ( $890^\circ\text{C}$ ). In addition, the reaction layer at  $\text{Al}_2\text{O}_3/\text{AgCuTi}$  interface in Fig. 3(b) is continuous and the thickness increases to  $1.4\ \mu\text{m}$  due to the violent interfacial metallurgical reaction that also caused by the rise of brazing temperature. Comparing the three samples brazed at different temperatures, the thickness of reaction layer adjacent to  $\text{Al}_2\text{O}_3$  ceramic increases with the rise of brazing temperature, and it reaches  $1.7\ \mu\text{m}$  at a brazing temperature of  $910^\circ\text{C}$ , as shown in black-bordered box in Fig. 3(c).

Fig. 4 shows the elemental diffusion of the cross section of  $\text{Al}_2\text{O}_3/\text{AgCuTi}/\text{Fe–Ni–Co}$  joint. The results reveal that Ti element aggregates to  $\text{Al}_2\text{O}_3$  surface (zone A) and in the banded structure (zone C), and Ni distributes in the whole brazing joint and present higher concentration in banded structure and near Fe–Ni–Co alloy. It is noted that Al and O aggregate in the layer at  $\text{Al}_2\text{O}_3/\text{AgCuTi}$  interface and Cu distributes in the whole brazing seam.

In order to identify the formed phases in the  $\text{Al}_2\text{O}_3/\text{AgCuTi}/\text{Fe–Ni–Co}$  brazing seam, an EDS analysis was conducted to investigate the composition of the phases as shown in Fig. 5. Table 1 shows the EDS analysis results of the  $\text{Al}_2\text{O}_3/\text{AgCuTi}/\text{Fe–Ni–Co}$  joint in Fig. 5. It can be seen that the lump phases (as marked by “D”) are Cu-rich and the concave phases are Ag-rich. As a result of

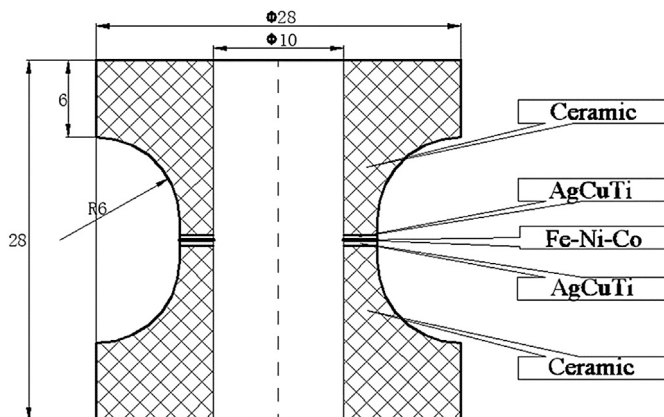


Fig. 1. Sketch of assembling jig for samples.

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