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Characterization of the Effects of Active Filler-Metal Alloys in Joining Ceramic-to-Ceramic and Ceramic-to-Metal Materials

D. Aliya^{1,a}, L. W. Walker^{2,b}, E. Montz^{2,c}, S. Pastor^{2,d}, A. Abad^{3,e},
F.A. Hashim^{4,f}, A. Abdul-Latif^{5,g}, A. O. Al-Roubaiy⁶, Y.S. Oh⁷,
H. Garmestani^{7,h}, N. Ula^{8,i}, and O. S. EsSaid^{2,j}

¹Aliya Analytical Inc., Grand Rapids, MI 49501, USA

²Mechanical Engineering Department, Loyola Marymount University, Los Angeles, CA 90045-8145, USA

³Northrop Grumman Corporation, One Hornet Way, El Segundo, CA 90425, USA

⁴Production Engineering and Metallurgy Department, University of Technology, Baghdad, Iraq

⁵Laboratoire d'Ingénierie des Systèmes Mécaniques et des Matériaux (LISMMA), Supméca, 3, rue Fernand Hainaut - 93407 St Ouen Cedex, France

⁶College of Engineering, University of Babylon, Babylon, Iraq

⁷Materials Science and Engineering Department, Georgia Institute of Technology, Georgia, USA

⁸Electrical Engineering Department, Loyola Marymount University, Los Angeles, CA 90045-814, USA

^aDaaliya@itothen.com, ^bweswalker19@gmail.com, ^cemontz@lion.lmu.edu,
^dspastorm20@hotmail.com, ^eadeleenabad@hotmail.com, ^fhashembsb@yahoo.com,
^gaal1958@gmail.com, ^hgarm@gatech.edu, ⁱNazmul.Ula@lmu.edu, ^joessaid@lmu.edu

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Abstract. This study investigated actively brazing Alumina-to-Alumina with Ag-Cu-Ti as the filler metal system and Alumina-to-Copper with Cu-Ti-Co as the filler-metal system. The research was conducted on four samples, two of which were alumina brazed to alumina (Samples 1 & 2), and the other two were alumina brazed to copper (Samples 3 & 4). The filler metal composition for each sample was as follows: Sample 1 consisted of Cu-96%, and Ti-4%; Sample 2 consisted of Ag-70%, Cu-26%, and Ti-4%; Sample 3 consisted of Cu-85%, Ti-10%, and Co-5%; and Sample 4 consisted of Cu-55%, Ti-40%, and Co-5%. The phase transformations between the filler and base metal of each brazed joint were studied using EDS, SEM, optical microscopy, and X-ray diffraction.

Introduction

Producing reliable ceramic joints when brazing ceramics-to-ceramics or ceramics-to-metals, known as the single stage actively brazed method, has recently been attracting a lot of interest from researchers. Much work has recently emerged to establish a joining technology that is based on sound, fundamental procedures, along with an understanding of the reactions that occur during joining [1-5]. There are two major challenges encountered when brazing metals to ceramics. The first challenge lies in that fact that the surface of a ceramic is very stable; consequently, metals in their molten phase do not wet well with ceramics due to the higher energy of the molten metal as compared to the bare ceramic surface. This causes the molten metal to bead up on the ceramic/metallic interface [3-5,6-8], which greatly restricts the uniform dispersion of the filler metal. Flow of the filler metal is then facilitated by the capillary attraction, which results from surface energy effects [3, 5, 7, 9]. The compositions and the surface energies of liquids and solids are assumed to remain constant on the interface but in real systems, interactions occur [5, 7, 10, 11]. The second challenge is due to the residual stresses caused by the mismatch in coefficients of thermal expansion between ceramics and metals [2, 3, 10]. This effect, combined with the fact that ceramics have low specific heat values, may result in cracking of the base ceramic [2, 11].

Hashim [1] and Xiong et al. [3] found that when joining ceramic to ceramic, Cu-Ti filler metal alloys demonstrate high joining strength. Additionally, the bonding phase of $\text{Cu}_2(\text{Al}, \text{Ti})_4\text{O}$

shows excellent bonding strength with the Cu-Ti active-filler metal alloys in comparison to the TiO phase, while AgCuTi filler metal alloys also demonstrate excellent joining strength when joining ceramic to ceramic [4]. Kuzumaki et al. [12] investigated the effect of cobalt and niobium on Ag-Cu-Ti alloy active system to join copper with aluminum nitride. They found that small amounts of cobalt (5 wt %) and niobium (5 wt %) added to active fillers are sufficient enough to improve the joint properties. They also claimed that cobalt exhibited the same behavior as titanium when brazing ceramic to copper.

For this study, high purity alumina (Al_2O_3) was chosen as the ceramic base because it is a more applicable ceramic in several industrial applications. The copper alloy base was chosen because of its availability, low price, and its melting temperature is between that of molybdenum and silver [1]. Xiong et al. [3] also suggested that the use of ceramics with low thermal expansion coefficients (such as Al_2O_3) would help relieve residual stresses produced during brazing. The aim of this research was to expand upon the previous findings of Hashim and Al-Roubaiy [1, 6]. This study focused on characterizing actively brazed alumina-to-alumina using Cu-Ti systems and alumina-to-copper using a Cu-Ti-Co filler-metal system.

Experimental Work

Materials used in this investigation were 99.89% pure polycrystalline alumina manufactured by ALM-41-Sumito Chemical Company, Japan, 99.8% pure copper and active filler metal alloys composed of: Cu-4wt%Ti, eutectic Ag-Cu-4wt%Ti, Cu-10wt%Ti-5wt%Co and Cu-40wt%Ti-5wt%Co. The dimensions of the alumina and copper at room temperature were 14 x14 x10 mm and were prepared by means of grinding with a series of emery papers (800 to 1200 grit). The samples were then degreased in an acetone bath by use of an ultrasonic vibration container and were then dried. In order to braze alumina to alumina or alumina to copper, suitable concentrations of (Ti, Cu, Ag, Co) powders were premixed and then ball milled for 8 hours to achieve uniform particle size. The mixed filler metal powders were weighed on a 0.001 mg sensitivity digital balance. Once uniform particle size of the powders was achieved, they were mixed with approximately 0.05 grams of glycerin which yielded approximately 0.15 grams of brazing filler alloy in the form of a paste suitable for producing one joint. The compositions of the four samples studied are as follows: In sample 1, alumina (Al_2O_3) was brazed to alumina by an active filler metal composed of 96% copper (Cu) and 4% titanium (Ti). In sample 2, alumina was again brazed to alumina by an active filler metal composed of 70% silver (Ag), 26% copper (Cu), and 4% titanium (Ti). In sample 3, alumina was brazed to copper by an active filler metal composed of 85% copper (Cu), 10% titanium (Ti), and 5% cobalt (Co). In sample 4, alumina was brazed to copper by an active filler metal composed of 55% copper (Cu), 40% titanium (Ti), and 5% cobalt (Co).

The clean surfaces of the alumina and copper squares were coated with the brazing paste approximately 0.1 mm thick. All joints were secured by using a simple fixture which utilized two sheets of molybdenum to hold the assembly together. Alumina samples were joined by a vacuum furnace brazing technique [1], which imparts a good standard of cleanliness. Alumina was then brazed to copper by means of an electrical resistance furnace in an argon atmosphere where the duration time of the brazing cycle was 2 hours. The samples were prepared with two holes to charge an argon atmosphere, delivered at a rate of 2 to 4 lit/min. in a vacuum brazing pressure environment of approximately 5×10^{-4} torr. Samples were heated to the brazing temperature of 1025°C and maintained at this temperature for 15 minutes. The holding time was one hour and the brazing time was 4 hours [1]. After brazing, each sample was cut evenly and perpendicular to the joint. Lastly, each sample was prepared for optical microscopy, SEM, EDS, and X-ray diffraction in order to characterize the bonding phases and identify the elements of the filler metals within the braze joint interface. An Olympus PME 3 optical microscope, a JEOL 6400 scanning electron microscope (SEM), an Inca EDS detector, and an X'Pert PRO MPD, PANalytical Co., Holland x-ray diffractometer were used in this study.

Results and Discussion

Optical Microscopy, SEM, EDS. For samples 1-4 the braze thicknesses were estimated to be 70 μm , 40 μm , 220 μm , and 230 μm respectively. The only significant defect that was visible in the microstructure was a crack running parallel to the brazed joint in sample 3. It was suspected that this crack resulted from the mismatched thermal expansion coefficients between the alumina and copper. This cracking phenomenon also occurred in the experiments of Singh and Asthana [4], Bobzin et al. [7], and Chen [11]. In Fig. 1, box A displays a standard SEM image in backscatter mode of the brazed joint of sample 1. Box B highlights the copper element present in the lighter areas. Box C highlights the aluminum present in the ceramic (alumina). Box D highlights the titanium present, indicated by the light areas. High concentrations of titanium are observed along the ceramic braze interface of box D (Fig. 1), suggesting a diffusion phenomenon. As seen in box B, copper is mostly concentrated in the middle of the brazed joint.

In Fig. 2, the diagonal line represents the path taken by the line scan, approximately 112 μm in length. Fig. 3 is a quantitative analysis of the line scan taken in Fig. 2. Box A (Fig. 3) is a general image of all reflections within the sample. Boxes B, C, and D show the frequency of finding copper, aluminum, and titanium, along the line scan of Fig. 2. The numbers found next to the elemental symbol on the top left corner of the boxes represent the amount of signals received from the element of interest. The crosshair represents the beginning of the line scan. It can be seen from Fig. 2 that the crosshair starts and the line scan ends in the ceramic (alumina) region where, as expected, a high frequency of aluminum is observed. Box C in Fig. 3 supports this observation in that there are high aluminum peaks at the beginning and the end of the chart. As the line scan enters the brazed joint, approximately 6 μm across, a high concentration of titanium is observed along the ceramic-braze interface (dark gray region) in addition to lesser concentrations of copper (both supported by Fig. 3, boxes B and D). As the line scan moves into the lighter gray region, approximately 18 μm across, boxes B and D demonstrate high concentrations of copper and a significant drop in titanium concentration. This pattern continues along the remainder of the line scan and boxes B and D demonstrate that areas rich in titanium are poor in copper, and vice versa. This can be confirmed by Chang et al. [10] findings related to the poor solubility of Cu in Ti.

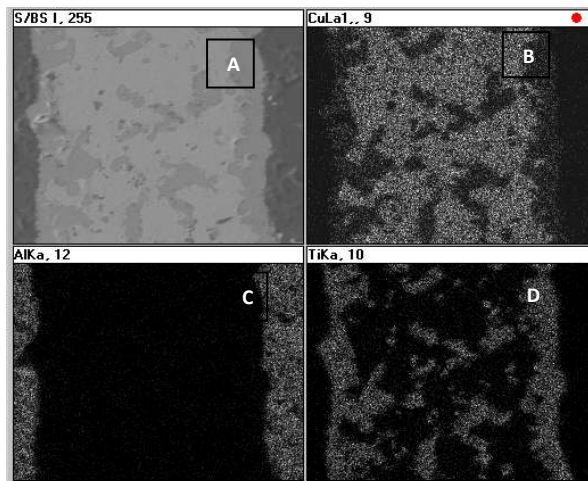


Fig. 1 Speed map on SEM backscatter mode 25 keV focused on braze joint at 1000X, sample 1

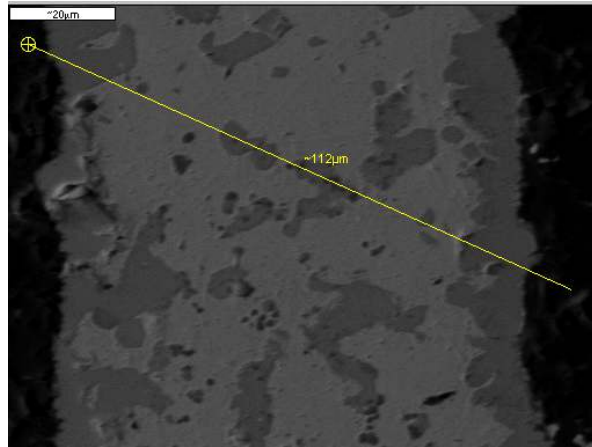


Fig. 2 25 keV Line Scan at 1000X over a length of 112 μ m (indicated by the line) of sample 1

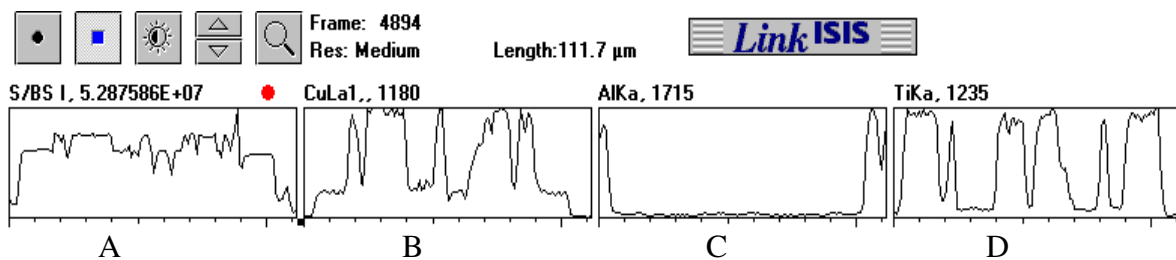


Fig. 3 Elemental Composition as a Function of Length along the Line Scan (each tick mark represents 10 μ m) sample 1

This analysis was also carried out on samples 2-4. In sample 2, Ti clearly diffuses into the ceramic and this suggests that Cu participates principally in the center of the bonding zone, while Ag is not involved there. According to the findings of Mandal and Ray [13], the activity of Ti increases in the presence of Ag and has a tendency to dissociate from the other filler metals, thus increasing the overall activity of the alloy in the brazing process. This is similar to what Liu et al. [4] found with Nb and Lee et al. [7] found with Zr. These observations appear to be consistent with Sample 2 because the EDS results show a higher concentration of Ti at the interface in sample 2 than was observed in sample 1, which did not contain Ag. Silver does not appear anywhere near the braze interface, which is consistent with Bobzin et al. findings [5] that Ag does not readily wet with ceramics, it only melts.

The EDS results for sample 3 indicate that high oxygen and alumina peaks are present at the beginning of the chart. Just before the brazed joint begins it was observed that there was a small amount of diffusion of Ti, Co, and Cu into the alumina. As the line scan entered the brazed joint, approximately 50 μ m across, a high concentration of Ti and Co is found with slightly lesser concentrations of Cu. As the line scan continues across the braze joint (approximately 250 μ m across) it was shown that a large amount of Cu was present as well as small concentrations of Ti and Co towards the ceramic braze interface. Alumina appeared to have diffused across the brazed joint with a slight diffusion of Ti and Co into the base metal.

The EDS results for sample 4 indicate that Ti, Co, and Cu are found within the brazed joint. It is again observed that Ti and Co have an affinity for one another. The presence of Co improves the wetting of the joint, which is in accord with the work of Hashim [1]. As expected, Ti is detected along the braze interface as a result of the attraction between Ti and Co. However, the intensity of the attraction seems to be less due to the higher concentration of Ti. In sample 4 several pores were detected. Hashim [1] and Singh and Asthana [4] also observed porosity when brazing ceramics. It was shown that unlike sample 3, the Co does not have an affinity for Ti and is rather uniform throughout the braze interface.

X-Ray Diffraction. X-ray diffraction was performed on all four samples. The presence of CuO and AlCu peaks were identified in samples 1 and 2, Fig. 4. This means that CuO and AlCu were formed during brazing process. Despite the compositions of the filler materials being different from each other, the XRD patterns are almost identical. Even though 70% of Ag exists in the filler metal of sample 2, no evidence of Ag or Ag-compounds can be found. This indicates that the majority of the Ag exists in the solid-solution, as Bobzin et al. [7] suggested. X-ray diffraction of sample 3 shows that peaks for CuO and AlCu are also present. As a result of the addition of Co to the filler material, Co-compounds, CoTi, AlCo and CoO, were detected. The peaks for CuO and AlCu are also confirmed in sample 4. The presence of CoO was not seen in sample 4, indicating that the higher concentration of Ti hindered the formation of CoO, as was similar to the findings of Kuzumaki et al. [12].

Future work could mainly focus on performing mechanical testing such as tensile testing, shear stress, and microhardness tests.

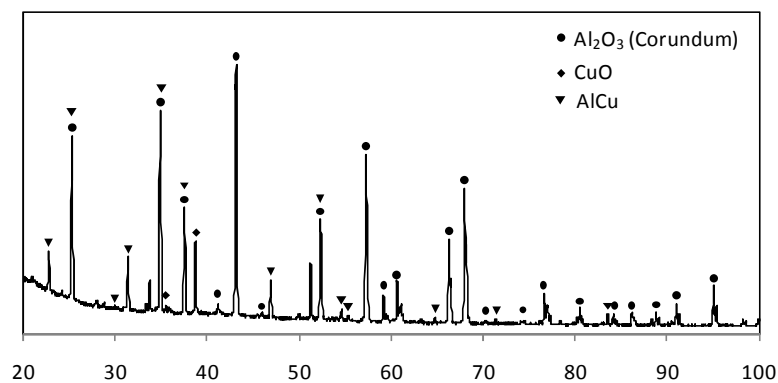


Fig. 4 X-ray diffraction pattern of sample 1

Conclusions

1. Sample 1 showed a concentration of Ti along the ceramic-braze interface. Cu was present mainly in the middle of the brazed joint, while Ti showed an affinity for the ceramic interface. Cu was principally in the bonding zone due to its chemical affinity with Ti. X-ray diffraction showed the creation of CuO and AlCu compounds indicating diffusive reactions between elements.
2. X-ray diffraction of sample 2 indicated that the presence of Ag does not appear to have much effect on the brazed joint. Hence, Ag is not involved in the bonding zone due to the chemical affinity between Ti and Al₂O₃. This is supported by the EDS scan as well. Again, Ti has a high affinity for the ceramic braze joint. Despite the different concentrations for the materials between samples 1 and 2, the compounds appear in the same patterns. It appears that this sample would provide a stronger brazed joint as compared to sample 1. The activity and dispersion of Ti and Cu is more intense which can be assumed to be a result of the addition of Ag. However, Ag does not react or form compounds with any of the elements in the sample.
3. In sample 3, the EDS scans showed the affinity of Co for Ti. Again, CuO and AlCu compounds were formed. X-ray diffraction also shows the formation of Co compounds, mainly CoTi, AlCo, and CoO. It is hypothesized that this filler metal would provide an overall stronger braze than sample 4 because of the increased activity between Ti and Co. However, there is some concern with the oxides formed in this compound that they might increase the brittleness of the brazed joint.

4. Sample 4 exhibited similar conditions as sample 3, however the CoO compound are not detected. This was most likely a result of the higher concentration of Ti. Porosity was also seen in the analysis of the EDS scan.

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