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Brazing of Boron Carbide by Cu-Alloys: Interface Interaction and Mechanical Properties of Joints

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Abstract

Boron carbide is one of the most promising structural ceramics; therefore its joining to other ceramics and metals is of technological importance. In this study, interface interactions and joints strength were studied in various cases of B_4C/Cu based fillers (Cu-9at%B, Cu-4.5at%Ti and Ticusil®). The fillers partially wet B_4C ($\theta < 50^0$), but the interface composition and morphology are different. In the B_4C/Cu-9at.%B system no new phases are detected at the interface, while the formation of TiB_2 takes place in the case B_4C/Cu-4.5at%Ti, and a double layer containing TiB_2 and TiC is formed at the B_4C/Ticusil® interface.

The maximal shear stress for B_4C/B_4C joints breakage was obtained for the Cu-9at.%B brazing alloy which displays partial wetting without the formation of brittle phases at the ceramic/metal interface.

This study shows that improved wetting is essential but not sufficient in order to obtain strong bonding between B_4C and the fillers examined.

Keywords: brazing, boron carbide, thermodynamics, mechanical properties, interfaces

1. Introduction

Successful performance of structural ceramic components depends on the quality and reliability of ceramic-to-ceramic and ceramic-to-metal joints. Direct brazing using active filler metals is one technique that can fulfill such requirements. In order to minimize the effect of thermal stresses it is desired to develop brazing alloys with relatively low melting temperature (based on Cu for instance), which display appropriate wetting properties. Adequate wetting of boron carbide may be achieved by using active brazing alloys. Yet, the major drawback of B_4C is related to its' low coefficient of thermal expansion ($4\text{-}8\cdot10^{-6}$ K^{-1}) compared to brazing metals ($16.6\cdot10^{-6}$ K^{-1} and $19.1\cdot10^{-6}$ K^{-1} for Cu and Ag, respectively) which might cause residual stresses at the interface (Naidich et al., 2008). The results of a systematical investigation of boron carbide wettability by various metals were reported in previous publications (Froumin et al., 2003; Frage et al., 2004; Aizenshtein et al., 2005a; Aizenshtein et al., 2005b; Aizenshtein et al., 2008a; Aizenshtein et al., 2008b) and the wetting mechanism is well understood. Experimental results for Ti and B alloyed Cu and Ti alloyed Ag are presented in Figure 1. In the B_4C/(Cu,Ag)-Ti systems, the improved wetting is achieved due to the formation of new phases at the interface, while in the B_4C/Cu-B system, the contact angle decreases as a result of the composition shift of boron carbide towards higher boron contents (Froumin et al., 2003).

In the present work the mechanical properties of brazed boron carbide using three Cu-based fillers, namely Cu-9at.%B, Cu-4.5at.%Ti and commercial Ticusil® alloy (68.8wt.% Ag, 26.7wt.% Cu, 4.5wt.% Ti) were investigated in order to clarify the correlation between the interface structure and mechanical properties of the joints.

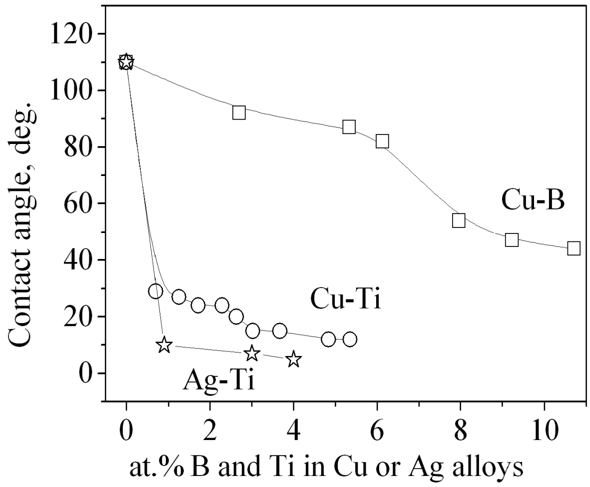

Figure 1. The final contact angle in the B_4C/(Cu-B), B_4C/(Cu-Ti) and B_4C/(Ag-Ti) systems after 30 min at 1423K (Aizenshtein et al., 2005a; Froumin et al., 2003)

2. Experimental Procedure

Two parts of B_4C (15×5×5 mm^3 and 5×5×5 mm^3) were cut from a block (99.8 % of the theoretical density). The surfaces of the samples were polished down to 1 μm using a SiC paper and diamond paste and cleaned ultrasonically in ethanol. Three types of brazing alloys were used in this study: Cu-9at.% B, Cu-4.5at.% Ti and the commercial Ticusil®. The Cu-9at.% B and Cu-4.5at.% Ti alloys were prepared in an arc furnace and rolled down to a final thickness of 150 μm. The Ticusil® brazing alloy was received in the form of a rolled plate with a thickness of 250 μm. The rolled sheet was cut to 5×5 mm^2 squares and placed between B_4C specimens. Brazing procedure was performed in a vacuum furnace under $2 \cdot 10^{-3}$Pa. Brazed samples were heated to approximately 50°C above the melting temperature of the brazing alloys, at the heating rate of 10°C/min and held for 30 minutes at the set point temperature. The cooling rate was about 30°C/min.

The maximum load value for joints brakeage was determined using an Instron LRX Plus instrument with the strain rate of 2 mm/min. Four samples for each brazing alloys were tested.

The microstructure and the composition of the interfaces were examined using optical microscopy and scanning electron microscopy (SEM) equipped with Energy Dispersive (EDS) and Wavelength Dispersive (WDS) Spectrometers.

3. Results and Discussion

3.1 Interface Characterization of the B_4C Joints

General view of the specimens after brazing and their SEM (SE and BSE) images are presented in Figures 2-5. For Cu-9at.%B joined B_4C, the interface of the ceramic specimens remains flat and no evidence of new phases was detected (Figure 3). This observation is in a good agreement with the previously reported results on the wetting behavior in the B_4C/Cu-B system (Froumin et al., 2003).

The structure of the B_4C joint obtained by using Cu-Ti filler (Figure 4) indicates the formation of small craters, which reflect boron carbide dissolution in the melt. According to WDS analysis, the continuous layer (up to ~ 5μm thick) is TiB_2 which was formed as a result of the interaction between dissolved B and Ti in the melt. Yet, this layer is detached from the B_4C/Cu-Ti interface. These observations are similar to that reported for interface interactions in the B_4C/Cu-Ti system (Aizenshtein et al., 2005a).

For the Ticusil® filler the dissolution of B_4C in the melt was not observed and only a thin (less than 1 μm) continuous Ti-rich layer was formed at the interface (Figure 5).

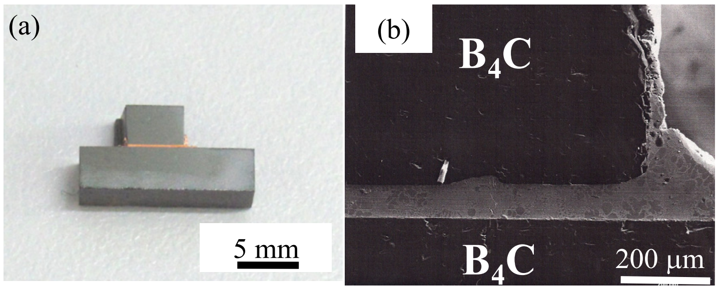

Figure 2. B_4C after brazing (a) macroscopic image Cu-4.5at.%Ti brazed B_4C (b) SE image, Ticusil® brazed B_4C

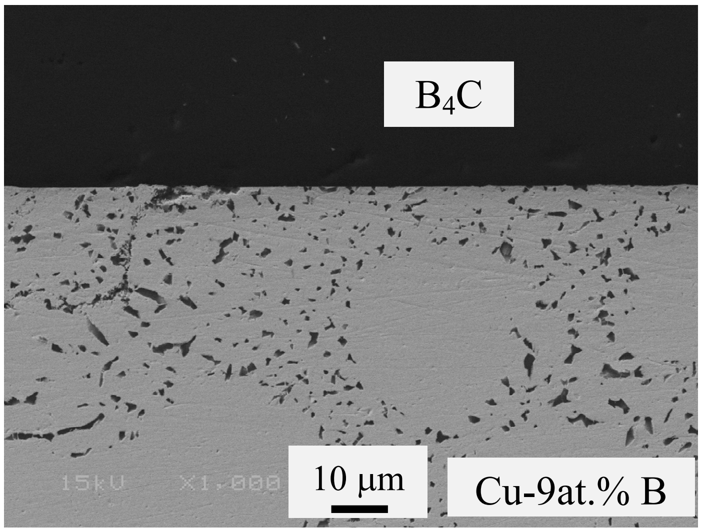

Figure 3. BSE image of B_4C specimens joined by Cu-9at.%B filler alloy

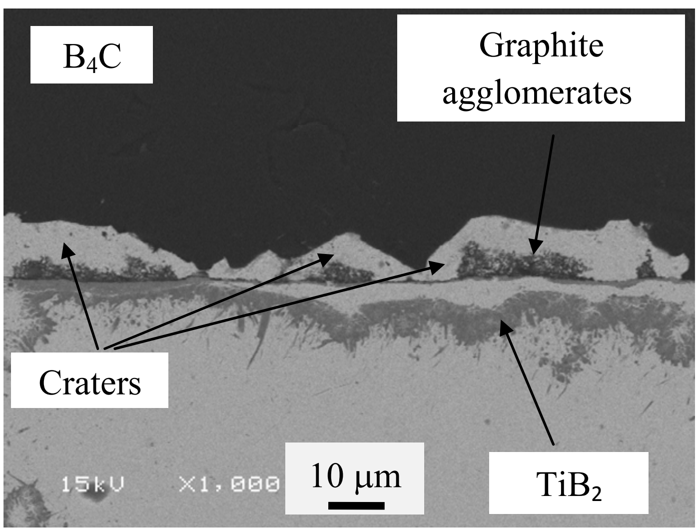

Figure 4. BSE image of B_4C specimens joined by Cu-4.5at.%Ti filler alloy

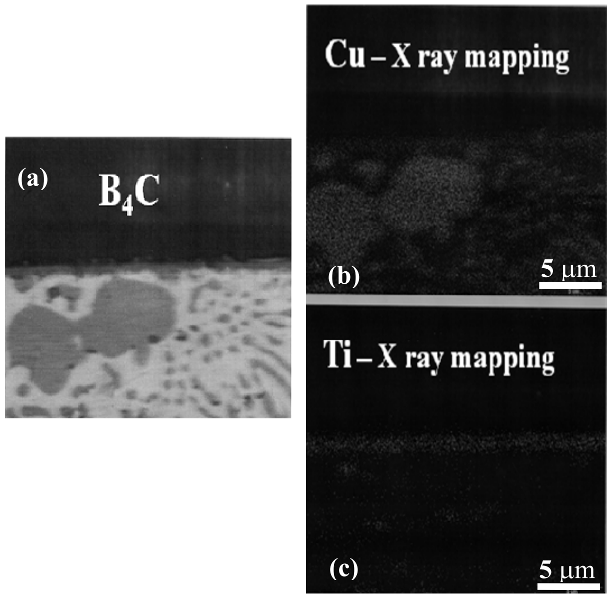

Figure 5. The morphology and spatial distribution of Cu and Ti at the B_4C/ Ticusil® interface (a) BSE image, (b) Cu X-ray mapping, (c) Ti X-ray mapping

According to the WDS line scan across the interface (Figure 6), the composition of this layer, close to the ceramic could be attributed to the TiB_2 phase, while TiC sub-layer is in contact with the melt. The same feature was reported for the B_4C/Ag-Ti system (Aizenshtein et al., 2005a). The formation of such double layer structure at the ceramic-filler interface was also observed in other systems (see for instance Mandal et al., 2004; Kar et al., 2007; Kar & Ray, 2007).

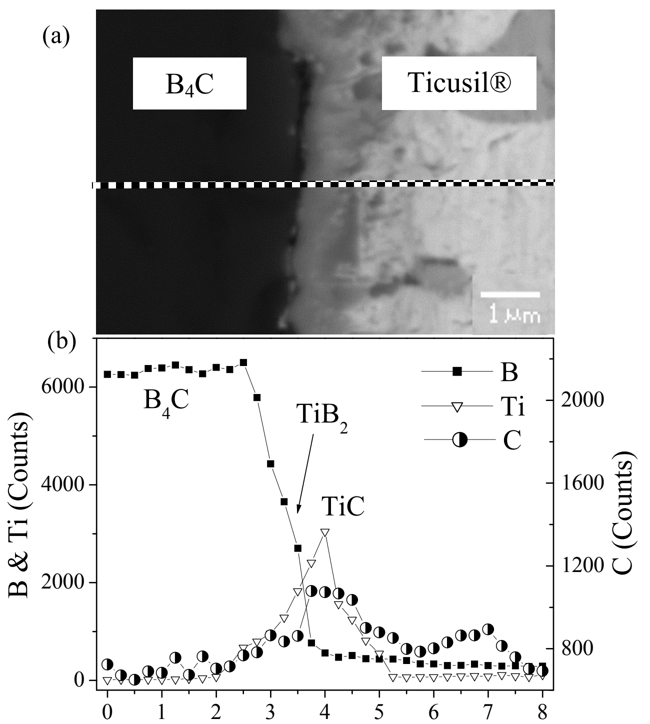

Figure 6. Microstructure and elements distribution at the B_4C/Ticusil® interface (a) BSE image of the scanned area (b) WDS line scans of B, Ti and C across the interface

We suggest that the differences between interface morphology in the B_4C/Cu-Ti and B_4C/Ticusil® systems reflect the thermodynamic properties of the fillers. The solubility of carbon in the liquid metals (Cu and Ag) is extremely low (Oden & Gokcen, 1992; Karakaya & Thompson, 1988). The solubility of B in liquid Cu at 1070^0C is about 20at.%, while this value for Ag is negligible (Chakrabarti & Laughlin, 1982; Okamoto, 1992). Thus, the dissolution of boron carbide in liquid Cu leads to boron transfer to the melt, which reacts with Ti and forms TiB_2, and free carbon precipitation. The dissolution of boron carbide in liquid Ag is very limited. Thus, we assume that the interface morphology in the B_4C/Ticusil® systems reflects the effect of Ag on the interface interaction. This assumption could be supported by thermodynamic considerations.

The boron activity in liquid Ag-Cu (Figure 7) was calculated using the Redlich-Kister approach. The interaction parameter, L^0, of the Ag-B and Cu-B liquid solutions was estimated using the binary phase diagrams (Okamoto et al., 1992; Froumin et al., 2003). At 1423K, the values of L^0 of the Cu-B and Ag-B systems are 20 kJ/mol and 90 kJ/mol respectively. The interaction parameters, L^0 and L^1, of the Ag-Cu liquid system were reported by Hayes, Lukas, Effenberg and Petzow (1986). Boron activity in Ag-Cu alloys is significantly higher than that in pure liquid Cu and provides the conditions for the formation of a continuous TiB_2 layer, attached to the ceramic, which was clearly observed in the case of B_4C/Ticusil® system (Figure 6). The formation of the double layered morphology is attributed to the Ti interaction with released free carbon resulting in the formation of the titanium carbide phase as was explained by Aizenshtein et al. (2005a).

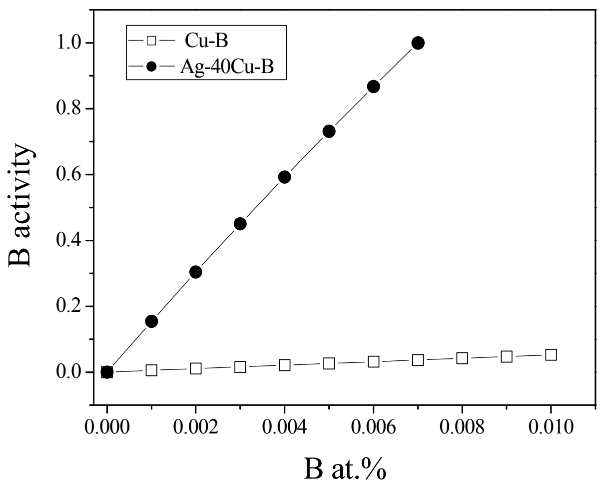

Figure 7. Boron activity in Cu and Ag-40at.% Cu liquid solutions as a function of B content. The B activity was calculated using Reldlich-Kister approach

3.2 Joint Strength and its Relation to Microstructure of the Interface

The mechanical properties strongly depend on the interface structure and residual stresses developed at the interface due to differences of thermal expansion coefficients (CTE). The measured shear forces and stresses for joints brakeage are presented in Table 1. The results obtained for Cu-9at.% B are in the range of the results (131MPa) reported for Fe-Ni-Co/Al_2O_3 joints brazed with Ticusil® (Do Nascimento et al., 1999).

Table 1. Measured shear force for different joints

Brazing alloys	Shear force, kN	Calculated shear stress [MPa]
Ticusil®	0.59 ± 0.15	23.6 ± 6.0
Cu 4.5 at.%Ti	0.76 ± 0.04	30.4 ± 1.6
Cu 9at.%B	2.44 ± 0.27	97.6 ± 10.8

Failure of the B_4C/Cu-9at%B joints take place due to a crack propagation in boron carbide (Figure 8a), while failure of the B_4C/Ticusil® and B_4C/Cu-4.5at%Ti joints occur through the brazing alloy (Figure 8b). The fracture path through the boron carbide body reflects a strong bonding at the metal/ceramic interface and the fact that the boron carbide composition at the interface is modified during the brazing process.

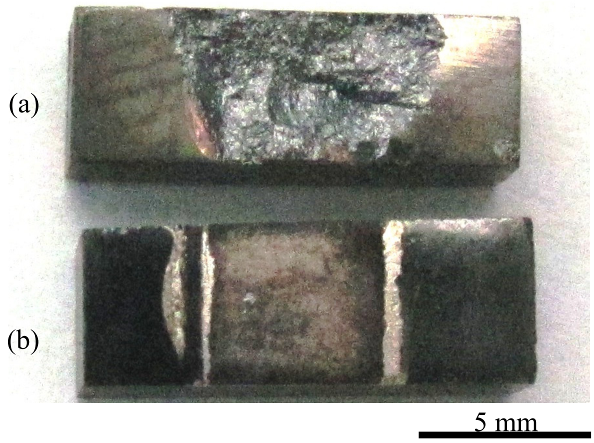

Figure 8. B_4C joints after shear force measurements (a) Cu-9at.%B brazing alloy (b) Ticusil® brazing alloy

In the two other cases the interface is weaker. In the case of B_4C/Ticusil®, the weakening of the interface is directly correlated to the formation of the new brittle phases TiB_2 and TiC. In the case of B_4C/Cu-4.5at%Ti, the brittle TiB_2 phase was detached from the interface and the weak interface is a result of the poor bonding between B_4C and Cu.

In this study it is demonstrated that improved wetting properties are essential, but not sufficient in order to guarantee adequate bonding. In fact, the contact angle in the B_4C/Cu-B is the highest compared to B_4C/(Cu, Ag)-Ti systems (Figure 1), yet better joint strength was obtained in the first case.

4. Conclusion

Joining of boron carbide specimens by Cu based fillers (Cu-9at%B, Cu-4.5at%Ti and Ticusil®) was studied. These alloys wet B_4C well ($\theta < 50^0$), whereas the interface composition and morphology are different. In the B_4C/Cu-9at.%B system no new phases were detected at the interface, while the formation of new brittle phases takes place in the B_4C/Cu-4.5at%Ti and B_4C/Ticusil® systems. The maximal strength of the joint close to 100MPa was obtained for the Cu-9at%B filler which could be correlates with the interface composition and structure. This study shows that improved wetting is essential but not sufficient in order to obtain strong bonding between B_4C and the fillers examined.

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