Joining of tungsten carbide to nickel by direct diffusion bonding and using a Cu–Zn alloy

José Lemus-Ruiz · Leonel Ceja-Cárdenas · J. A. Verduzco · Osvaldo Flores

Received: 3 August 2007/Accepted: 22 July 2008/Published online: 13 August 2008 © Springer Science+Business Media, LLC 2008

Abstract The objective of this work was to study various aspects of liquid and solid state diffusion bonding of cylindrical samples of WC (with 6% Co) and commercially pure nickel (99.5%) produced by direct bonding and brazing using a 25 µm thick 70Cu 30Zn (wt%) alloy as joining element. Joining experiments were carried out on WC/Ni and WC/Cu Zn/Ni combinations at temperature of 980 °C using 1, 15, 25 and 35 min holding times in argon (Ar). The results show that it is possible to create a successful joint at temperature and times used. Joining occurred by the formation of a diffusion zone. The joining interface is feasible because it presents a homogeneous interface with no several interfacial cracking and porosity. In both combinations, it can be observed a diffusion of cobalt decreasing in the direction of the metal, as well as, the diffusion of nickel decreasing in the direction of the ceramic.

Introduction

The use of ceramics and ceramics based composites in industrial applications, mainly for high temperature due to their good corrosion resistance and strength at elevated

O. Flores

temperatures, has received extensive attention recently. Cemented tungsten carbide is one of the most important materials used as cutting tools, wear parts and, as replacement of standard materials for tools, dies and machine components, due to its hardness and abrasion resistance combined with good high temperature mechanical properties [1]. However, in most applications, ceramic materials are used in combination with metals, and this has generated a continued interest in the use of joining technologies to produce complex configurations from assemblies of simple shapes [2]. Various techniques for joining ceramic to metal are available, some need an intermediate liquid phase, brazing, and others are produced by solid state diffusion bonding [3, 4]. One widely used method for joining ceramics consists of brazing with a reactive metal alloy, however, the highest obstacle of successful brazing of ceramics to metals is the fact that most conventional brazing materials, in general, do not wet ceramic surfaces [5, 6].

In diffusion bonding, diffusion or chemical reaction takes place between the metal and the ceramics, and the properties of the reaction product layer dictate the usefulness of the bond and thus of the whole assembly at high temperature. The reaction product layer should provide a strong bond between the two dissimilar materials, which means: i) it should accommodate for the thermo-mechanical mismatch resulting from differences in thermal expansion coefficients, and ii) the reaction product layer should not consist of compounds that have mechanical properties significantly inferior to those of the metal and ceramic [7, 8]. All joining techniques must take into account the difference in coefficient of thermal expansion, CTE, between the metal and ceramic. The misfit in the CTE of the joining materials can result in areas of high residual stresses at the interface during the cooling process [9, 10]. Ceramic-metal interfaces are a critical feature in

J. Lemus-Ruiz (⊠) · L. Ceja-Cárdenas · J. A. Verduzco Instituto de Investigaciones Metalúrgicas, Universidad Michoacana de San Nicolás de Hidalgo, Apdo. Postal 888, C.P. 58000 Morelia, Michoacan, Mexico e-mail: jlruiz@umich.mx

Instituto de Ciencias Físicas, Universidad Nacional Autónoma de México, Av. Universidad S/N, Col. Chamilpa, C.P. 62251 Cuernavaca, Morelos, Mexico

many systems, and processing of these interfaces is fundamental to fabrication of a wide range of materials and devices. The practical use of advanced ceramics depends on the reliability of ceramic/metal joining techniques and the properties of the resulting interfaces [11, 12].

Several problems have been associated with the joint strength influencing the reliability of a joint [13]. From a microscopic view, the reaction microstructure caused by wetting or by chemical and physical bond-ability between two faces may be of concern. These factors will reflect the distribution of unjoined or weakly bonded island-like defects on interfaces resulting in substantial reduction in joint strength. On the other hand, from the more macroscopic view, when a reaction layer grows, cracking occurs in the layer, which frequently influences joint strength. The final goal for joining research will be in establishing a technique producing a tightly bound interface by eliminating these defects and by accommodating thermal stress. These residual stresses reduce the strength of the bonded material and in some cases lead to joint failure during or after the joining process [14-16]. Therefore, in order to understand the mechanical performance of joints, it is important to understand the mechanisms of interface formation between the metal and ceramic. This research is focused on the fabrication and characterization of WC/Ni and WC/Cu Zn/Ni combinations by solid and liquid state diffusion bonding, respectively.

Experimental procedures

The materials used in this work were WC/6Co (wt%) (Goodfellow, England), commercially pure Ni (99.5%) supplied in rod shape, and commercially Cu70 Zn30 brass foil (Johnson Matthew, USA). The diffusion bonding experiments started with the preparation of the materials to be joined. The original rods of 25 mm length and 6.35 mm diameter were cut into small blocks of cylindrical geometry having thickness of 3 mm. The success of diffusion bonding processes depends on a combination of factors: one of the most important is the surface roughness of the materials because it controls the initial contact area between the diffusion couples. In order to assure reproducibility of the surface preparation of the samples, a polishing procedure was established. The surfaces were ground using a diamond-grinding disc, followed by silicon carbide paper of 600 and 1000 grit. Subsequently, polishing was carried out using diamond paste (1 µm) and alumina suspensions of 0.3 and 0.05 µm. Before the joining experiments, the samples were cleaned with isopropanol in an ultrasound bath for 5 min.

Dissimilar joints combination, WC/Ni and WC/Cu Zn/Ni, were mounted axially such that their polished



Fig. 1 Schematic representation of the sample assembly and furnace chamber

surfaces were in contact. The specimens to be joined were placed in a graphite die embedded in a boron nitride (99.5% pure) powder bed; this die consists of a screw that on turning clockwise can impart pressure to the sandwich assembly in order to prevent movements of the pieces by keeping the joints in contact. The purpose of the powder bed was to avoid contact and reaction between the sample and the internal walls of the die. The experimental apparatus used to join the sample combinations consists of a resistance furnace with one size closed-end alumina-tube chamber of 80 cm long and 10 cm in diameter shown in Fig. 1. The sample assembly was positioned in the furnace and this was filled with argon. Next, the furnace was heated up to the preset joining temperature of 980 °C using 1, 15, 25 and 35 min holding times. Microstructural examination was performed on polished cross-sections of the experimental couples using scanning electron microscopy and micro analysis.

Results and discussion

The experimental results show a successful joining, achieved at 980 °C for the different bonding times for both combinations, WC/Ni and WC/Cu Zn/Ni. Joints of WC/Ni samples are formed through the formation of a diffusion interface on the metal side of the sample as a result of solid state diffusion of Co to Ni, as well as diffusion of Ni to the cermet WC/Co. On the other hand, liquid formation occurs during joining of WC/Cu Zn/Ni at 980 °C (melting point of Cu–Zn \sim 950 °C) and joining takes place by liquid state diffusion of Co and Ni and interaction of these species with Cu and Zn of the liquid Cu Zn alloy. Figure 2 shows a cross-section of the interface obtained for a sample of WC/Cu Zn/Ni bonded at 980 °C for 1 min. It can be seen a continuous and homogeneous diffusion interface on the metal side free of thermal cracks, however un-joined islands are observed on the bonding line with the metal Ni. On the other hand, Fig. 3 shows a cross-section of the interface observed in brazing samples produced at (a) 980 °C for 25 min and (b) 980 °C for 35 min, it can be observed that increasing the bonding time the diffusion



Fig. 2 Cross section of the interface obtained in a WC/Cu Zn/Ni sample bonded at 980 $^{\circ}\mathrm{C}$ for 1 min



Fig. 3 Cross section of the interface observed in a WC/Cu Zn/Ni sample produced at 980 °C for (a) 25 min and (b) 35 min

increases forming a continuous bonding layer free of porosity. Electron probe micro analysis performed on these samples indicated that Ni, Co, Cu, and Zn are in the diffusion interface, however no phases were detected and the components are in solution in the bonding interface. According to the thermodynamic, WC is stable at the bonding temperature; therefore W and C interactions with Ni or Cu Zn are not expected in the joining zone. Diffusion is the dominating reaction mechanism in diffusion joining, consequently, the high affinity of Co and Ni for Cu and Zn resulted in immediate diffusion and phase transformations may or may not be observed depending on the joining parameters, such as bonding temperature and time, because these parameters affect the concentration of diffusion of the components at the interface, and therefore, the nature of the resulting interface. The thickness of these interfaces increases when the bonding time increases. The average thickness was 36 µm for samples joined at 980 °C for 25 min and 37 µm for samples joined at 980 °C for 35 min. The driving force for the formation of an interface between materials is the free energy decrease of the system resulting from joining. Since joining of WC/Cu Zn/Ni occurred at relatively high temperature and liquid formation occurred during the process, this promoted an interfacial interactions and bonding between the materials.

A qualitative overview of the different components across the WC/Cu Zn/Ni interface was studied using atomic distributions for a sample joined at 980 °C for 15 min. The results are illustrated in Fig. 4, where the interface is aligned in the vertical direction with the WC on the left and Ni on the right side. It can be observed clearly that the average thickness of the interface was 33 µm. The main elements analyzed were Cu, Zn, Co, Ni, and W. The different contrast from dark to white corresponds to the increase in the concentration of the specific element. In the Co-map, the different contrast from left to right corresponds to the decrease in concentration of Co from de WC. Diffusion of Co into the interface can be observed. For the Ni-map, a decrease in the intensity corresponding to a decrease in the concentration of Ni observed in the direction of WC, passing through the interface clearly demarking the extent of Ni diffusion. For the Cu-map, the concentration decreased close to the Ni and ceramic interface clearly delineating the extent of Cu diffusion. For the Zn-map, homogeneous distribution can be observed across the bonding interface. No diffusion of W into the interface can be observed. These results confirmed that, Co, Ni, and Cu were the main diffusing elements into the interface. On the other hand, an overview of the different components in the interface was obtained in a WC/ZN Cu/Ni sample joined at 980 °C for 15 min by line analysis using electron probe micro analysis. The results are illustrated in Fig. 5 where the WC and Ni are on the left and right, respectively. The scan line was chosen to start on the WC side of the sample through the interface, Cu Zn, finishing on the Ni side. The Ni signal reached its maximum at the Ni Cu braze. Interdiffusion of Cu Ni and Cu Co



Fig. 4 Qualitative analysis by atomic distributions across the interface obtained in a sample joined at 980 $^{\circ}$ C for 15 min



Fig. 5 Line analysis through the interface obtained in a WC/Cu Zn/ Ni sample joined at 980 $^{\circ}\text{C}$ for 15 min

could be observed. The micro analysis profile indicates the presence and even distribution of Zn concentration. In the region corresponding to diffusion zone, high levels of Cu and Zn were observed.

In contrast, it is important to point out that because no good joints were obtained on WC/Ni samples for times shorter than 25 min, therefore no further characterization was performed on such samples; however, for times of 25 and 35 min, successfully bondings were attained in a solid state process. A cross-section of WC/Ni interface of samples joined at 980 °C for 35 min is shown in Fig. 6. A continuous and homogeneous bonding line interface can be seen. The bonding process is governed by diffusion of Ni through the Co of the WC and diffusion of Co through Ni, forming a thin interdiffusion zone. Figure 7 shows an



Fig. 6 Cross section of the interface observed in a WC/Ni sample joined at 980 $^{\circ}$ C for 35 min



Fig. 7 Line analysis through the interface obtained in a WC/Ni sample joined at 980 $^\circ$ C for 25 min

overview of the Co, W, and Ni components through the interface obtained in a WC/Ni sample joined at 980 °C for 25 min by line analysis using electron probe micro analysis. WC and Ni are on the left and right, respectively, starting the scan line on the WC side of the sample and finishing on the Ni side. It can be clearly observed from the micro analysis profiles of Ni and Co that the accumulation of these components in the bonding line with interdiffusion of Ni to Co and Co to Ni. In the present work, the joining temperature and time were the main parameters studied. The first step in creating an interface is to achieve intimate contact, and subsequently bonding can occur. In this sense, it has been reported [17] that the effect of a reaction layer on the interface strength depends on a number of factors such as the mechanical properties of the reaction layer, its thickness and morphology; therefore the choice of suitable conditions to prepare ceramic/metal joints requires knowledge concerning the mechanism of reaction between the materials and the evolution of the interface.

Conclusions

- It is possible to join tungsten carbide to nickel by direct diffusion bonding and brazing using a Cu Zn interlayer as joining element.
- Successful bonding was observed at a temperature of 980 °C on WC/Ni and WC/Cu Zn/Ni sample combinations.
- Joining of tungsten carbide to nickel occurred by the formation of a homogenous diffusion interface on the metal side of the joint.
- 4) Thickness of the diffusion layer increases when bonding time increases, and the diffusion zone in the WC/Cu Zn/Ni samples has a larger width than that of WC/Ni samples combinations.

Acknowledgements The authors would like to thank to CONA-CYT-México and Universidad Michoacana de San Nicolás de Hidalgo (UMSNH) for the financial support and facilities of this research.

References

- 1. Fernie JA, Sturgeon AJ (1992) Bonding & joining. Joining Ceramic Materials, USA
- 2. Nicholas MG (1998) Joining processes: introduction to brazing and diffusion bonding
- 3. Tomsia AP (1993) J Phys IV 3:1317. doi:10.1051/jp4:19937203
- 4. Tinsley ND, Huddleston J, Lacey MR (1998) Mater Manuf Process 13:491
- Janickovic D, Sebo P, Duhaj P, Svec P (2001) Mater Sci Eng A 304–306:569. doi:10.1016/S0921-5093(00)01536-7
- Carim AH, Mohr CH (1997) Mater Lett 33:195. doi:10.1016/ S0167-577X(97)00098-0
- Wang L, Aldinger F (2002) Mater Lett 54:93. doi:10.1016/ S0167-577X(01)00536-5
- Heikimheimo E, Isomaki I, Kodentsov AA, Van Loo FJJ (1997) J Eur Ceram Soc 17:25. doi:10.1016/S0955-2219(96)00082-9
- Locatelli MR, Dalgleish BJ, Nakashima K, Tomsia AP, Glaeser AM (1997) Ceram Int 23:313. doi:10.1016/S0272-8842(96)00024-7
- Marks RA, Chapman DR, Danielson DT, Glaeser AM (2000) Acta Mater 48:4425. doi:10.1016/S1359-6454(00)00229-9
- 11. Lemus J, Drew RAL (2003) Mater Sci Eng A352-306:169
- Jadoon AK, Ralph B, Hornsby PR (2004) J Mater Proc Tech 152:257. doi:10.1016/j.jmatprotec.2003.10.005
- 13. Suganuma K (1993) Mater Res Soc Sym Proc 314:51
- 14. Anderson RM (1989) Adv Mater Proc 3:31
- Richerson DW (1992) Modern ceramic engineering, 2nd edn. Marcel Decker, New York
- Osendi MI, De Pablos A, Miranzo P (2001) Mater Sci Eng A 308:53. doi:10.1016/S0921-5093(00)02027-X
- Lemus-Ruiz J, León-Patiño CA, Drew RAL (2006) Metall Mater Trans A 37A:69. doi:10.1007/s11661-006-0153-4