

Interfacial Reactions and Joint Strengthening Mechanisms in Pulsed Current-Assisted Liquid Phase Diffusion Bonding of Ceramic Matrix Composites (Postprint)

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Abstract

Liquid phase diffusion bonding experiments were conducted on Ti(C, N)-Al₂O₃ ceramic matrix composites using Cu-Zr foil/Cu foil/Cu-Zr foil interlayers to investigate the effects of auxiliary pulsed current on element diffusion, interfacial reaction products, and joint strengthening mechanisms. The results indicate that under auxiliary pulsed current conditions during liquid phase diffusion bonding, higher joint strength can be achieved at lower bonding temperatures and with shorter bonding times. The auxiliary pulsed current liquid phase diffusion bonding process significantly alters the diffusion behavior of Zr and Cu in the Ti(C, N)-Al₂O₃ ceramic matrix composites and the brazing seam, reducing Zr activity and inhibiting its vigorous chemical reaction with Al₂O₃ ceramic particles. Additionally, auxiliary pulsed current can suppress the dissolution of ceramic particle phases into the weld seam and reduce the thickness of both the interfacial diffusion transition layer and the Zr-Cu reaction layer, thereby ensuring weld seam strengthening and interfacial strengthening. This is the key to achieving the high strength levels observed in joints produced by auxiliary pulsed current liquid phase diffusion bonding.

Full Text

Preamble

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Ceramic Matrix Composite Joints Using Liquid Phase Diffusion Bonding with Auxiliary Pulse Current: Interfacial Reaction and Strengthening Mechanism

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Abstract

Ceramic matrix composites (CMCs) have attracted significant interest in aerospace and nuclear engineering applications due to their excellent high-temperature resistance, corrosion resistance, and wear resistance. However, their practical usage is limited by challenges in joining CMCs with metals, which exhibit obvious physical and chemical incompatibility. This work investigates liquid phase diffusion bonding (LPDB) of Ti(C, N)-Al₂O₃ CMC to Cu using a Cu-Zr foil/Cu foil/Cu-Zr foil sandwich interlayer, with auxiliary pulse current applied to control elemental diffusion and interfacial reaction during the bonding process. The element diffusion and reaction products at the interface were analyzed using SEM, EPMA, and EDS, and joint strength was evaluated using four-point bending tests. The results demonstrate that auxiliary pulse current during LPDB achieves higher joint strength at lower bonding temperatures and shorter holding times. The diffusion behavior of Zr and Cu elements in the CMC and interfacial region is significantly altered, and the activity of Zr element and its chemical reaction with Al₂O₃ are suppressed by the auxiliary pulse current. The diffusion of ceramic particles into the weld seam and the thickness of both the diffusion transition zone (DTZ) and Zr-Cu interfacial reaction zone (IRZ) are also suppressed, which strengthens both the weld seam and the interface and maintains higher joint strength.

KEY WORDS Ti(C, N)-Al₂O₃, liquid phase diffusion bonding, auxiliary pulse current, interfacial reaction, joint strengthening

Introduction

Ceramic materials exhibit excellent high-temperature resistance, corrosion resistance, and wear resistance, making them promising for applications in aerospace,

aviation, and nuclear energy fields. However, their intrinsic brittleness makes it difficult to fabricate complex structural components, limiting their widespread application. Typically, they must be joined with other metals to form composite structures that can fully utilize their unique properties. When joining ceramics with dissimilar materials, particularly metals, their brittle nature often results in significant residual stresses in the welded joints, severely weakening the load-bearing capacity and even causing cracking in the ceramic during the joining process. Currently, numerous studies have been reported on ceramic-metal joining, with methods primarily involving active metal brazing [1-3], transient liquid phase diffusion bonding [4-6], solid-state diffusion bonding [7-9], and self-propagating high-temperature synthesis [10-12]. Although researchers have conducted extensive work in this area, further investigation is still needed regarding the theoretical understanding of interfacial reactions, accuracy of residual stress analysis, reliability of joint performance evaluation, and practicality of joining processes [13].

Ti(C, N)-Al₂O₃ ceramic matrix composites are multiphase composites composed primarily of Ti(C, N) and Al₂O₃ ceramic particle hard phases, along with Ni, Mo, Co, and Mo₂C binder phases. These materials exhibit high hardness and wear resistance while maintaining good oxidation resistance and chemical stability, showing broad application prospects as cutting tool materials and high-temperature structural materials. Like monolithic ceramics, the biggest bottleneck for their practical application remains the reliability of welded joints with metallic materials. Currently, few studies have been reported on joining ceramic matrix composites with metals. Drawing on relevant research results from ceramic-metal joining [14-16], this work proposes an auxiliary pulse current liquid phase diffusion bonding method for Ti(C, N)-Al₂O₃ ceramic matrix composites, aiming to control the interfacial reaction process and elemental diffusion behavior through pulse current, reduce the joining temperature to alleviate peak residual stresses in the welded joints, and thereby improve and enhance joint performance.

Experimental Procedures

The base material used in this study was Ti(C, N)-Al₂O₃ ceramic matrix composite, with a flexural strength of 850 MPa, hardness of 94.7 HRA, and coefficient of thermal expansion of $9 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$. The original dimensions were 19 mm \times 19 mm \times 20 mm, which were cut into cylindrical rods with a diameter of 8.5 mm and height of 20 mm using wire electrical discharge machining.

Regarding the interlayer materials, their purpose was to achieve metallurgical bonding with the Ti(C, N)-Al₂O₃ ceramic matrix composite and reduce residual stresses. The authors previously investigated Ti-containing alloy foils [17,18] and found that Ti was overly active, reacting violently with the Ti(C, N)-Al₂O₃ ceramic matrix composite, making the interfacial reaction process difficult to control and resulting in relatively low joint strength. In this work, the interlayer filler material selected was amorphous 53Cu-47Zr alloy foil (mass fraction, %)

with a thickness of 50 μm . Zr is less active compared to Ti, and a 500 μm thick Cu foil was used as a stress-relief interlayer.

All materials were ground with metallographic sandpaper and ultrasonically cleaned in acetone before welding. The joint was assembled in a Ti(C, N)-Al₂O₃/53Cu-47Zr alloy foil/Cu foil/53Cu-47Zr alloy foil/Ti(C, N)-Al₂O₃ butt joint configuration and placed in a heating furnace for auxiliary pulse current liquid phase diffusion bonding experiments. The vacuum level was not lower than 13 Pa, with an axial pressure load of 1 MPa applied to the sample. The DC pulse duty cycle (ON/OFF) was 12/2, the heating rate was 100 °C/min, and the samples were furnace-cooled after welding. The bonding temperatures were 850 and 900 °C, with holding times of 3–10 min. For comparative study, conventional brazing experiments were conducted in a vacuum furnace with the same joint configuration at a vacuum level not lower than 1×10^{-2} Pa. The process parameters were: heating temperature 980 °C, holding time 5–30 min, axial pressure load 1 MPa, heating rate not exceeding 3 °C/min, and cooling rate not exceeding 1 °C/min after welding.

After welding, samples were sectioned using wire electrical discharge machining for microstructural analysis and four-point bending test specimens. Microstructural analysis samples measured 8 mm \times 8 mm \times 3 mm and were etched with HCl-75% HNO₃ solution (volume fraction) to reveal the microstructure. Four-point bending test bars measured 3 mm \times 4 mm \times 40 mm. Joint strength testing was conducted according to the GB6569-86 standard for engineering ceramic flexural strength testing methods. The microstructure, elemental distribution, and fracture analysis of the welded joints were performed using a JSM6480 scanning electron microscope (SEM), JXA8100 electron probe micro-analyzer (EPMA), and energy dispersive spectroscopy (EDS).

2.1 Element Diffusion and Reaction Mechanism

Figure 1 [Figure 1: see original paper] shows the microstructure and EPMA results of Ti(C, N)-Al₂O₃/53Cu-47Zr alloy foil/Cu foil/53Cu-47Zr alloy foil/Ti(C, N)-Al₂O₃ joints bonded with auxiliary pulse current at 900 °C for 3 and 10 min under an axial pressure load of 1 MPa. Table 1 presents the EDS composition analysis of characteristic points. As shown in Figure 1a, for auxiliary pulse current liquid phase diffusion bonding, despite a holding time of only 3 min, a continuous and dense reaction layer has formed at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface with a thickness of approximately 1–2 μm , hereafter referred to as the interfacial reaction zone (IRZ). After melting of the 53Cu-47Zr alloy foil, Zr is significantly enriched at the interface and has diffused into the Ti(C, N)-Al₂O₃ ceramic matrix composite to a depth of 2–3 μm , hereafter referred to as the diffusion transition zone (DTZ). In this region, the Al content is very low while the Ti content remains essentially unchanged, indicating that Zr diffused into the Ti(C, N)-Al₂O₃ ceramic matrix composite and chemically reacted with Al₂O₃ ceramic particles. In the Zr-enriched region at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface, the Cu content from

the melted 53Cu-47Zr alloy foil decreases sharply, but increases significantly after entering the Ti(C, N)-Al₂O₃ ceramic matrix composite. Dissolution of Ti(C, N) and Al₂O₃ ceramic particle phases into the weld seam is essentially not observed. Figure 1b shows the effect of extended holding time on elemental diffusion and interfacial reaction. The Zr enrichment at the interface remains essentially unchanged, while the diffusion depth into the Ti(C, N)-Al₂O₃ ceramic matrix composite increases slightly to 4–5 μm. The thickness of the interfacial reaction layer at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface also increases slightly to approximately 3–4 μm, while the diffusion behavior of Cu remains essentially unchanged. Notably, with increased holding time, the distribution of Zr in the weld seam becomes continuous, corresponding to significant fluctuations in Cu content.

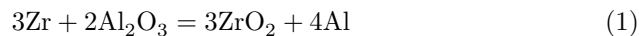
Figure 2 [Figure 2: see original paper] shows the microstructure and EPMA results of Ti(C, N)-Al₂O₃/53Cu-47Zr alloy foil/Cu foil/53Cu-47Zr alloy foil/Ti(C, N)-Al₂O₃ joints bonded without auxiliary pulse current at 980 °C for 10 and 30 min under an axial pressure load of 1 MPa. The EDS composition analysis of characteristic points is presented in Table 1. Comparing Figures 1 and 2 reveals that, aside from the common phenomenon of Zr enrichment at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface, other aspects of elemental diffusion are significantly different. For conventional brazing, the diffusion depth of Zr into the Ti(C, N)-Al₂O₃ ceramic matrix composite increases significantly (with almost no Al detected in this region), reaching 30–35 μm. The distribution pattern of Zr in the weld seam does not change substantially with increased holding time, appearing as discontinuous peak-like features. Under the same holding time conditions, the interfacial reaction layer thickness is significantly greater than that in auxiliary pulse current liquid phase diffusion bonding. The diffusion trend of Cu element also changes, with its content decreasing sharply in the Zr-enriched region at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface and very little diffusion into the Ti(C, N)-Al₂O₃ ceramic matrix composite, being almost undetectable in EPMA elemental line scan profiles.

These experimental results demonstrate that compared with conventional brazing, auxiliary pulse current liquid phase diffusion bonding significantly changes the elemental diffusion rate, interfacial reaction process, and dissolution behavior of Ti(C, N)-Al₂O₃ ceramic matrix composite particles: (1) the diffusion migration rate of active element Zr into the Ti(C, N)-Al₂O₃ ceramic matrix composite is significantly reduced; (2) the chemical reaction process between active element Zr and Cu at the interface is suppressed; and (3) dissolution of Ti(C, N)-Al₂O₃ ceramic matrix composite particle phases into the weld seam during welding is significantly reduced. From this, it can be inferred that the application of auxiliary pulse current during the liquid phase diffusion bonding process is responsible for these phenomena, i.e., the auxiliary electric field formed by the pulse current in the bonding region alters the mass transfer process and chemical reaction activity between elements during liquid phase diffusion bonding.

As shown in Table 1, in the diffusion transition zone (characteristic points A

and D), for auxiliary pulse current liquid phase diffusion bonding, because Cu migrates in large quantities into this region and chemically reacts with active element Zr, the activity of Zr is reduced, effectively controlling the reaction process between Zr and Al_2O_3 ceramic particles. Therefore, Al can still be detected in this region. Under conventional brazing conditions, because only trace amounts of Cu diffuse into this region, active element Zr undergoes intense chemical reaction with Al_2O_3 ceramic particles, generating large amounts of free Al atoms that diffuse into the weld seam, resulting in drastically reduced Al content in the diffusion transition zone and significantly increased Al content in the weld seam. In the interfacial reaction zone (characteristic points B and E), the main reaction product is Zr-Cu intermetallic compound for both auxiliary pulse current liquid phase diffusion bonding and conventional brazing. Notably, under brazing conditions, dissolution of Ti(C, N)- Al_2O_3 ceramic matrix composite is evident, with measurable Ti content detected at the interface. In the weld seam region (characteristic points C and F), the main phase is Cu solid solution for both joining methods, with the only difference being that the brazed weld seam contains small amounts of Al, which confirms that during brazing, Zr underwent relatively complete chemical reaction with Al_2O_3 ceramic particles in the diffusion transition zone.

According to literature [19], Zr reacts with Al_2O_3 through the following chemical reaction:



Based on literature [20-22], the standard Gibbs free energy (ΔG_T) expression for the above chemical reaction is calculated as:

$$\Delta G_T = 69400.916 - 25.163T - 4.862 \times 10^{-3}T^2 - 27.28 \times 10^{-5}T^{-1} + 89.22 \quad (2)$$

where T is the bonding temperature. Substituting $T = 900$ °C into the equation yields $\Delta G_T = -43559.87$ J/mol for the reaction between Zr and Al_2O_3 . Therefore, from a thermodynamic perspective, reaction (1) is highly likely to occur.

To further compare the differences in elemental diffusion behavior between conventional heating and auxiliary pulse current heating, diffusion experiments were conducted on Zr/Cu diffusion couples under both conditions, with interfacial microstructure and elemental distribution shown in Figure 3 [Figure 3: see original paper]. The solid-state diffusion process parameters for both auxiliary pulse current heating and conventional heating were: temperature 870 °C, holding time 10 min. As shown in Figure 3, compared with conventional heating, the diffusion and reaction behavior of Zr and Cu change significantly under auxiliary pulse current heating. The diffusion distances of Zr into Cu and Cu into Zr are noticeably shortened, and the diffusion reaction zone (DRZ) composed of DTZ and IRZ becomes smaller, indicating that auxiliary pulse current suppresses the interdiffusion capability of Zr and Cu. EDS analysis shows that for auxiliary pulse current heating, the atomic fraction of Zr in the diffusion

reaction layer on the Cu side is generally below 10%, whereas for conventional heating, the atomic fraction of Zr exceeds 20%, which verifies the above conclusion and demonstrates that auxiliary pulse current can effectively control the chemical reaction between Zr and Cu to form intermetallic compounds. For conventional heating diffusion, Cu penetrates through the initial Zr/Cu diffusion couple interface, forming an enrichment zone at the Zr substrate interface with a thickness of several micrometers, whereas no such phenomenon occurs under auxiliary pulse current heating. Literature [23] uses phenomenological theory to analyze how electric fields can enhance intragranular solute solid solution capacity and hinder solute and vacancy segregation at grain boundaries, thereby strengthening grain boundaries. The method of using pulse current to improve solidification microstructure has attracted attention because the electromagnetic force generated by electric pulses in the molten metal refines the grain structure. Additionally, electric pulses can interfere with the normal migration patterns of metal atoms, thereby achieving grain refinement of the solidified microstructure [24,25]. The results of this study confirm that auxiliary pulse current significantly suppresses the diffusion migration and chemical reaction rates between metal atoms.

2.2 Joint Strengthening Mechanism

To compare the effectiveness of auxiliary pulse current liquid phase diffusion bonding versus conventional brazing for Ti(C, N)-Al₂O₃ ceramic matrix composites, four-point bending strength tests were conducted at room temperature, with results shown in Figure 4 [Figure 4: see original paper]. For auxiliary pulse current liquid phase diffusion bonding at a bonding temperature of 900 °C, the joint strength reaches a high level; even with a holding time of only 3 min, the average four-point bending joint strength already reaches 315.5 MPa, demonstrating that auxiliary pulse current liquid phase diffusion bonding can significantly improve welded joint strength compared with conventional brazing under appropriate heating temperature conditions.

Typical joint fracture paths for conventional brazing and auxiliary pulse current liquid phase diffusion bonding are shown schematically in Figure 5 [Figure 5: see original paper]. For conventional brazing, microcracks generally initiate and propagate in the Zr-enriched region of the weld seam during four-point bending, whereas for auxiliary pulse current liquid phase diffusion bonding, microcracks typically initiate at the Ti(C, N)-Al₂O₃ ceramic matrix composite interface and propagate unstably within the composite.

Based on the above analysis, the joint strength weakening mechanism in conventional brazing and the strengthening mechanism in auxiliary pulse current liquid phase diffusion bonding can be summarized as follows: During liquid phase diffusion bonding, the auxiliary pulse current alters the diffusion behavior of Cu and Zr in the liquid filler metal 53Cu-47Zr, suppresses the diffusion migration rate of Zr into the Ti(C, N)-Al₂O₃ ceramic matrix composite, and effectively controls the reaction amount between active element Zr and Al₂O₃

ceramic particles. This significantly reduces the dissolution of both Ti(C, N) and Al₂O₃ ceramic particles from the composite, decreases the Al content in the weld seam, and thereby strengthens the weld seam. Additionally, it reduces the activity of Zr, promotes uniform interaction between Zr and Cu elements in the weld seam, and controls the migration amount of Zr to the Ti(C, N)-Al₂O₃ ceramic matrix composite interface, thereby effectively reducing the thickness of the interfacial reaction layer and enhancing interfacial strength.

The key findings can be summarized as: (1) Auxiliary pulse current liquid phase diffusion bonding significantly changes the diffusion behavior of Zr and Cu in the Ti(C, N)-Al₂O₃ ceramic matrix composite and weld seam, reduces Zr activity, and plays an important role in suppressing intense chemical reactions between Zr and Al₂O₃ ceramic particles. (2) Controlling the dissolution of ceramic particle phases into the weld seam and the thickness of the interfacial Zr-Cu reaction layer ensures weld seam strengthening and interfacial strengthening, which is the key to achieving higher joint strength levels in auxiliary pulse current liquid phase diffusion bonding.

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