Effect of Interlayer Metal on Microstructure and Shear Strength of Al2O3/1Cr18Ni9Ti Brazed Joints (Postprint)

Authors: Liu Yi, Jiang Guofeng, Xu Kun, Luo Ximing, Chen Dengquan, Li Wei

Date: 2023-03-19T00:00:00+00:00

Abstract

Al2O3 ceramic and 1Cr18Ni9Ti stainless steel were joined using Ag-Cu-Ti active filler metal, and the effects of three interlayer metals–Cu, Ni, and Ni-plated Cu–on the microstructure and shear strength of the brazed joints were investigated. The results showed that when Cu was used as the interlayer, a good interface reaction could be formed between the ceramic and the filler metal; when Ni was used as the interlayer, a large amount of Ni3Ti intermetallic compounds formed in the weld seam, which prevented the formation of a good reaction layer between the ceramic and filler metal and reduced the shear strength of the joint. When Ni-plated Cu sheet was used as the interlayer metal, the presence of a small amount of Ni did not affect the content of the active element Ti in the filler metal, and the filler metal could form a good interface reaction with the ceramic; meanwhile, the presence of the Ni layer reduced the erosion of Cu by the filler metal, and this type of interlayer could more effectively relieve the residual stress in the brazed joint. When the thickness of the Ni layer was 30 mm and the thickness of the Cu sheet was 0.2 mm, the shear strength of the joint could reach 109 MPa.

Full Text

Preamble

ACTA METALLURGICA SINICA
Vol. 51, No. 2, February 2015, pp. 209-215

Effect of Interlayers on the Microstructure and Shear Strength of Al_2O_3/1Cr18Ni9Ti Brazed Joints

LIU Yi[1,2]**), JIANG Guofeng**[2]**), XU Kun**[1]**), LUO Ximing**[1]**), CHEN Dengquan**[1]**), LI Wei**[1]**)**
[1]) Sino-Platinum Metals Co. Ltd, Kunming 650106, China
[2]) Kunming Institute of Precious Metals, Kunming 650106, China

Correspondent: LIU Yi, Associate Professor, Tel: (0871)68328480, E-mail: liuyi@ipm.com.cn

Supported by: Applied Basic Research Project of Yunnan Province (No. 2010ZC56)

Received: June 27, 2014; **Revised:** November 3, 2014

Abstract

Al_2O_3 ceramic was joined to 1Cr18Ni9Ti stainless steel using Ag-Cu-Ti active filler metal. The effects of three interlayer metals—Cu, Ni, and Ni-coated Cu—on the microstructure and shear strength of the brazed joints were investigated. The results indicated that when Cu was used as the interlayer, a good interfacial reaction formed between the ceramic and filler metal. When Ni served as the interlayer, numerous Ni_3Ti intermetallic compounds formed in the weld seam, preventing the formation of a proper reaction layer at the ceramic/filler interface and reducing the joint' s shear strength. When a Ni-coated Cu sheet was employed as the interlayer, the small amount of Ni did not affect the active Ti element content in the filler metal, allowing a good interfacial reaction between filler and ceramic. Simultaneously, the Ni coating reduced the erosion of Cu by the filler metal, making this interlayer more effective at relieving residual stress in the brazed joint. A maximum shear strength of 109 MPa was achieved when the Ni coating thickness was 30 m and the Cu sheet thickness was 0.2 mm.

Keywords: interlayer metal, Al_2O_3 ceramic, 1Cr18Ni9Ti stainless steel, shear strength

1. Introduction

In recent years, significant research efforts have focused on joining ceramics to metals for a wide range of industrial applications. However, several critical challenges remain unresolved. Among these, achieving atomic bonding at ceramic/metal interfaces and minimizing residual stresses caused by large thermal expansion mismatches between the two materials are the most crucial. The thermal expansion mismatch is particularly problematic because even joints with strong interfaces can fail easily when large residual stresses are present. Therefore, reducing residual stress magnitude is highly desirable. Previous researchers have successfully achieved strong alumina-stainless steel joints using soft metallic interlayers. This study investigates the effects of nickel, copper, and nickel-coated copper interlayers on the microstructure and shear strength of alumina ceramic and 1Cr18Ni9Ti stainless steel joints brazed with Ag-Cu-Ti filler metal.

2. Experimental Procedures

[Figure 2: see original paper]

[Figure 3: see original paper]

The shear tests were conducted at a loading rate of 0.5 mm/min, with joint shear strength values reported as the average of three valid specimens.

2.1 Effect of Interlayer on Weld Seam Microstructure

[Figure 4: see original paper]

Figure 4 shows the overall microstructure of brazed joints between Al_2O_3 and 1Cr18Ni9Ti using 0.4 mm thick interlayers of Cu, Ni, and Cu coated with 50 m Ni. Regardless of the interlayer metal used, dense weld seams formed between Al_2O_3 and 1Cr18Ni9Ti without microcracks at the ceramic side or voids in the seam. With a Cu interlayer, the reaction interface between the Cu sheet and Ag-Cu-Ti filler exhibited a serrated morphology (Fig. 4a) with a thick reaction layer reaching 200 m, primarily due to significant erosion of Cu by the silver-based filler at the brazing temperature (870°C). With a Ni interlayer, the interface between the Ni sheet and filler was smooth (Fig. 4b) with a reaction layer thickness of only about 100 m. According to the Ni-Ag binary phase diagram, Ni and Ag are mutually insoluble in both solid and liquid states, resulting in minimal erosion of Ni by the silver-based filler. With a Ni-coated Cu interlayer, the reaction interface between filler and interlayer was flat and smooth (Fig. 4c), because the Ni coating effectively blocked filler erosion of the Cu sheet, leaving the Cu layer intact within the joint. These results demonstrate that good metallurgical bonding occurred between filler and all three interlayer metals, indicating excellent wettability of the filler on each interlayer surface and producing sound joints. However, severe filler erosion of Cu occurred when using a Cu interlayer, potentially reducing its effectiveness for residual stress relief. With a Ni interlayer, no significant erosion occurred, leaving the Ni skeleton intact. With Ni-coated Cu, the central Cu layer remained complete within the joint.

2.2 Effect of Interlayer Metal on Ceramic-Side Weld Seam Microstructure and Phase Distribution

[Figure 5: see original paper]

Figure 5 shows the ceramic-side microstructure of joints brazed with 0.4 mm thick interlayers of Cu, Ni, and Cu coated with 40 m Ni. A reaction layer approximately 10 m thick formed between filler and ceramic when using Cu (indicated by arrows in Fig. 5a). No distinct reaction layer was observed with a Ni interlayer. With Ni-coated Cu, a reaction layer about 5 m thick formed at the ceramic boundary (indicated by arrows in Fig. 5c).

To further investigate the effect of interlayer metal on reaction products and ceramic-side microstructure, EDS analysis was performed to determine elemental distributions in the reaction layers, with results summarized in Table 2. The weld seam composition varied significantly depending on the interlayer metal used.

With a Cu interlayer, the ceramic-side weld seam exhibited a distinct eutectic structure, primarily comprising dark red Cu-based solid solution (marked 1 in Fig. 5a), bright white Ag-based solid solution containing Cu (marked 2), and a gray interfacial reaction layer between filler and Al_2O_3 (marked 3). Based on the composition of the gray reaction product (Table 2) and literature reports [20,21], the reaction product was identified as $Ti_3(Cu, Al)_3O$, formed through the following reactions [22]:

$1/3Al_2O_3 + Ti \rightarrow TiO + 2/3Al$
$4Cu + Ti \rightarrow Cu_4Ti$
$3Cu_4Ti + 2TiO \rightarrow Cu_3Ti_3O + Ti_2O + 9Cu$

According to literature [22], $Ti_4(Cu, Al)_2O$-type compounds may form under higher Ti and O activities, but such compounds were not observed under the present experimental conditions.

With a Ni interlayer, the ceramic-side weld seam consisted mainly of dark red Cu-based solid solution (marked 1 in Fig. 5b), grayish-white Ag-based solid solution containing Cu (marked 2), and numerous gray Ti-Ni intermetallic compounds distributed throughout the seam (marked 3). EDS analysis (Table 2) identified these gray compounds as Ni_3Ti, consistent with literature [20]. Based on interactions between Ti and Cu, Ni, and Ag, the presence of Ni significantly reduces Ti activity (aTi) due to strong Ni-Ti interactions [20,21], thereby inhibiting formation of $Ti_3(Cu, Al)_3O$ compounds and preventing significant interfacial reaction between Ti and the ceramic. Literature [20] reports formation enthalpies of -10, 39, and -187 kJ/mol for Ti-Cu, Ti-Ag, and Ti-Ni, respectively, showing Ti-Ni formation enthalpy is substantially lower than that of Ti-Cu.

With Ni-coated Cu interlayer, the ceramic-side weld seam comprised dark red Cu-based solid solution (marked 1 in Fig. 5c), white Ag-based solid solution containing Cu (marked 2), Ti-Ni intermetallic compounds (marked 3), and gray interfacial reaction products at the ceramic boundary (marked 4). Combined with EDS results (Table 2), point 3 was identified as Ni_3Ti compound and point 4 as $Ti_3(Cu, Al)_3O$ compound.

These results demonstrate that ceramic/filler interfacial reactions depend on interlayer metal type. With Cu interlayer, a distinct reaction layer forms and no intermetallic compounds appear in the seam. With Ni interlayer, no obvious ceramic reaction layer forms but numerous brittle Ni_3Ti intermetallics appear, which is detrimental to joint performance. With Ni-coated Cu interlayer, although Ni_3Ti intermetallics are present, a distinct ceramic/filler reaction layer forms. However, Ni coating thickness must be controlled, as excessive thickness would affect Ti activity and consequently the interfacial reaction.

2.3 Effect of Interlayer Metal Type on Joint Shear Strength

Using 0.4 mm thick interlayers of Cu, Ni, and Ni-coated Cu (30 m Ni layer), the shear strengths of ceramic/stainless steel joints were 85 MPa, 70 MPa, and 95 MPa, respectively.

Literature [7-10] indicates that soft metal Cu relieves residual stress primarily through plastic deformation and creep, thereby improving joint shear strength. Cu's low strength and elastic modulus (Table 1) make it suitable as an interlayer for ceramic/stainless steel brazing. Microstructurally, the ceramic-side seam with Cu interlayer consists mainly of Ag-based and Cu-based solid solutions (Fig. 5a) with minimal effect on joint performance. Cu does not affect the effective Ti content in the filler, allowing the active element to form a good reaction layer ($Ti_3(Cu, Al)_3O$ compound) with the ceramic. According to Torvund and Grong [23], ceramic/brazed joint mechanical properties depend on reaction layer characteristics and thickness, with maximum strength achieved at an optimal reaction layer thickness of 2-6 m. Therefore, Cu interlayer effectively relieves residual stress in Al_2O_3/1Cr18Ni9Ti brazed joints.

Ni sheet has good elastic-plastic properties that can absorb and relieve residual stress [9]. However, when Ni serves as the interlayer, although good metallurgical bonding occurs between filler and Ni, the Gibbs formation enthalpy of Ti-Ni is much lower than that of Ti-Cu [20], making Ni more reactive with active Ti element and generating large amounts of brittle Ni_3Ti intermetallics. Literature [7,8,24-26] notes that ceramic/Ag-Cu-Ti filler reactions depend primarily on active Ti element. With Ni interlayer, dissolved Ni consumes the active Ti, reducing Ti activity and inhibiting ceramic/filler interfacial reaction, thereby decreasing joint shear strength.

With Ni-coated Cu interlayer, the Cu substrate' s elastic-plastic deformation and creep absorb residual stress while the thin Ni coating blocks filler erosion of Cu, maintaining the effective stress-relief thickness. Simultaneously, the small Ni amount does not affect Ti content or the ceramic/filler interfacial reaction.

Therefore, an optimal Ni coating thickness exists that reduces Cu erosion without affecting Ti activity.

2.4 Effect of Substrate and Coating Thickness on Joint Performance with Ni-Coated Cu Interlayer

[Figure 6: see original paper]

Figure 6 shows the overall joint morphology with varying Ni coating thicknesses. With 20 m Ni coating, severe filler erosion of Cu occurred (Fig. 6a). With 30 m Ni coating, the Ni layer reduced filler erosion of Cu (Fig. 6b), though some erosion remained. With 40 m Ni coating, filler erosion of Cu was completely blocked (Fig. 6c). However, as previously analyzed, excessive Ni consumes active Ti, forming intermetallics that affect the ceramic/filler reaction layer. Therefore, an optimal Ni coating thickness exists where filler just begins to erode Cu but essentially maintains Cu sheet thickness, maximizing joint shear strength.

Using Cu sheets (0.6 mm thick) coated with Ni layers of 20, 30, and 40 m thickness, joint shear strengths were 65 MPa, 80 MPa, and 40 MPa, respectively. The results show that joint shear strength initially increases then decreases with Ni layer thickness, reaching a maximum of 80 MPa at 30 m Ni thickness. At small Ni thicknesses, filler erosion of Cu is not effectively reduced. At large Ni thicknesses, although Cu erosion is blocked, excessive Ni adversely affects the ceramic/filler interfacial reaction. At 30 m Ni thickness, the Ni coating neither affects active Ti content nor blocks Cu erosion, maximizing shear strength.

[Figure 7: see original paper]

Figure 7 shows the shear strength of joints brazed with different Cu sheet thicknesses (with 30 m Ni coating). Shear strength decreases with increasing Cu sheet thickness, reaching 109 MPa at 0.2 mm Cu thickness and dropping sharply when Cu thickness exceeds 0.6 mm. Although Cu’ s good plasticity and toughness absorb residual stress, the large thermal expansion coefficient mismatch between Cu and ceramic (Table 1) increases residual stress with thicker Cu sheets. Consequently, residual stress increases with Cu thickness under these competing factors, reducing joint shear strength.

[Figure 8: see original paper]

Figure 8 shows the ceramic-side microstructure and corresponding EPMA analysis for a joint brazed with 0.2 mm Cu coated with 30 m Ni. A good reaction layer formed between ceramic and filler. EPMA results show minimal Ni in the interlayer, with Ni distributed mainly in the filler forming small, dispersed Ni_3Ti intermetallic particles (black particles indicated by arrows) approximately 1-3 m in size. Ti distributed primarily in the ceramic/filler reaction layer, indicating that the small Ni amount had minimal effect on active Ti content. Therefore, using 0.2 mm Cu coated with 30 m Ni as the interlayer material maximizes Cu thickness while minimally affecting Ti content, effectively relieving residual stress and serving as a rational and effective interlayer for ceramic/stainless steel brazing.

---

3. Conclusions

1. With Cu interlayer, Ag-Cu-Ti filler forms a reaction layer approximately 10 m thick with Al_2O_3. With Ni interlayer, no distinct reaction layer forms between Al_2O_3 ceramic and filler. With Ni-coated Cu interlayer, the reaction layer thickness depends on the Ni coating thickness.
2. Ni-coated Cu sheet serves as a rational and effective interlayer for Al_2O_3 ceramic/1Cr18Ni9Ti stainless steel brazing. A 30 m Ni coating reduces filler erosion of Cu while minimally affecting active Ti content, effectively relieving residual stress in ceramic/stainless steel joints.
3. Joint shear strength depends on both Cu thickness and Ni coating thickness. A maximum shear strength of 109 MPa was achieved with 0.2 mm Cu thickness and 30 m Ni coating thickness.

References

[1] Nicholas M G. Joining of Ceramics. London: Chapman and Hall Ltd, 1990: 1
[2] Schwartz M M. Ceramic Joining. Material Park, Ohio: ASM International, 1990: 1
[3] Park J W, Mendez P F, Eagar T W. Acta Mater, 2002; 50: 883
[4] Zhang J X, Chandel R S, Chen Y Z, Seow H P. J Mater Process Technol, 2002; 122: 220
[5] Li Z R, Fan J X, Feng J C. Aerosp Mater Technol, 2008; (4): 6
[6] Xian A P, Si C Y. J Mater Sci, 1992; 27: 1560
[7] Huang X L, Lin S, Xiao J M. Trans China Weld Inst, 1998; 19: 216
[8] Huang X L, Lin S, Xiao J M, Chu J X. Mater Sci Eng, 1996; 14(3):
[9] Zou G S, Ren J L, Wu A P, Peng Z S, Zeng W J. Mater Rev, 1999; 13(2): 16
[10] Xian A P, Si C Y. Acta Metall Sin, 1991; 27: 421
[11] Zhou Y, Bao F H, Ren J L, North T H. Mater Sci Technol, 1991; 7:
[12] Wang Y, He P, Feng J C, Zhang L X, Gu X L, Wang D Y. Trans Chin Weld Inst, 2007; 28(4): 13
[13] Suganuma K, Okamoto T, Koizumi M, Kamachi K. J Mater Sci, 1987; 22: 3561
[14] Suganuma K. J Mater Sci, 1991; 26: 6144
[15] Zhu M U, Chung D D L. J Mater Sci, 1997; 32: 5321

[16] Shirzadi A A, Zhu Y, Bhadeshia H K D H. Mater Sci Eng, 2008; A496: 501
[17] Tang Q, Chu J X, Zhang X Y. Electros Proc Technol, 2001; 22:
[18] Hao H Q, Wang Y L, Jin Z H, Wang X T. J Mater Sci, 1995; 30:
[19] Zou X. Brazing. Beijing: China Machine Press, 1997: 392
[20] Valette C, Devismes M F, Voytovych R, Eustathopoulos N. Scr Mater, 2005; 52: 1
[21] Arróyave R, Eagar T W. Acta Mater, 2003; 51: 4871
[22] Jocelyn L W. Master Thesis, Massachusetts Institute of Technology, Cambridge, USA, 2001
[23] Torvund T, Grong Ø, Akselsen O M, UlvensØen J H. J Mater Sci, 1997; 32: 4437
[24] Zhu D Y, Wang Y L, Jin Z H. Ordnance Mater Sci Eng, 1998; 21 (20): 22
[25] Wang X Y, Li Y, Wei S Z, Xu L J, Ma X D. New Technol New Proc, 2010; (5): 85
[26] Li Z R, Fan J X, Feng J C. J Mater Eng, 2008; (9): 1

Note: Figure translations are in progress. See original paper for figures.

Source: ChinaXiv —Machine translation. Verify with original.