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Active brazed Invar-SiO_{2f}/SiO_2 joint using a low-expansion composite interlayer

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ABSTRACT

An AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite interlayer was designed to braze the Invar-SiO_{2f}/SiO_2 joints. Ti contents in the interlayer between the tungsten foil and the Invar alloy was optimized. The effects of the tungsten-foil thickness on the microstructure, elemental distribution and mechanical properties of the brazed joints were investigated. Compared with a single AgCu-4.5 wt.%Ti interlayer, a sufficient metallurgical reaction happened on the SiO_{2f}/SiO_2 composite side and the formation of Fe_2Ti, Ni_3Ti compounds which are detrimental to the joint properties, was greatly inhibited by using a low-expansion composite interlayer (LECI). The tungsten interlayer can reduce the CTE (coefficient of thermal expansion) mismatch in the Invar-SiO_{2f}/SiO_2 joint. The highest shear strength of joints brazed with an LECI was 33 MPa, which was 1.75 times higher than the joints brazed with a single AgCu-4.5 wt.%Ti interlayer.

1. Introduction

SiO_{2f}/SiO_2 composites possess low dielectric constants and the physical properties are insensitive to high temperatures (Chen et al., 2003). They have become good candidates for protection materials on missiles and recoverable satellites etc. to substitute quartz ceramics with intrinsic brittleness and low fracture strain (Xu et al., 2007). To further expand the application of quartz fiber-reinforced silica composites, it is necessary to join them with metallic parts.

Active brazing was selected in this research due to its simplicity and good wettability on most ceramics (Voytovych et al., 2006). AgCuTi alloys with fine plasticity have been studied thoroughly and widely adopted in brazing dissimilar materials. Neverheless, a single AgCuTi interlayer could hardly relieve the large CTE mismatch between the base metal (CTE_{Invar}: $17.5 \times 10^{-6}\,K^{-1}$) and the composite ceramic ($CTE_{SiO2f/SiO2}$: $0.56 \times 10^{-6}\,K^{-1}$) well (Sun et al., 2011). Besides, the formation of wave-like Ni-Ti and Fe-Ti compounds with poor plasticity degrades the mechanical performance of the joints when brazing the Invar alloy using an AgCuTi alloy (Yang et al., 2012). To alleviate the residual stress of the joints, many efforts have been made such as adding particles with low CTEs like SiC (Blugan et al., 2007), Al_2O_3 (Yang et al., 2001) and Mo (He et al., 2010). Other efforts include inserting buffer interlayers during brazing such as Cu/Ni porous composite (Zaharinie et al., 2014), a V/Cu barrier interlayer (Cai et al., 2015), and a reticular Cu interlayer (Tian et al., 2015). However, it is difficult to inhibit the formation of the large number of intermetallic compounds with poor plasticity during brazing.

To address the two issues above simultaneously, a tungsten interlayer with a low-expansion coefficient was selected and an AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite interlayer was designed to braze the Invar alloy and the SiO_{2f}/SiO_2 composite. The thickness effect of the tungsten interlayer on the microstructure and mechanical properties of the joint was researched. The mechanism of the residual stress relaxation using an AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite interlayer was analyzed using a finite element method (FEM).

2. Experimental procedure

The original SiO_{2f}/SiO_2 composite ceramics were synthesized by immersion sintering of quartz fibers together with silica sol braided in two dimensions. Commercial Invar alloy with a nominal composition of Fe-36 Ni wt.% was chosen as the parent material in the experiment. The samples of the SiO_{2f}/SiO_2 composite ceramics were sliced into dimensions of $5 \times 5 \times 3\,mm^3$. The sizes of the Invar alloy were $20 \times 10 \times 2\,mm^3$ for shear tests and $10 \times 10 \times 2\,mm^3$ for metallographic observation. The surfaces of the specimens were polished by 800 grid papers before joining. The LECI is comprised of an AgCu-4.5 wt.%Ti foil with a thickness of 100 μm, a tungsten foil with a thickness of 40, 70 and 100 μm respectively and 0.1 g AgCu-1 wt.%Ti tablet pressed by AgCu and TiH_2 powders. All materials were cleaned before joining by ultrasonic cleaning in acetone for 10 min. The comparative experiments were conducted using a single AgCu-4.5 wt.%Ti

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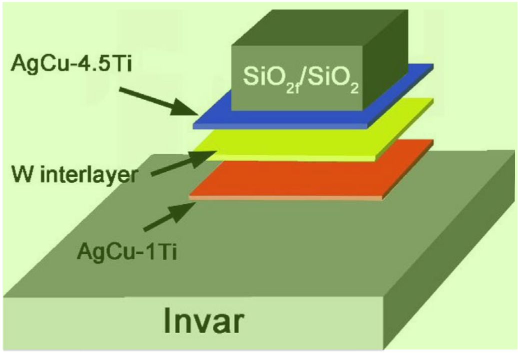

Fig. 1. Sketch of assembly configuration of SiO_{2f}/SiO_2-Invar joints brazed with AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite foils.

interlayer sandwiched between the parent materials. For the joint brazed with a LECI, the sample assembly was Invar-AgCu-1 wt.%Ti-W-AgCu-4.5 wt.%Ti-SiO_{2f}/SiO_2 as shown in Fig. 1. A load of 1.2 kPa was applied to keep the specimens in close contact. The samples were heated to 1123 K with a heating rate of 10 K min^{-1} first and isothermally held for 10 min and then cooled down to room temperature at a rate of 5 K min^{-1} with a vacuum of 3.0×10^{-3} Pa.

Microstructures of the brazed joints were examined by a Helios Nanolab600i scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). Shear strength tests were conducted at a constant speed of 0.5 mm/min using an Instron-1186 universal testing machine and the average strength was determined by five shear specimens conducted at the same temperature. A laser scanning confocal microscope (LSCM, Keyence VHX-1000E) was adopted to investigate the fracture morphologies of the brazed joints.

3. Results and discussion

3.1. Wettability

The assembly sequence of the joint was carefully designed as follows. A 100 μm AgCu-4.5 wt.%Ti foil was placed between the SiO_{2f}/SiO_2 composite and the tungsten interlayer. According to previous experimental results (Yang et al., 2012), the high proportion of wave-like brittle Fe-Ti and Ni-Ti compounds would significantly decrease the plasticity of the braze filler, causing a large stress in the joint. To avoid these brittle compounds, the Ti content in the braze filler should be reduced. An AgCu and an AgCu-1 wt.%Ti braze filler was the suitable candidate, sandwiched between the tungsten interlayer and the Invar alloy respectively. To verify which interlayer was more suitable and reliable, before brazing, a wetting experiment was carried out to examine the interfacial bonding between the braze filler and the tungsten interlayer.

Fig. 2 shows the wetting results and the Ti-W binary phase diagram. The contact angle of AgCu on the tungsten foil was approximately 135°, demonstrating a poor wettability. Gaps can be observed at the interface, indicating a poor bonding between the AgCu and the tungsten foil. In contrast, the interface between the AgCu-1 wt.%Ti and the tungsten foil was quite compact and showed no voids or cracks. The contact angle of the AgCu-1 wt.%Ti/W system was about 40°. The solubility of Ag and Cu in W is extremely low (Vijayakumar et al., 1988) and the metallurgic reaction or interdiffusion could hardly happen between AgCu and tungsten. Therefore, the wettability of AgCu on the tungsten interlayer was poor and the interfacial bonding was extremely weak. In comparison, when AgCu-1 wt.%Ti melted and contacted with the tungsten interlayer, Ti will first adsorb at the solid-liquid interface. Then the molten braze filler sufficiently contacted with the tungsten and fully spread and wetted well on it. According to the Ti-W binary diagram (Lin et al., 2012), tungsten showed an extremely high Ti solubility and Ti can diffuse into tungsten substrate, leading to a strong bonding at the AgCu-1 wt.%Ti/tungsten interface. A minor addition of active Ti would significantly improve the wettability, which is the design principle for the assembly form of the brazed joint as shown in Fig. 1.

3.2. Microstructures of the joints brazed with and without a LECI

Fig. 3 shows the microstructure of the Invar-SiO_{2f}/SiO_2 joints brazed at 1123 K for 10 min using two types of interlayers.

It can be seen from Fig. 3(a) that the brazed seam mainly consisted of AgCu eutectic combined with a large number of wave-like compounds. Compositions of positions A–E in Fig. 3 are listed in Table 1. The wave-like compounds were identified to be Fe_2Ti and Ni_3Ti intermetallics according to the chemical compositions of Point A and B shown in Table 1. This is consistent with previous research results (Sun et al., 2016). These intermetallic compounds formed in the joints were quite brittle and could reduce the plastic deformability of joints (Arróyave and Eagar, 2003). As a result, the brazed seam could hardly offer sufficient plastic deformation to accommodate the large thermal stress generated during the cooling process. Cracks could thus be easily found in the reaction layer on the SiO_{2f}/SiO_2 composite side as shown in Fig. 3(b). Fig. 3(d) shows the interfacial microstructure of the joint brazed using a LECI. A 70 μm thick tungsten interlayer was in the middle of the brazed seam and AgCu eutectic mainly formed on both sides of the tungsten interlayer. The replacement of brittle compounds by the AgCu eutectic with fine plasticity will be beneficial to the mechanical properties of the brazed joint. The reaction layer on the SiO_{2f}/SiO_2 composite side was quite dense and no voids or cracks could be found in Fig. 3(e).

As shown in Table 1, the reaction layers on the SiO_{2f}/SiO_2 composite side in the joints brazed with and without the tungsten interlayer were both composed of double-layer reaction products, i.e., a thinner layer of dark gray Ti-O and Ti-Si phases formed adjacent to the SiO_{2f}/SiO_2 composite and a thicker layer of light gray Cu-Ti-O phase formed adjacent to the braze filler. However, the thickness of the Ti-O, Ti-Si + Cu-Ti-O reaction layer brazed with a single AgCu-4.5 wt.%Ti foil was 1.8 μm, which was much thinner than that brazed with AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite foils (2.4 μm). Fe and Ni atoms from Invar alloy severely dissolved into the liquid alloy. The partial enthalpies of Ti solution in molten Ni and Fe at infinite dilution are −187 kJ mol^{-1} (Arróyave and Eagar, 2003) and −54 kJ mol^{-1} (Valette et al., 2005), indicating that both Ni and Fe would interact strongly with Ti. The active Ti would be trapped and the reaction between Ti and the SiO_{2f}/SiO_2 composite weakened. The reaction layer on the SiO_{2f}/SiO_2 composite side became thinner, which could hardly withstand the thermal stress and cracks thus propagated easily. In contrast, Ti interacted much weaker with tungsten and sufficient metallurgical bond could be achieved on the SiO_{2f}/SiO_2 composite side. That is why the Ti-O, Ti-Si + Cu-Ti-O reaction layer brazed with AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite interlayers was thicker than that brazed with a single AgCu-4.5 wt.%Ti interlayer.

To better understand each phase's distribution, elements' area distribution was obtained as shown in Fig. 4. Ag(s,s) and Cu(s,s) were the major phases that distributed in the areas on both sides of the tungsten interlayer. In spite of the existence of Fe and Ni in the base alloy, only a small proportion of Fe and Ni distributed on the Invar side compared with the case using a single AgCu-4.5 wt.%Ti interlayer. Combined with the distribution of Ti, the contents of the brittle Fe-Ti and Ni-Ti compounds were extremely low and distributed dispersedly, which was due to the suppression of the Ti concentration in the liquid AgCu-1 wt.%Ti alloy. The small proportion of Ti could promoted the wetting and bonding in the AgCu-1 wt.%Ti/W system, and at the same time, suppressed the formation of wave-like brittle compounds.

Fig. 5 displays the effect of the tungsten-foil thickness on the microstructure evolution of the brazed joints. The joints are mainly composed of AgCu eutectic with fine plasticity together with a small proportion of dispersive Fe_2Ti and Ni_3Ti intermetallic compounds. It is

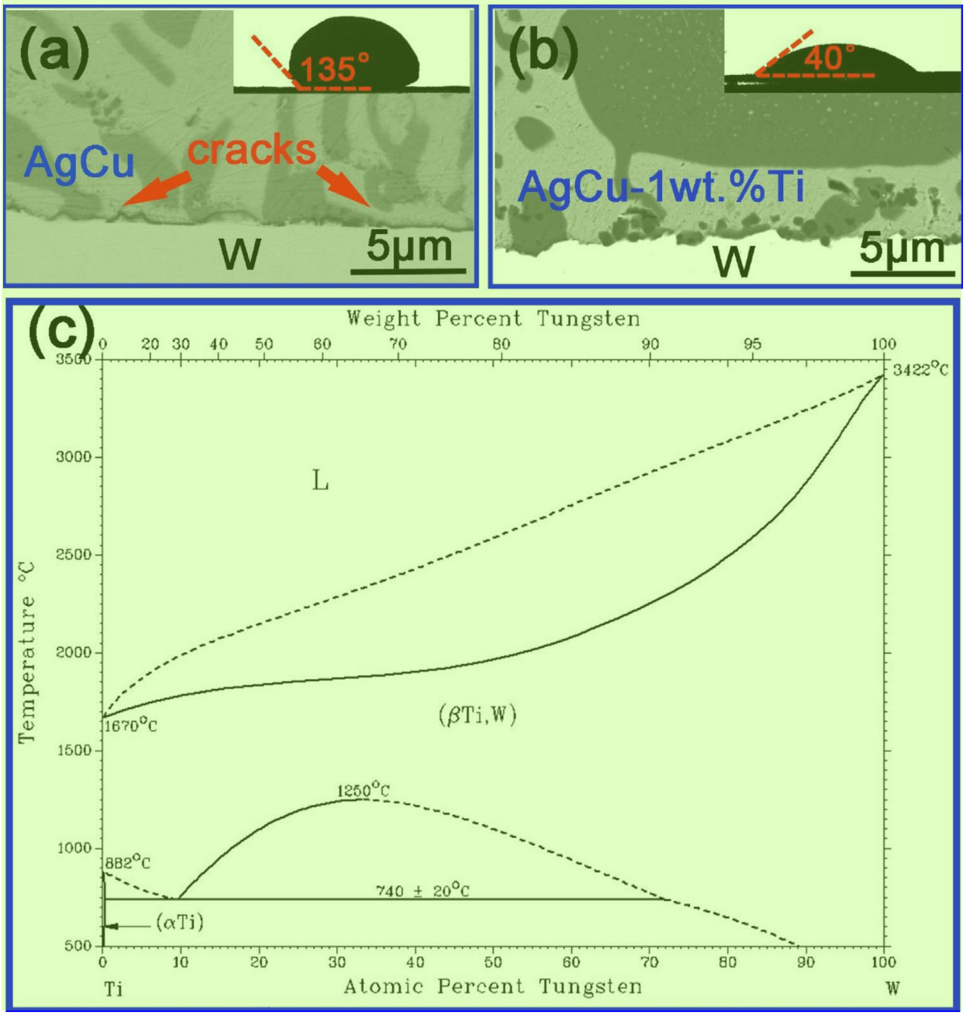

Fig. 2. Contact angle profile and interfacial morphology: (a) AgCu/W, (b) AgCu-1 wt.%Ti/W and (c) Ti-W binary phase diagram.

notable that cracks propagated inside the tungsten interlayer when the thickness increased to 100 μm. Fig. 5(d) shows the thickness of various tungsten interlayers as well as their corresponding brazed seams. The thicknesses of tungsten interlayers were 40, 70 and 100 μm respectively. The tungsten interlayer/brazed seam thickness ratio (TBTR) are 0.30, 0.50 and 0.65 respectively. The TBTR will have a strong effect on the mechanical property of the brazed joint, which will be discussed in the following text.

3.3. Mechanical properties of the brazed joint

Fig. 6 shows the effect of TBTR on the shear strength of the brazed joints. The shear strength increased with the increase of the TBTR. But when the TBTR increased from 0.50 to 0.65, cracks could be found inside the tungsten interlayer (see Fig. 5(c)) and the enhancement of the shear strength was not significant. The maximum shear strength of the joint reached 33 MPa, which was 175% higher than that brazed with a single AgCu-4.5 wt.%Ti interlayer.

There are three reasons responsible for the improvement of the shear strength by using composite interlayers. Firstly, as was discussed above, Fe and Ni interacted strongly with Ti, which would significantly weaken the reaction between Ti and the SiO_{2f}/SiO_2 composite. The reaction layer (Ti-O, Ti-Si + Cu-Ti-O layer) on the SiO_{2f}/SiO_2 composite side was too thin to suffer high shear forces. Cracks could be found in Fig. 3(b) probably due to the high thermal stress generated during cooling process when using a single AgCu-4.5 wt.%Ti interlayer. In contrast, when a tungsten interlayer with a high melting point was used, it would basically maintain its initial shape during brazing and inhibit Fe and Ni from interacting with AgCu-4.5 wt.%Ti on the SiO_{2f}/SiO_2 composite side. As a result, the sufficient reaction between Ti and the SiO_{2f}/SiO_2 composite could be achieved and the reaction layer was strong to bear high shear forces.

Secondly, the plastic deformation ability of the brazed seam containing large-area wave-like brittle compounds was poor when a single AgCu-4.5 wt.%Ti was adopted. It can hardly benefit the stress accommodation and the shear strength of the joint was thus relatively low as shown in Fig. 6. To prove this point, nanoindentation tests were carried out and the results are shown in Fig. 7. We define A_{OPB} (the area of OPB) as the work of the plastic deformation (W_p) and A_{OPC} (the area of OPC) as the total work (W_t). The plastic factor could be defined as follows:

$$\eta_p = \frac{A_{OPB}}{A_{OPC}} \tag{1}$$

After calculation, the plastic factors of Ag(s,s), Cu(s,s) and Fe_2Ti were 89.4%, 84.5% and 77.4% respectively. It demonstrates that both Ag(s,s) and Cu(s,s) possessed better ductility than Fe_2Ti. Therefore, suppressing the formation of the Fe_2Ti phase was beneficial to maintain the fine plastic deformation ability of the brazed seam. When the tungsten interlayer was adopted, the diffusion of the Fe and Ni to the SiO_{2f}/SiO_2 composite side could be prohibited. As a result, the reaction between Fe, Ni and Ti will not happened. No brittle Fe-Ti and Ni-Ti intermetallic compounds formed in the brazed seam between the SiO_2/SiO_2 composite and the tungsten interlayer. Besides, because of the optimum Ti content in the AgCu-1 wt.%Ti braze filler between the tungsten and the Invar alloy, the number of brittle Fe-Ti, Ni-Ti

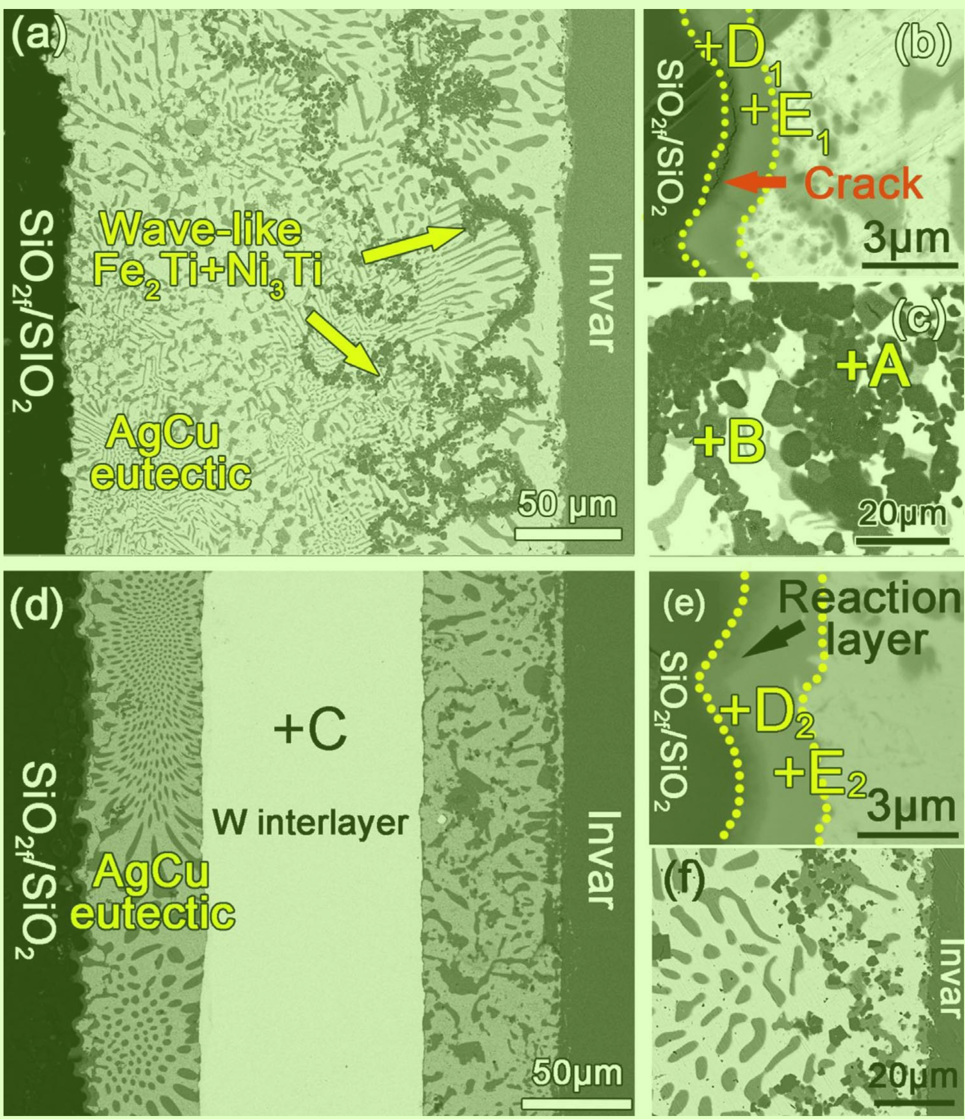

Fig. 3. Microstructure of the Invar-SiO_{2f}/SiO_2 joints brazed at 1123 K for 10 min using: (a-c) AgCu-4.5 wt.%Ti foil, (d-f) AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite foils.

Table 1
Composition (at%) at the points A-E in Fig. 3.

	Fe	Ni	Ti	Ag	Cu	Si	O	W	Possible phase
A	59.70	4.40	29.94	1.76	4.20	–	–	–	Fe_2Ti
B	2.67	65.71	23.37	0.83	7.42	–	–	–	Ni_3Ti
C	–	–	–	–	–	–	–	100	Tungsten
D_1	0.71	1.06	40.53	0.42	3.66	23.48	30.14		Ti-Si + Ti-O
E_1	2.30	4.11	38.72	0.66	21.64	3.26	29.31	–	Cu-Ti-O
D_2	–	–	37.59	0.30	4.13	16.72	41.26	–	Ti-Si + Ti-O
E_2	–	–	37.67	3.12	27.25	3.33	28.63	–	Cu-Ti-O

intermetallic compounds significantly decreased, which made the AgCu eutectic with fine ductility dominate the brazed seam. For the brazed seam with and without a tungsten interlayer, image pro plus software was adopted to characterize the number of the Fe-Ti, Ni-Ti phases. It can be seen from Fig. 8(a) and (b) that the ratio of the brittle Fe-Ti, Ni-Ti compounds to the brazed seam decreased from 10.17% to 3.87%, i.e., a 62% reduction of the intermetallics contents after adopting a LECI, which would significantly increase the plastic deformation ability of the brazed seam.

The strains of the brazed joints could be roughly calculated as follows:

$$\Delta T\Delta\alpha_{Invar-SiO2f/SiO2} = 1.7 \times 10^{-2}$$

$$\Delta T\Delta\alpha_{Invar-tungsten} = 1.25 \times 10^{-2}$$

The results seem to exceed the yield strain of the AgCuTi alloy (an order of 10^{-3}) indicated by Singh et al. (2005). The AgCu eutectic could relax the thermal stress of the joint to some extent during the cooling process due to its own fine ductility. So the suppression of Fe-Ti, Ni-Ti compounds is indeed beneficial to relieve the residual stress of the joint and a reliable joint with high shear strength is reasonable.

Finally, the CTE mismatch between the SiO_{2f}/SiO_2 composite and the Invar alloy is large. The CTE of the SiO_{2f}/SiO_2 composite is $0.56 \times 10^{-6}\,K^{-1}$ from 293 to 1073 K and the CTE of the Invar alloy is $17.5 \times 10^{-6}\,K^{-1}$ at 1000 K which is an order of magnitude higher than that of the composite ceramic. The large CTE mismatch (Δa) between them will induce high thermal stress at the brazed interface on the SiO_{2f}/SiO_2 side. In contrast, the tungsten interlayer has a medium CTE of $4.44\text{-}5.14 \times 10^{-6}\,K^{-1}$ over 293–1293 K. Thus, Δα between SiO_{2f}/SiO_2 and tungsten, between tungsten and Invar were significantly lower than that between SiO_{2f}/SiO_2 and Invar, respectively. The stress gradient on the joint was correspondingly reduced.

The fracture morphologies and fracture modes of the joints brazed with and without a tungsten interlayer are shown in Fig. 9. It can be seen that cracks mainly propagated along the reaction layer when using a single AgCu-4.5 wt.%Ti brazing filler. In contrast, when a LECI containing a 70 μm tungsten foil were adopted, cracks mainly located

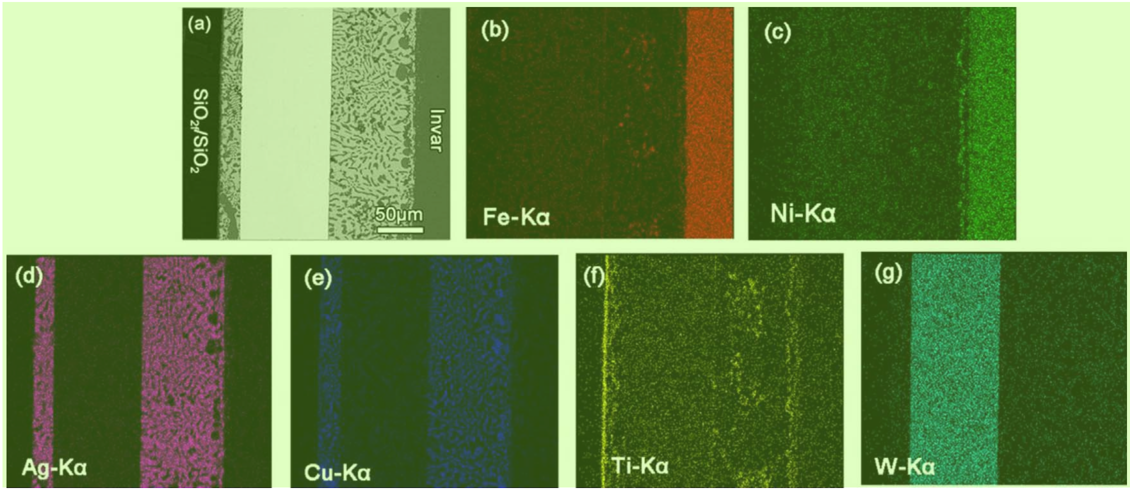

Fig. 4. Microstructure and EDS compositional maps of the joint brazed with the composite interlayer: (a) joint microstructure and (b-g) EDS compositional maps of Fe, Ni, Ag, Cu, Ti and W.

inside the SiO_{2f}/SiO_2 composite as shown in Fig. 9(b). To better understand the crack path in detail, LSCM was utilized. Compared with the straight area in Fig. 9(c), (d) shows that there was an apparent projection on the fracture surface, which means that there was a deflection for the fracture.

The difference in fracture path is well decided by both microstructure and residual stress of the joints. When a single AgCu-4.5 wt.%Ti interlayer was used, a relatively thin reaction layer formed on the SiO_{2f}/SiO_2 composite side due to the strong interaction between Ti and dissolved Fe, Ni atoms. The reaction layer on the SiO_{2f}/SiO_2 composite side (Ti-O, Ti-Si + Cu-Ti-O layer) became the weak zone and cracks could even propagate inside the reaction layer due to the large residual stress as discussed above. Therefore, when shear stress was exerted on the brazed joint, the reaction layer was no doubt preferential to crack.

When a LECI with 70 μm tungsten foil was adopted, AgCu-4.5 wt.%Ti could sufficiently react with the SiO_{2f}/SiO_2 composite due to the barrier effect of the tungsten foil on the diffusion of Fe and Ni atoms. The reaction layer thus became strong enough to bear high shear stress and cracks preferentially propagated inside the SiO_{2f}/SiO_2 composite.

Generally, a bowed crack mode tends to form on the ceramic side when brazing the ceramic using a metal interlayer, caused by a large compressive stress occurred near the brazed seam (Park et al., 2002). However, the fracture of the joint brazed using a LECI was quite flat in most area and there was a reflection at the end of the fracture, leaving a sharp edge there. This type of fracture mode was totally different from the typical bow-like fracture mode, which could indicate a stress relief

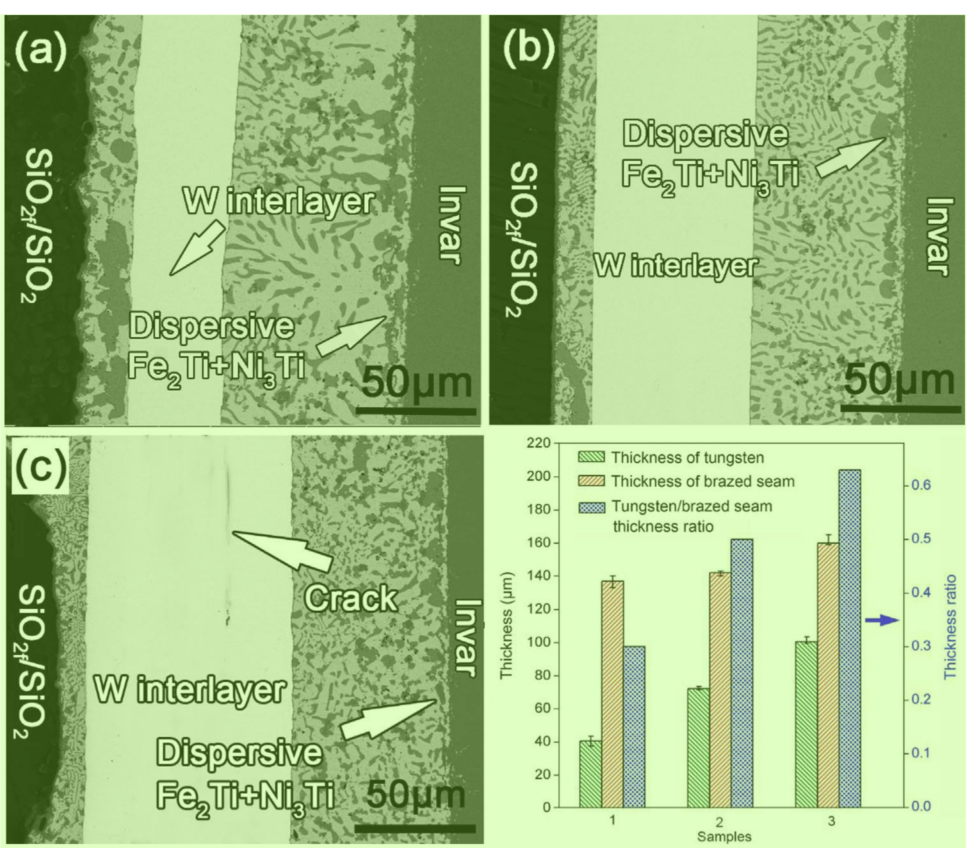

Fig. 5. Effect of tungsten-foil thickness on the microstructure of the joint: (a) 40 μm, (b) 70 μm, (c) 100 μm and (d) thickness of the interlayer and brazing seam.

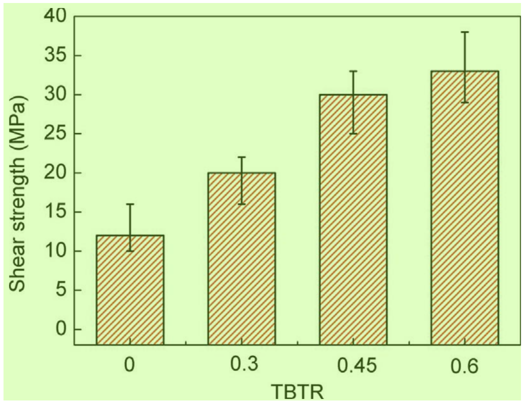

Fig. 6. Effect of the TBTR on the shear strength of the brazed joint.

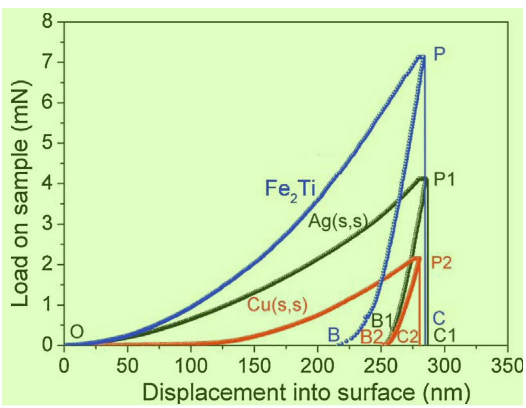

Fig. 7. Load-Displacement curves of the Fe_2Ti, Ag(s,s) and Cu(s,s) phases.

and a stress distribution change might happen as well, which further contributed to the improvement of the joint property.

3.4. The results of FEM simulation

To understand the variation of the stress level and distribution of the joints brazed with and without the LECI, FEM was employed, focusing on the cooling process of the brazed joint.

780 °C was set as the initial temperature point, which is the melting point of the Ag-Cu eutectic. Stress started to form after the temperature dropped lower than 780 °C. The simulation was conducted using a COMSOL Multiphysics software. Fig. 10 shows how the joint was divided by meshes. Note that the meshes were refined in the braze seam zone to get a more accurate result. The model created was elasto-plastic and its dimension was equal to the size of the actual brazed samples. Some boundary conditions were set up at first. The whole configuration does not move or rotate. To improve the simulation efficiency, a half symmetrical model was set up. The assembly was a union form, which means the whole joint was a single part for the simulation. The SiO_{2f}/SiO_2 composite and the tungsten interlayer were an ideal elastomer. The AgCuTi and the Invar alloy were an elasto-plastic body. No additional pressure was applied during simulations.

Table 2 shows the related physical parameters of the bonded materials which were used in the simulation. The effect of the reaction layer on the residual stress of the joint was ignored since the reaction layer thickness was far much thinner than that of the braze seam. Except for the tungsten interlayer, the brazed seam was simplified to be composed of AgCuTi alloy.

Fig. 11 shows the equivalent von mises distribution of the joints brazed with two different interlayers. It can be seen that for the joint brazed with a single AgCuTi interlayer, the high-stress zone (red zone) mainly located at the AgCuTi-SiO_{2f}/SiO_2 interface as shown in the magnified picture in Fig. 11(a). Large differences happened after a LECI was used. The high-stress zone at the braze filler-SiO_{2f}/SiO_2 interface narrowed down significantly as shown in the magnified picture in Fig. 11(b). Another noteworthy point was that a new high-stress zone formed, which mainly located at the tungsten interlayer. While the stress inside the AgCuTi zone in the brazed seam was still relatively low.

To get a detailed understanding of the stress distribution variation after using the LECI. A line was drawn from the bottom of the Invar alloy to the top of the SiO_{2f}/SiO_2 composite along the Z direction as shown in Fig. 12(a).

Fig. 12(b) shows the equivalent Von Mises stress distribution along the Z direction. There were two stress peaks in the joints, which located at the AgCuTi-SiO_{2f}/SiO_2 and the AgCuTi/Invar interfaces. The highest stress located at the AgCuTi-SiO_{2f}/SiO_2 interface, which was around 350 MPa. For the joint brazed with a LECI, four stress peaks existed,

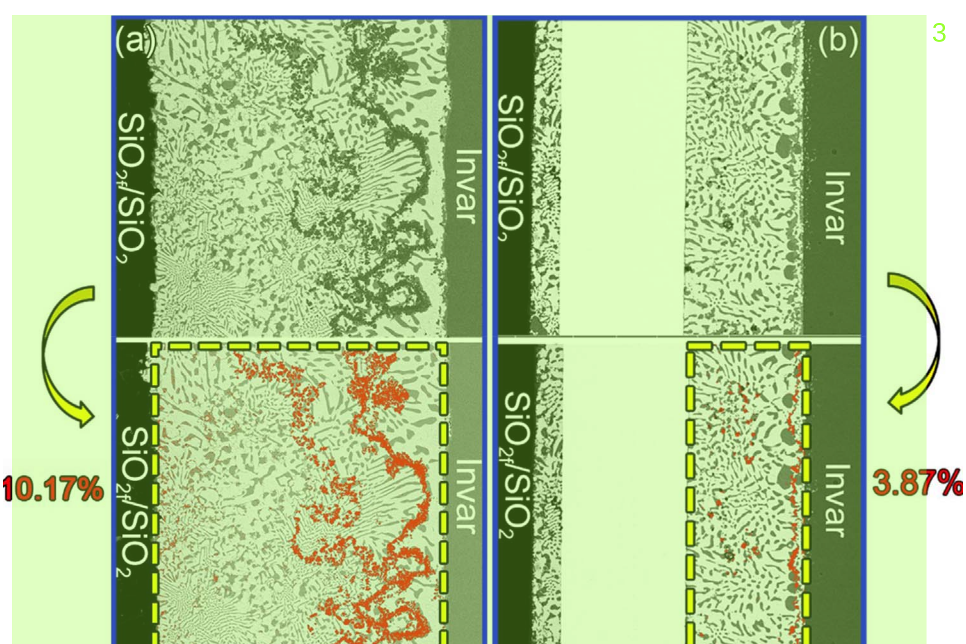

Fig. 8. The ratio of the Fe-Ti, Ni-Ti to the brazed seam: (a) without tungsten interlayer; (b) with tungsten interlayer.

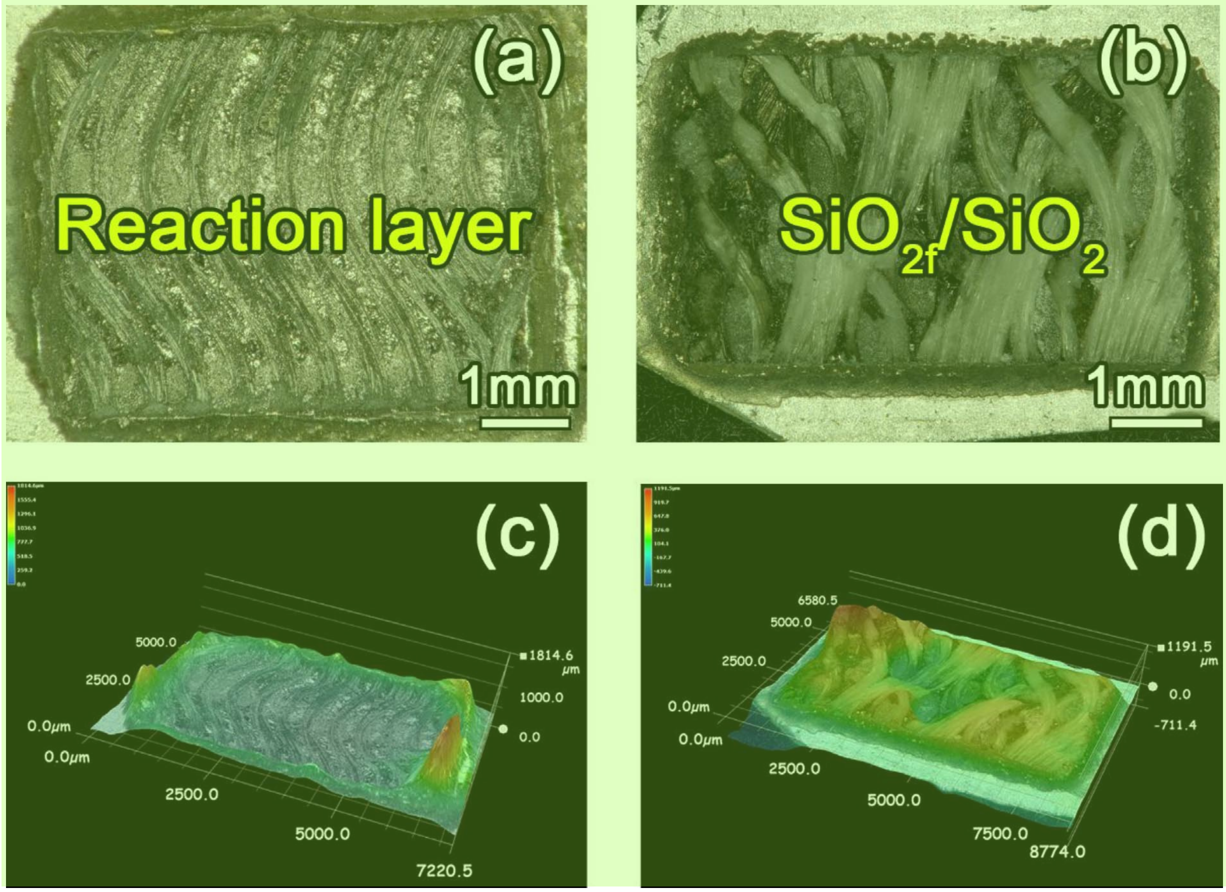

Fig. 9. Fracture morphologies, LSCM images and fracture schematic of the joints brazed with (a), (c) AgCu-4.5Ti and (b), (d) composite interlayer with a 70 μm tungsten foil.

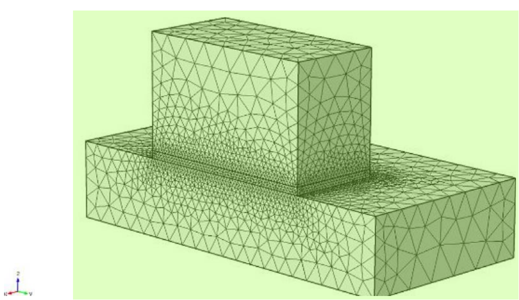

Fig. 10. FEM mesh configuration of Invar-SiO_{2f}/SiO_2 brazed joint.

which located at the AgCu-1 wt.%Ti/Invar, AgCu-1 wt.%Ti/W, W/AgCu-4.5 wt.%Ti and the AgCu-4.5 wt.%Ti-SiO_{2f}/SiO_2 interfaces. It is interesting that the highest stress peak transferred from the AgCuTi-SiO_{2f}/SiO_2 interface (brazed with a single AgCuTi interlayer) to the tungsten interlayer, which was only 225 MPa. The stress of the weakest part of the joint, i.e., the AgCuTi-SiO_{2f}/SiO_2 interface, was only around 110 MPa, which was more than 2 times lower than case brazed with a single AgCuTi interlayer.

From the FEM results, it was clear that compared with the joint brazed with a single AgCuTi interlayer, the LECI showed its positive effects in two aspects. It could firstly transfer the highest stress peak from the AgCuTi-SiO_{2f}/SiO_2 interface to the tungsten interlayer, which meant the stress moved from the weakest part of the joint to a relatively stronger zone. Secondly, the stress at the AgCuTi-SiO_{2f}/SiO_2 interface, on which we paid lots of attention, showed a 2 times decrease. This is

Table 2
Physical and mechanical properties of the materials used in the FEM simulation.

Materials	Temperature (°C)	Elastic Modulus (GPa)	Yield stress (MPa)	CTE ($\times 10^{-6}$ K^{-1})	Poisson's ratio
SiO_{2f}/SiO_2	–	70	–	0.56	0.152
Invar	20	143	310	1.2	0.26
	200	141	115	7.8	
	400	138	90	17.1	
	600	120	75	17.5	
AgCuTi	20	100	230	19.0	0.36
	200	90	170	19.7	
	400	80	98	20.2	
	600	67	25	20.5	
W	20	400	–	4.45	
	200	392			0.28
	400	383			
	600	374			

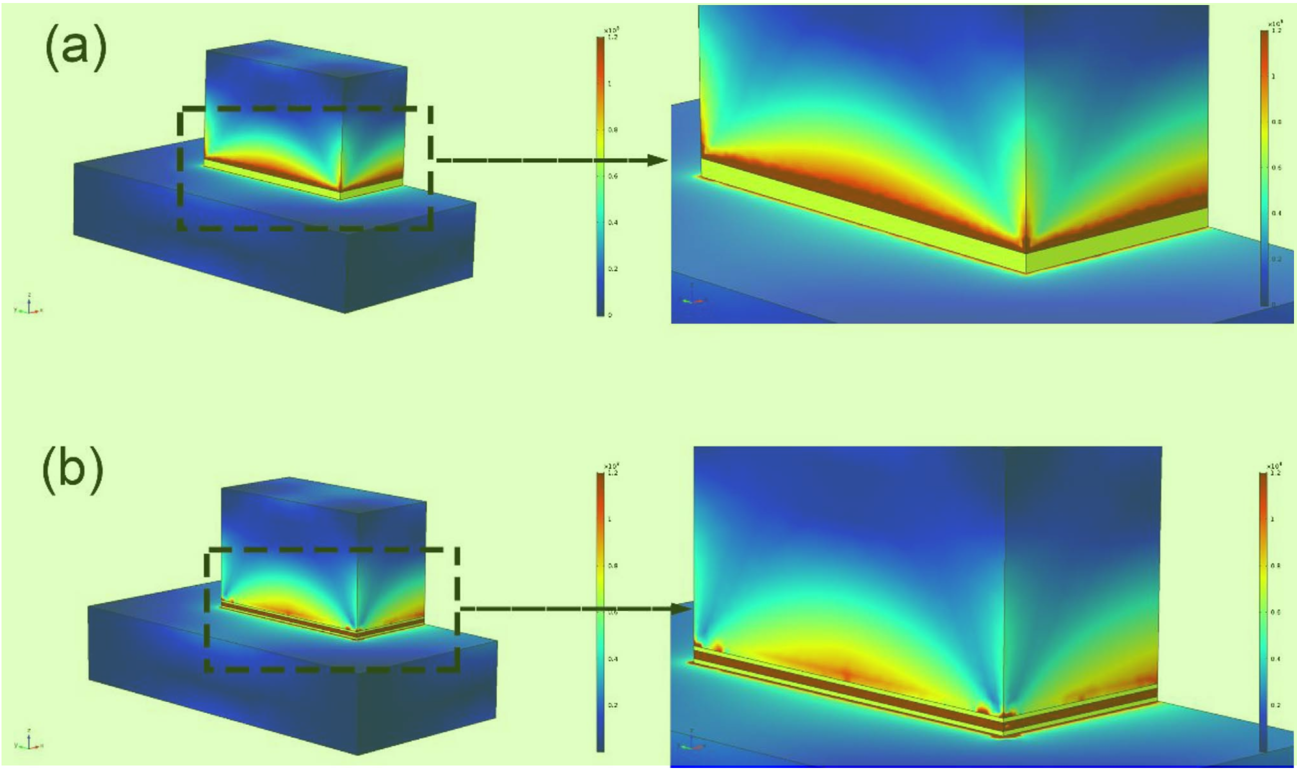

Fig. 11. Residual stress distribution of the Invar-SiO_{2f}/SiO_2 brazed joint with different filler metals: (a) A single AgCuTi interlayer; (b) AgCu-1 wt.%Ti/W/AgCu-4.5 wt.%Ti interlayer.

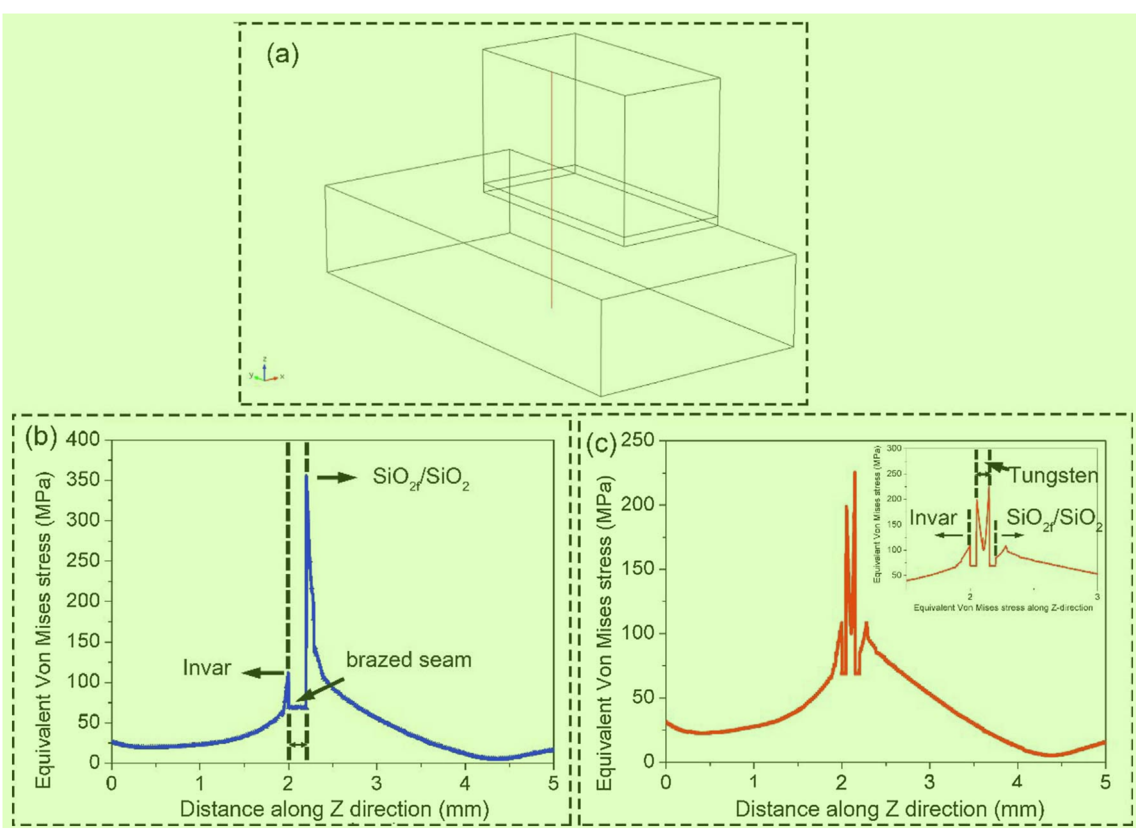

Fig. 12. Equivalent Von Mises stress distribution of the brazed joints (a)Schematic of the stress distribution path; Equivalent Von Mises stress versus distance along Z direction of the joint brazed with (b) A single AgCuTi interlayer; (c) A LECI.

quite consistent with the previous fracture analysis in this paper. For the joint brazed with a single AgCuTi interlayer, the stress at the AgCuTi-SiO_{2f}/SiO_2 interface was large and the reaction layer of the AgCuTi-SiO_{2f}/SiO_2 interface was too thin to suffer the double effects of the residual stress and the shear force during the shear tests. The fracture was thus preferentially located at the reaction layer. While for the joint brazed with a LECI, the stress at the AgCuTi-SiO_{2f}/SiO_2 interface reduced a lot. The strong reaction layer at the AgCuTi-SiO_{2f}/SiO_2 interface was not sensible to crack any more. The SiO_{2f}/SiO_2 composite with low bending strength became much easier to break down in this case. Meanwhile, put the zoomed-in pictures in Fig. 11(a) and (b) into comparison, the stress distribution at the AgCuTi-SiO_{2f}/SiO_2 interface did not show a typical bow-like mode after a LECI interlayer was used. That could be the reason why the fracture of the joint brazed with a LECI interlayer was flat instead of a bow-like one, which was a typical fracture mode often happened in the joint with a large residual stress (Park et al., 2002).

4. Conclusions

An AgCu-4.5 wt.%Ti/W/AgCu-1 wt.%Ti composite interlayer was successfully designed to braze Invar alloy and SiO_{2f}/SiO_2 composites. Based on the results obtained above, several conclusions are summarized as follows:

(1) An AgCu-1 wt.%Ti interlayer was successfully designed between the tungsten foil and the Invar alloy. It could wet the tungsten foil well without any voids along the interface compared with the AgCu interlayer. The AgCu-1 wt.%Ti interlayer could also suppress the formation of excessive brittle Fe_2Ti and Ni_3Ti compounds and AgCu eutectic mainly occupied in the brazed seam compared with a single AgCu-4.5 wt.%Ti interlayer.
(2) Compared with a single AgCu-4.5 wt.%Ti interlayer, a tungsten interlayer could effectively prevent Fe and Ni atoms from reacting with the AgCu-4.5 wt.%Ti on the SiO_{2f}/SiO_2 composite side. The reaction between the AgCu-4.5 wt.%Ti and the SiO_{2f}/SiO_2 composite was sufficient and no cracks were found inside the reaction layer.
(3) The maximum shear strength of the joints brazed with the composite interlayer was 33 MPa, which was 1.75 times higher than that of the joints brazed with a single AgCu-4.5 wt.%Ti interlayer. The shear strength of the joints improved with the increase of the TBTR. When the TBTR exceeded 0.45, the enhancement of shear strength was limited.
(4) FEM results indicated that the LECI can transfer the highest stress peak from the AgCuTi-SiO_{2f}/SiO_2 interface to the tungsten interlayer and the highest equivalent Von Mises stress reduced from 350 MPa to 225 MPa. The equivalent Von Mises stress at the AgCuTi-SiO_{2f}/SiO_2 interface decreased from 350 MPa to 110 MPa after the joint was brazed using a LECI.

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References

Arróyave, R., Eagar, T.W., 2003. Metal substrate effects on the thermochemistry of active brazing interfaces. Acta Mater. 51, 4871–4880.
Blugan, G., Kuebler, J., Bissig, V., Janczak-Rusch, J., 2007. Brazing of silicon nitride ceramic composite to steel using SiC-particle-reinforced active brazing alloy. Ceram. Int. 33, 1033–1039.
Cai, Q., Liu, W., Ma, Y., Liu, H., 2015. Microstructure, residual stresses and mechanical properties of diffusion bonded tungsten-steel joint using a V/Cu composite barrier interlayer. Int. J. Refract. Met. Hard Mater. 48, 312–317.
Chen, H., Zhang, L.M., Jia, G.Y., Luo, W.H., Yu, S., 2003. The preparation and characterization of 3D-silica fiber reinforced silica composites. Key Eng. Mater. 249, 159–162.
He, Y.M., Zhang, J., Sun, Y., Liu, C.F., 2010. Microstructure and mechanical properties of the Si_3N_4/42CrMo steel joints brazed with Ag-Cu-Ti + Mo composite filler. J. Eur. Ceram. Soc. 30, 3245–3251.
Lin, C.C., Shu, C.H., Chen, C., Shiue, R.K., Shy, H.J., 2012. Brazing porous tungsten and molybdenum using palladium and titanium foils brazing porous tungsten and molybdenum using palladium and titanium foils. Int. J. Refract. Met. Hard Mater. 31, 284–287.
Park, J.W., Mendez, P., Eagar, T., 2002. Strain energy distribution in ceramic-to-metal joints. Acta Mater. 50, 883–899.
Singh, M., Shpargel, T., Morscher, G., Asthana, R., 2005. Active metal brazing and characterization of brazed joints in titanium to carbon-carbon composites. Mater. Sci. Eng. A 412, 123–128.
Sun, Y., Zhang, J., Geng, Y.P., Ikeuchi, K., Shibayanagi, T., 2011. Microstructure and mechanical properties of an Si_3N_4/Si_3N_4 joint brazed with Au–Ni–Pd–V filler alloy. Scr. Mater. 64, 414–417.
Sun, Z., Zhang, L.X., Zhang, Z.H., Hao, T.D., Feng, J.C., 2016. Microstructure and mechanical response of the SiO_{2f}/SiO_2 composite and Invar alloy joints brazed with an AgCuTi alloy. Mater. Des. 111, 239–247.
Tian, X., Feng, J., Shi, J., Li, H., Zhang, L., 2015. Microstructure, residual stresses and mechanical properties of diffusion bonded tungsten–steel joint using a V/Cu composite barrier interlayer. Ceram. Int. 41, 145–153.
Valette, C., Devismes, M.F., Voytovych, R., Eustathopoulos, N., 2005. Interfacial reactions in alumina/CuAgTi braze/CuNi system. Scr. Mater. 52, 1–6.
Vijayakumar, M., Sriramamurthy, A.M., Naidu, S.V., 1988. Calculated phase diagrams of Cu-W, Ag-W and Au-W binary systems. Calphad 12, 177–184.
Voytovych, R., Robaut, F., Eustathopoulos, N., 2006. The relation between wetting and interfacial chemistry in the CuAgTi/alumina system. Acta Mater. 54, 2205–2214.
Xu, C.M., Wang, S.W., Huang, X.X., Guo, J.K., 2007. Processing and properties of unidirectional SiO_{2f}/SiO_2 composites. Ceram. Int. 33, 669–673.
Yang, J., Fang, H., Wan, X., 2001. Effects of Al_2O_3-particulate-contained composite filler materials on the shear strength of alumina joints. Mater. Sci. Technol. 9, 676–678.
Yang, Z., Zhang, L., Xue, Q., He, P., Feng, J., 2012. Interfacial microstructure and mechanical property of SiO_2-BN ceramics and Invar joint brazed with Ag-Cu-Ti active filler metal. Mater. Sci. Eng. A 534, 309–313.
Zaharinie, T., Moshwan, R., Yusof, F., Hamdi, M., Ariga, T., 2014. Vacuum brazing of sapphire with Inconel 600 using Cu/Ni porous composite interlayer for gas pressure sensor application. Mater. Des. 54, 375–381.