Materials Research Express

PAPER

Microstructure and mechanical properties of Ti6Al4V alloy and sapphire joint brazed with graphene-AgCuTi

Zhou Fan¹, Kun Zhang¹, Jian-yi Liu¹, Min Hu¹ and Chun-feng Yang¹

¹ School of Materials Science and Engineering, Southwest Petroleum University, Chengdu, 610000, People’s Republic of China
² Petroleum Engineering School, Southwest Petroleum University, Chengdu, 610000, People’s Republic of China
³ Journal center of Southwest Petroleum University, Southwest Petroleum University, Chengdu, 610000, People’s Republic of China

E-mail: fazhou505@163.com

Keywords: graphene, sapphire, Ti6AI4V alloy, coefficient of thermal expansion

Abstract

The sapphire (α-Al₂O₃) and Ti6Al4V (TC4) alloy was successfully brazed with GNSs (Graphene)-AgCuTi composite filler. The effects of different GNSs content and different brazing temperatures on the interface and mechanical properties of brazed joints were studied. The typical interfacial microstructure of the brazed joint was: Ti6Al4V/Ti₂Cu/TiCu/Ti₃Cu₄/Ag(s.s)/TiC + GNSs/TiCu/Ag(s.s)/Ti₃Cu₃O/sapphire. The result shows the diffusion layer (I) and Ti-Cu layer (II) got thickened gradually as the brazing temperature rose. At the same time, the thickness of Ti₃Cu₃O reaction layer increased from 1.9 m to 4.7 m when the brazing temperature increased from 810 °C to 870 °C. The thermal expansion coefficient of the composite filler and the mechanical properties of the joint were calculated and analyzed. The addition of GNS can reduce the mismatch of thermal expansion coefficient between the brazing joint and Ti₃Cu₃O reaction layer. When the 0.1 wt% GNSs-AgCuTi composite filler was put into use, the shear strength of the joint reached 29.1 MPa. However, the agglomeration of excess graphene hindered the diffusion of the elements, making the Ti₃Cu₃O layer discontinuous and causing a mismatch in the coefficient of thermal expansion among the materials, thus reducing the shear strength of the joint.

1. Introduction

Sapphire (single crystal α-Al₂O₃) is a high-purity single crystal and optical material with high hardness, high temperature resistance and corrosion resistance [1–3]. It is widely used in aerospace, electrical and electronic fields and new energy devices [4–6]. Ti6Al4V boasts high strength, good heat resistance and corrosion resistance, which is widely used in the aviation field [7, 8]. Therefore, the sapphire and the metal are connected to obtain a subject with both excellent performances, thereby maximizing the performance of the sapphire.

However, in practical engineering applications, sapphire has the characteristics of high hardness, brittleness and poor toughness. There remains great difficulties in processing and manufacturing of complex and large-sized parts, which has certain limitations in the application of sapphire in the engineering field [2, 6, 9]. Sapphire and metal materials have quite a chemical bond difference, and there are large differences in microstructure, chemical properties and physical properties, especially in thermal expansion coefficient [10]. The brazed joint during the brazing process produces large residual stresses which greatly weaken the mechanical properties of the joint. On the other hand, the problem of wettability of ceramics and metals occurs during brazing. The first condition is to ensure that the brazing alloy should have good wettability on the ceramic surface [11]. The current solution is to metallize the ceramic surface or to add active elements to the solder [12]. However, the role of traditional active solders is limited, so it is urgent to study new solders to improve the wettability between ceramics and metals so as to solve the residual stress of the joints [13, 14].

As a new type of two-dimensional nanomaterial, graphene boasts good thermal properties, large specific surface area, high thermal tensile strength and extremely stable chemical properties [15, 16]. Graphene is often used as a reinforcing phase to improve the properties of metal matrix composites [17, 18]. Graphene is added to a
conventional brazing filler metal to achieve the purpose of improving the performance of the brazed joint. Numerous studies have shown that the addition of low coefficient of thermal expansion materials to brazing alloys helps to relieve residual stress and improve the mechanical properties of ceramic-metal joints [19–21]. Liu et al reported that the addition of GNSs in the brazing seam could alleviate the residual stress and improve the joint strength of TC4/C/C composite [22].

In order to study the effect of the addition of GNSs in the composite filler on the brazing of Ti6Al4V alloy and Sapphire, GNSs-AgCuTi composite filler with different GNSs content was prepared by mechanical ball grinding. Furthermore, the effects of GNSs content and brazing temperature on the interfacial microstructure, mechanical properties and fracture mode of the brazed joints were investigated in details.

2. Experimental procedures

The sapphire (purity >99.9%) used in this study was supplied by Chengdu Technology Co., Ltd The Ti6Al4V plates were supplied by Aviation Industry Corporation of China, Ltd All sample surfaces were grinded by SiC paper and ultrasonic-cleaned in acetone prior to vacuum brazing. The commercial Ag–Cu–Ti powders with the average diameter of 10 μm were supplied by Taizhou City Eric Advanced Materials Co., Ltd The graphene was supplied by Deyang Carbon Technology Co., Ltd The GNSs-AgCuTi composite filler was prepared by adding GNSs into Ag–Cu–Ti powders. The contents of GNSs in the composite filler were 0 wt%, 0.1 wt%, 0.3 wt%, and 0.5 wt%, respectively. Firstly, graphene with different mass fractions was weighed and placed in alcohol and then dispersed in an ultrasonic-cleaning machine for 30 min. Secondly, the ultrasound-treated graphene and AgCuTi alloy were grinded for 6 h at a speed of 400 r min\(^{-1}\) with a QM-SB planetary ball mill. Then, the GNSs-AgCuTi composite filler is dried in a vacuum-drying oven for 4 h at a temperature of 40 °C. Lastly, the dried filler was stored in a vacuum storage cabinet and taken out when needed. Figure 1 shows the morphology and XRD analysis of the GNSs-AgCuTi composite filler. It can be seen from figure 1(a) that GNSs is evenly distributed around the AgCuTi filler and no metallurgical reaction occurred during mechanical ball milling. The GNSs-AgCuTi composite filler was mixed with a small amount of acetone for preparing a composite filler paste. The composite filler paste was coated on the surface of sapphire with a thickness of 100–200 μm. Brazing experiments were carried out at 810–870 °C for 10 min in a vacuum furnace with the vacuum level up to 5 × 10\(^{-3}\) Pa to avoid the oxidation of active Ti during heating. To ensure uniform temperature in the furnace, the sample assembly was firstly heated to 750 °C at a rate of 20 °C min\(^{-1}\) and held for 30 min Then it was heated to the brazing temperature at a rate of 5 °C min\(^{-1}\) and held for 10 min Finally, in order to reduce the generation of joint internal stress, the sample was cooled down to 300 °C at a rate of 2 °C min\(^{-1}\) and then furnace-cooled down to the room temperature.

After brazing, the microstructure of the brazed joints was analyzed with a scanning electron microscope (SEM, ZEISS EVO MA15) equipped with an energy dispersive spectrometer (EDS). The reactant phase formed in the joints was identified by x-ray diffractometer (XRD, X Pert PRO MPD) with Cu-Kα radiation. The shear strength of the brazed joints was tested with a microcomputer controlled electronic universal testing machine (CWT6104). At least 3 specimens brazed at the same brazing parameter were tested to obtain average shear strength. The fracture surface was observed with SEM and EDS.
3. Results

3.1. Interfacial structure of the brazed joints

Figure 2 shows the typical microstructure of sapphire and Ti6Al4V joint brazed with 0.1% GNSs-AgCuTi composite filler at 860 °C for 10 min. Figure 2(a) shows the microstructure of the complete joint, which indicates the successful connection of the Ti6Al4V and sapphire. The formation of such a good joint indicates that the liquid composite filler has good wettability and sufficient fluidity. The typical joint interface can be divided into four zones: (I) diffusion layer of the Ti6Al4V and GNSs-AgCuTi composite filler, (II) reaction layer of the Ti6Al4V and GNSs-AgCuTi composite filler, (III) brazing seam zone, (IV) a continuous reaction layer adjacent to sapphire. The reaction product in the joint can be clearly understood by EDS analysis, as listed in Table 1.

Table 1. Chemical compositions and possible phase of each spot marked (at%) in figure 2.

| Elements | Ag  | Cu  | Ti  | V   | Al  | O   | C   | Possible phase                      |
|----------|-----|-----|-----|-----|-----|-----|-----|--------------------------------------|
| A        | 1.45| 10.53| 71.51| 7.78| 8.763| —   | —   | Ti(s,s) + Ti2Cu                      |
| B        | 3.06| 26.38| 61.75| 2.33| 3.95 | —   | 2.53| Ti2Cu                               |
| C        | 9.54| 40.04| 43.07| 0.64| 2.28 | —   | 4.43| TiCu                                 |
| D        | 6.52| 52.23| 38.11| 0.25| 0.21 | 2.68| Ti3Cu4                               |
| E        | 83.42| 10.16| 4.33 | —   | —   | —   | 2.09| Ag(s,s)                              |
| F        | 42.99| 9.10 | 8.04 | 0.93| 10.18| 28.76| TiCu + Ag(s,s)                       |
| G        | 9.21| 11.31| 18.95| 6.23| 5.16 | 49.12| TiC + GNSs                           |
| H        | 3.13| 6.99 | 9.55 | 1.66| 2.64 | 76.03| TiCu + GNSs                          |
| I        | 16.58| 44.27| 30.16| 1.35| 2.26 | 5.38 | Ti3Cu4                               |
| J        | 10.52| 42.11| 37.69| 1.24| 1.97 | 6.47 | TiCu                                 |
| K        | 0.35| 30.90| 32.31| 12.91| 13.16| 10.37| Ti5Cu3O                              |

Figure 2. Microstructure of Ti6Al4V/sapphire joint brazed with 0.1% GNSs-AgCuTi composite filler at 860 °C for 10 min; (a) the entire joint; (b) magnification of Ti6Al4V/composite filler interface; (c) magnification of local composite filler interface; (d) magnification of composite filler/sapphire.
Zone I and II can be divided into four layers marked as A, B, C and D respectively from TC4 side to composite filler side, as shown in figure 2 (b). The region I was mainly the diffusion layer and the region II was a series of Ti-Cu compound layers. During the brazing process, Cu accumulates on the Ti6Al4V side and diffuses into the TC4 substrate \([23, 24]\). Therefore, region A was mainly composed of Ti(s,s) + Ti2Cu. Since the partial enthalpy of the Ti solution in molten Cu is infinitely diluted \((-10 \text{ kJ mol}^{-1})\), Cu has a stronger tendency to react with Ti than Ag \([25]\). As a result, a continuous Ti-Cu composite layer with different Ti/Cu stoichiometry was formed in region II. According to the EDS analysis result listed in table 1 and the reference reported, the phases marked by B-D were Ti2Cu, TiCu, and Ti3Cu4, respectively. The zone II can be marked as Ti-Cu layers for convenience in the following paragraphs. Additionally, white Ag solid solution \((\text{Ag(s, s)})\) is formed at point E due to copper consumption during brazing.

Figure 2 (c) shows the local magnified microstructure of brazing seam. It can be seen that there are not only white phase, but also gray and black phase distributed in the brazing seam. According to the EDS analysis result, the gray phase (point F) was TiCu, and the black phase (point G, H) was GNSs. Additionally, it was also found that TiC (point G) was distributed near GNSs. To further determine the product phase, XRD was used for analysis, as shown in figure 3. XRD analysis results indicate the formation of TiC. During brazing, GNSs reacting with Ti elements and promoting the formation of TiC particles has been reported \([22]\). In addition, the free energy formed by TiC in the liquid alloy \((\Delta G = -184 + 1.26 \times 10^{-2} T \text{ [26]} )\) promotes the reaction of Ti with GNSs and induces the growth of TiC. In order to further verify the existence of TiC, the first principle is used for calculation with Materials Studio. For the first-principle calculations, the self-consistent convergence conditions are set as follows: total energy less than \(2.0 \times 10^{-5} \text{ eV atom}^{-1}\), force on each atom less than 0.05 eVÅ\(^{-1}\), and stress deviation less than 0.1 GPa. The results calculated show that the adsorption energy and the adsorption height between Ti atom located in the H position of GNSs (figure 4) and GNSs are 2.351 eV and 2.165 Å respectively, which indicates that the adsorption type between Ti atoms and GNSs is chemisorption. This adsorption results consistent with reports in the literature \([27, 28]\).

Figure 2 (d) shows the magnified microstructure of zone IV. A continuous reaction layer was observed near the sapphire in figure 2 (d). According to the EDS and XRD analysis result, the continuous reaction layer on the sapphire side is Ti3Cu3O phase. The formation of Ti3Cu3O comes into being due to the reaction between Al2O3.
and the liquid composite filler. The existence of Ti$_3$Cu$_3$O is the key to ensure the connection between Ti6Al4V and sapphire. Additionally, Ti$_3$Cu$_3$O is a very stable oxide with a reaction free energy of about $-170$ KJ mol$^{-1}$ [29].

### 3.2. Effect of brazing temperature and addition of GNSs on the microstructure of Ti$_6$Al$_4$V/sapphire brazed joints

Figure 5 shows the interfacial microstructure of Ti$_6$Al$_4$V/sapphire joints brazed at different temperature for 10 min with 0.1 wt%GNSs-AgCuTi: (a) 810 °C, (b) 830 °C, (c) 850 °C, (d) 870 °C, (e) the thickness of the phases in the joint.

![Interfacial microstructure of Ti6Al4V/sapphire joints brazed at different temperature for 10 min with 0.1 wt%GNSs-AgCuTi](image)

*Figure 5.* Interfacial microstructure of Ti$_6$Al$_4$V/sapphire joints brazed at different temperature for 10 min with 0.1 wt%GNSs-AgCuTi: (a) 810 °C, (b) 830 °C, (c) 850 °C, (d) 870 °C, (e) the thickness of the phases in the joint.
the brazing temperature, which accelerates the diffusion of Ti and Cu atoms, thereby promoting the expansion and isothermal solidification of the Ti-Cu eutectic liquid phase. During the isothermal solidification process, the residual liquid phase decreased, with more Ti atoms diffusing into the brazing seam and Cu atoms into the base material, which increases the thickness of the Ti-Cu layer. Additionally, the reaction layer (Ti$_3$Cu$_3$O layer) on the sapphire side thickens as the temperature rises. From figure 5(a), the Ti$_3$Cu$_3$O layer was only 1.9 μm-thick when the brazing temperature was 810 °C. At 870 °C, the thickness of the Ti$_3$Cu$_3$O layer reached 4.7 μm as shown in figure 5(e). The increase of the thickness of the Ti$_3$Cu$_3$O reaction layer was attributed to an increase in the Ti concentration on the sapphire side due to the temperature rise. In addition, it is possible that higher brazing temperatures sped up the chemical reaction between Al$_2$O$_3$ and Ti $^{[31]}$.

Figure 6 shows the effect of 0.0 wt%, 0.1 wt%, 0.3 wt% and 0.5 wt% GNSs on the microstructure of Ti6Al4V/sapphire joints brazed with GNSs-AgCuTi composite filler at 860 °C for 10 min. The change of the interface microstructure significantly increased the content from 0.0 to 0.5 wt% GNSs. The thickness of diffusion layer (I) and Ti-Cu intermetallic compound (II) on the side of titanium alloy decreased with the increase of GNSs content. This indicates that the addition of graphene can effectively limit the growth of the diffusion layer and the Ti-Cu phase. Generally, most surface active substances are easily absorbed by crystal plane. The interface energy decreases with the increase of adsorption capacity $^{[7, 32]}$. As a result, the growth rate of the crystal plane decreases. Graphene as a nanomaterial has a higher specific surface area as well as a higher surface tension $^{[33, 34]}$. Therefore, graphene can be easily adsorbed on the surface of the solder, which prevents the diffusion of the elemental titanium and copper, so the growth of the titanium copper phase is suppressed. In addition, we can see that due to the action of graphene, the titanium-copper phase was evenly distributed in the brazing seam, and the interface structure was well-distributed. The reaction layer (Ti$_3$Cu$_3$O layer) on the sapphire side gradually decreased as the graphene increased. The Ti$_3$Cu$_3$O reaction layer was thin and discontinuous when GNS content was 0.5%, as seen in figure 6(d). This is due to the fact that too much graphene hinders the diffusion of the element titanium and copper, which slows down the formation of the reaction layer. However, this can seriously affect the metallurgical bonding of brazing seam and sapphire. When the content of graphene is large, graphene has a local agglomeration phenomenon due to a large surface energy in figures 6(c) and (d). This phenomenon hinders the diffusion of elements and results in the very uneven interfacial structure.

Figure 6. Interfacial microstructure of Ti6Al4V/sapphire joint brazed with GNSs-AgCuTi composite filler with different GNSs content at 860 °C for 10 min: (a)0.0; (b)0.1; (c)0.3; (d)0.5(wt%).
It can be concluded that excessive GNSs in the GNSs-\textit{AgCuTi} composite filler lead to a discontinuous Ti$_3$Cu$_3$O layer and uneven interface structure, which is not conducive to the bonding performance of brazing joints.

### 3.3. Mechanical properties of Ti6Al4V/sapphire brazed joints

Figure 7(a) shows the shear strengths of the Ti6Al4V/sapphire joints brazed at different temperature for 10 min. The shear strength of brazing joint increases first and then decreases with the increase of brazing temperature. The interaction between base materials and composite filler is not adequate when the brazing temperature is lower. Therefore, a thin Ti$_3$Cu$_3$O reaction layer formed adjacent to sapphire, which induced a lower joint strength. When the temperature increases further, the Ti$_3$Cu$_3$O reaction layer increases obviously and the joint strength decreases. That the CTEs of both Ti$_3$Cu$_3$O phase ($\sim 15.1 \times 10^{-6} \text{K}^{-1}$)\cite{25} and Ag-based brazing filler ($\sim 18.2 \times 10^{-6} \text{K}^{-1}$)\cite{31} are larger than that of the sapphire ($\sim 6.5 \times 10^{-6} \text{K}^{-1}$)\cite{35} causes a large residual stresses in the joints. In addition, a large number of brittle phase Ti-Cu compounds occupy the brazing seam, which is not conducive to relieving residual stress. Therefore, when the temperature is too high, the joint strength decreases.

Figure 7(b) shows the effect graphene content on joint properties. It can be seen that as the GNS content increases from 0 to 0.1 wt%, the joint strength increases from 17.5 MPa to 29.1 MPa. However, the strength of the joint decreased when the graphene content exceeded 0.1 wt%. When the GNS is added, the interface structure of the brazing seam becomes finer and more uniform, which is conducive to the joint strength. In addition, the addition of GNSs helps to relieve residual stress of joints.

In order to further explore the effect of graphene on the thermal expansion coefficient of composite filler, the coefficient of thermal expansion (CTE) and Young’s modulus of GNSs-\textit{AgCuTi} composite filler can be calculated separately, as in equations (1) and (2)\cite{36}, based on the theory of Eshelby and law of hybrid composites.

$$\alpha K = \alpha M - \frac{3fEG(\alpha M - \alpha G)}{(EG-EM)(1 + 2f) + 3EM}$$  \hspace{1cm} (1)
in which $\alpha_K$ is the CTE of the GNSs-AgCuTi composite filler; $f$ is the volume percentage of GNSs in composite filler; $\alpha_M$ is the CTE of AgCuTi filler; $\alpha_G$ is the CTE of GNSs, while $E_M$ and $E_G$ are the Young's modulus of AgCuTi filler and GNSs respectively and $E_K$ is the Young's modulus of the composite filler. As can be seen from table 2, the volume percentage of different GNSs content in composite filler $(f)$ is 0.939%, 2.765%, 4.526%, respectively. Therefore, the CTE of composite filler $(\alpha_K)$ is about $15.6 \times 10^{-6} K^{-1}$, $11.4 \times 10^{-6} K^{-1}$, $10.1 \times 10^{-6} K^{-1}$, respectively. The Young's modulus of composite filler $(E_K)$ is about 102.7 GPa, 121.4 GPa, 141.8 GPa respectively, as shown in figure 8. The addition of GNSs can reduce the coefficient of thermal expansion of the composite filler. The reduction of thermal expansion coefficient of composite filler can reduce the CTE mismatch between the brazing seam and the Ti$_3$Cu$_3$O reaction layer, and contribute to the improvement of the joint strength. Additionally, the addition of GNS can increase the Young’s modulus of the composite solder. When graphene content was 0.3 wt%, the Young’s modulus of the composite filler metal was also lower than both of Ti6Al4V and sapphire substrates. Therefore, the difference between the CTE, and the Young’s modulus of the composite solder, the reaction layer and the substrate is smaller when the graphene content is 0.1 wt%, resulting in less residual stress and greater joint strength. However, graphene agglomeration occurs in the brazed seam when the content of graphene exceeded 0.1%. Graphite agglomeration hinders the diffusion of elements and slows the growth of the reaction layer, reducing the strength of the joint. Even when the graphene content was 0.5%, the reaction layer was discontinuous and greatly reduced the strength of the joint. In conclusion, the exact amount of graphene can improve the shear strength of brazing joints.

Figure 9(a) shows the magnified microstructure of fracture surface of brazing joint. Two kinds of typical fracture characteristic (mark as 1 and 2) were observed at the surface. According to the EDS analysis listed in table 3. The zone ‘1’ could be identified as the Al$_2$O$_3$ phase and the GNSs (marked as A in figure 9(b)) while the zone ‘2’ should be the Ti-Cu phase (marked as B in figure 9(c)) and the GNSs (marked as C in figure 9(c)) near the sapphire substrate. In figure 9(b), we can see the obvious stepped cracks, which indicates that the cracks are derived from the interior of the sapphire during the shear process, and demonstrates a large residual stress between the sapphire and the composite solder [23]. In both Zone 1 and Zone 2, the presence of graphene was found and the distribution was relatively uniform. And it can be clearly seen that the fracture of zone 1 has a characteristic of toughness. Such a fracture mode indicates that graphene can effectively alleviate the residual stress between the composite solder and the sapphire. Therefore, the relief of residual stress further increases the shear strength of the joint.

### Table 2. Basic properties of substrates and composite filler [7].

| Materials | Density (g cm$^{-3}$) | $E$ (GPa) | $\alpha \times 10^{-6} K^{-1}$ |
|-----------|-----------------------|-----------|-------------------------------|
| TC4       | 4.5                   | 113       | 8.4                           |
| sapphire  | 3.98                  | 344       | 6.5                           |
| AgCuTi    | 9.48                  | 94        | 18.2                          |
| GNSs      | 1                     | 1000      | $-8.0 \pm 0.7$                |

![Figure 8. CTE and Young's modulus of the GNSs-AgCuTi composite filler with different GNSs content.](image)
4. Conclusions

Ti6Al4V alloy and sapphire were successfully brazed with GNSs-AgCuTi composite filler. This thesis studied the effect of GNSs content and brazing temperature on the microstructure and mechanical properties of the brazed joint, and the thermal expansion coefficient of graphene on composite brazing fillers. According to the research results, following conclusions can be drawn:

(1) The interface structure of Ti6Al4V/sapphire brazed joint is Ti6Al4V/Ti2Cu/TiCu/Ti3Cu4/Ag(s.s)/AgCuTi/Graphene/TiCu/Ag(s.s)/Ti3Cu5O/sapphire.

(2) The interface structure of the joint changes significantly with the brazing temperature. The dissolved titanium content increases as the temperature increases, and the diffusion layer (I) and Ti-Cu layer (II) got thickened gradually. Meanwhile, the thickness of Ti3Cu3O reaction layer increased from 1.9 μm to 4.7 μm at the brazing temperature from 810 °C to 870 °C.

(3) When the GNSs content in the composite filler increased, the microstructure of brazing seam became fine and homogeneous. Meanwhile, the diffusion layer (I) and Ti-Cu layer (II) reduced gradually. In addition, local agglomeration occurred when graphene content exceeded 0.1 wt%.

(4) The shear strength of the joint first increased and then decreased with the increase in the brazing temperature and GNSs content. The shear strength of the joint reaches to 29.1MPa with the composite filler containing 0.1 wt%GNSs at 860 °C for 10 min. The addition of graphene not only refines the interfacial structure, but also contributes to the release of residual stress.

Acknowledgments

Support by the National Natural Science Foundation of China (No.51474181) is much appreciated.

ORCID iDs

Zhou Fan https://orcid.org/0000-0002-0248-8631

References

[1] Nie H and Lu B 2005 Sapphire window and it’s application in military electro-optical equipment Ship. Electron. Eng. 25 131–133
[2] Yin S and Chu J 2010 Joining of sapphire dome and metal housing Aviat. Precis. Manuf. Tech. 46 54–57
[3] Wang G, Zuo H and Zhang H 2010 Preparation, quality characterization, service performance evaluation and its modification of sapphire crystal for optical window and dome application Mater. Des. 31 706–711
[4] Guo W, Wang T and Lin T 2018 Bismuth borate zinc glass brazing for bonding sapphire in air Mater. Charact. 137 67–76
[5] Fan Z, Liu J and Xiao H 2011 Research progress on growth technique and application of sapphire single crystal J. of the Chin. Ceram. Soc. 39 880–891
[6] Buhl S, Leinenbach C and Spolenak R 2010 Influence of the brazing parameters on microstructure, residual stresses and shear strength of diamond–metal joints J. of. Mater. Sci. 45 4358–4368
[7] Zhou Y, Liu D and Niu H 2016 Vacuum brazing of C/C composite to TC4 alloy using nano-\(\text{Al}_2\text{O}_3\) strengthened \(\text{AgCuTi}\) composite filler Mater. Des. 93 347–356
[8] Zhang J, Zhang L and Qi J 2013 Research on the interfacial structure and mechanical properties of \(\text{TC4-SiO}_2\) composite brazed joint Rare. Met. Mater. And. Eng. 42 2598–2601
[9] Shang J, Li N and Yan J 2013 Study on joining methods between sapphire and metals Weld. Join. 2013 45–48
[10] Buhl S, Leinenbach C and Spolenak R 2012 Microstructure, residual stresses and shear strength of diamond-steel-joints brazed with a \(\text{Cu–Sn}\)-based active filler alloy Intern. J. of. Refract. Hard. Mater. 30 16–24
[11] Yang Z, Lin J and Wang Y 2017 Characterization of microstructure and mechanical properties of \(\text{Al}_2\text{O}_3/\text{TiAl}\) joints vacuum-brazed with \(\text{Ag−Cu−Ti} + W\) composite filler Vac. 143 294–302
[12] Xin C, Li N and Yan J 2018 Effects of Ti Activity on Mechanical Properties and Microstructures of \(\text{Al}_2\text{O}_3/\text{Ag−Cu−Ti}/\text{Fe−Ni-Co}\) Brazed Joints Rare Met. Mater. Eng. 47 1031–1036
[13] Cui W, Li S and Yan J 2018 Microstructure and mechanical performance of composite joints of sapphire by ultrasonic-assisted brazing J. of. Mater. Process. Technol. 257 1–6
[14] Zaharinie T, Yusof F and Fadzil M 2015 Microstructural analysis of brazing sapphire and Inconel 600 for sensor applications Mater. Res. Innov. 18 56–58–70
[15] Yan S, Chen X and Hong Q 2016 Graphene reinforced aluminum matrix nanocomposites J. of. Aeronaut. Mater. 36 57–70
[16] Liu X, Wang H and Sun Q 2018 Research progress of graphene and 3D graphene composites Chem. Ind. Eng. Process. 37 168–174
[17] Ma Y, Li X and Zhou W 2017 Reinforcement of graphene nanosheets on the microstructure and properties of Sn38Bi lead-free filler Mater. Des. 113 264–272
[18] Wang Z, Ba J and Ma Q 2018 Research progress on nanomaterial reinforced composite brazing filler J. of. Netshape. Form. Eng. 10 82–90
[19] Cui B, Huang J and Xiong J 2013 Reaction-composite brazing of carbon fiber reinforced SiC composite and TC4 alloy using Ag-Cu-Ti (Ti + C) mixed powder Mater. Sci. and. Eng. A 562 203–210
[20] Yang Z, Zhang L and Ren W 2013 Interfaceal microstructure and strengthening mechanism of BN-doped metal brazed \(\text{Ti/SiO}_2\)-BN joints J. of. Eur. Ceram. Soc. 33 759–768
[21] Blugan G, Kuebler J and Bissig V 2007 Brazing of silicon nitride ceramic composite to steel using SiC-particle-reinforced active brazing alloy Ceram. Intern. 33 1033–1039
[22] Liu D, Song Y and Zhou Y 2018 Brazing of C/C composite and Ti-6Al-4V with graphene strengthened \(\text{AgCuTi}\) filler: effects of graphene on wettability, microstructure and mechanical properties Chin. J. of. Aeronaut. 31 1602–1608
[23] Qiu Q, Wang Y and Yang Z 2016 Microstructure and mechanical properties of \(\text{Al}_2\text{O}_3\) ceramic and \(\text{Ti6Al4V}\) alloy joint brazed with inactive Ag-Cu and Ag-Cu + B J. of. Eur. Ceram. Soc. 36 2067–2074
[24] Erelaefe A and Tillmann W 2009 Effect of brazing parameters on microstructure and mechanical properties of titanium joints J. of. Mater. Process. Technol. 209 4842–4849
[25] Voytovych R, Ljungberg L and Eustathopoulos N 2004 The role of adsorption and reaction in wetting in the CuAg: Ti/aluamina system Ser. Mater. 51 431–435
[26] Liang Y, Wang H and Yang Y 2008 Evolution process of the synthesis of TiC in the Cu–Ti–C system J. of. Alloy. And. Compd. 452 298–303.
[27] Zhou Q, Fu Z, Wang C and DFT A 2015 study of electronic and magnetic properties of titanium decorating point-defective graphite Appl. Surf. Sci. 356 1025–1031
[28] Zhang H, Luo X and Lin X 2013 Density functional theory calculations of hydrogen adsorption on Ti-, Zn-, Zr-, Al-, and N-doped and intrinsic graphene sheets Intern. J. of. Hydrog. Energ. 38 14269–14275
[29] Kelkar G, Spear K and Carm A 1994 Thermodynamic evaluation of reaction products and layering in brazed alumina joints J. of. Mater. Res. 9 2244–2250
[30] Ning H, Geng Z and Ma J 2003 Joining of sapphire and hot pressed \(\text{Al}_2\text{O}_3\) using Ag70.5Cu27.5Ti2 brazing filler metal Ceram. Intern. 29 689–694
[31] Niu G, Wang D and Yang Z 2017 Microstructure and mechanical properties of \(\text{Al}_2\text{O}_3/\text{TiAl}\) joints brazed with B powders reinforced Ag–Cu–Ti based composite fillers Ceram. Intern. 43 439–450
[32] Tao L, Chang S and Lee C 2010 Effects of nano-\(\text{Al}_2\text{O}_3\) additions on microstructure development and hardness of \(\text{Sn}_3\text{Ag}_3\text{Cu}\) solder Mater. Des. 31 4831–4835
[33] Meyer J, Geim A and Katsnelson M 2007 The structure of suspended graphene sheets Nat. 446 60–63
[34] Gleiter H 2000 Nanostructured materials: basic concepts and microstructure Acta Mater. 48 1–29
[35] Li C, Si X and Chen L 2019 Non-destructive measurement of residual stress distribution as a function of depth in sapphire/\(\text{Ti6Al4V}\) brazing joint via Raman spectra Ceram. Intern. 45 3284–3289
[36] Yang J, Ji S and Fang H 2006 Theoretical study and numerical simulation of the stress fields of the \(\text{Al}_2\text{O}_3\) joints brazed with composite filler materials Chin. Weld. 15 74–78