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Integrated filler metal/base metal manufacturing via cold spray additive manufacturing for brazing C_f/SiC and superalloy

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ABSTRACT

A new integrated filler metal/base metal manufacturing method by cold spray additive manufacturing is proposed. The integrated CuTi filler metal/GH3536 and CuTi + W composite filler metal/GH3536 are prepared by cold spray additive manufacturing techniques. The large plastic deformation of Cu and Ti particles and the tamping effect of W particles promote the interfacial bonding of particles, which improves the weldability of cold sprayed CuTi + W composite filler metals. Based on the cold sprayed CuTi + W composite filler metal, the C_f/SiC composites and GH3536 are successfully brazed, and the typical microstructure and brazing mechanism are investigated. As a result, the shear strength of C_f/SiC-GH3536 joint brazed by cold sprayed CuTi + W composite filler metal reaches 77 MPa. This study highlightes the great potential of cold spray additive manufacturing for integrated filler metal/base metal manufacturing in brazing.

1. Introduction

C_f/SiC composite as a structural material has shown great potential for applications in hot end components of aerospace engines [1,2]. C_f/SiC composite is difficult to be processed into complex components due to its brittleness and expensive price, so it needs to be combined with superalloys to form complex components to use. In practical applications, the connection of C_f/SiC composites and superalloys is usually achieved by bolting, which offsets the weight reduction advantages of C_f/SiC composites [3]. Active metal brazing is an effective method to achieve the connection of ceramic and metal materials. However, due to the different physical properties of C_f/SiC composites and superalloys, especially in the coefficient of thermal expansion, there is a large residual stress in the ceramic and metal brazed joint [4]. Therefore, it is important to develop a new method to achieve reliable connection of C_f/SiC composites and superalloys.

Currently, the traditional composite filler metals manufacturing methods include powder filler metals or foil filler metals. In order to alleviate the high residual stress, the ceramic or metal additives with low thermal expansion coefficient (CTE) are used to reduce the CTE of composite powder filler metals [5,6]. Song et al. [7] employed graphene nanoplates reinforced AgCuTi composite powder filler metal to braze SiC ceramic and $Al_{0.3}$CoCrFeNi high entropy alloy. When adding 0.3 wt% graphene nanoplates, the residual stress of brazed joint was effectively alleviated and the mechanical properties of the joint was significantly improved. On the other hand, metal or porous ceramic interlayer was used to release the high residual stress of the brazed joints [8,9]. Zhang et al. [10] brazed SiC_f/SiC and GH536 by assembling AgCuTi/Cu foam/AgCuTi foil filler metal. The results show that the Cu foam can enhance the shear strength of the joint by relieving residual stress. Although these methods relieve residual stress in ceramic/metal joints, they also increase the complexity of brazing process. A new method for both relieving residual stress in ceramic/metal joints and simplifying the brazing process is still urged to be developed.

Cold spraying is a new solid-state additive manufacturing technology, which is widely used to prepare pure metals, alloys and ceramic/metal composites [11,12]. In the cold spraying process, micro-sized particles are accelerated to supersonic speed by high-speed gas through the Laval nozzle, then the high-speed particles hit the substrate and deposit to form a coating. Due to the large plastic deformation of particles caused by ultra-high-speed collisions, cold spraying can achieve solid phase additive manufacturing at temperatures below their melting point. For active

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metal brazing, melting the active filler metal before welding will lead to metallurgical reaction inside the filler metal to reduce the active element content, thereby reducing the weldability of the filler metal [13]. However, most additive manufacturing techniques achieve deposition by melting the metal, and these methods are unsuitable for the additive manufacturing of active filler metal [14]. Therefore, cold spray additive manufacturing technology provides a unique method for manufacturing composite filler metals.

In this work, a new method for integrated filler metal/base metal manufacturing by cold spray additive manufacturing is presented. The CuTi and CuTi + W filler metal systems were selected as active brazing fillers for brazing C_f/SiC and GH3536 due to their excellent weldability and plasticity. Compared to the traditional manufacturing methods, such as powder filler metal and foil filler metal, the integrated filler metal/base metal manufacturing can not only relieve the residual stress of the joint but also simplify the brazing process.

The assembly method of brazing joint has been simplified from three-component assemblies (C_f/SiC-powder filler metal-superalloy, Fig. 1a) or five-component assemblies (C_f/SiC-foil filler metal-network interlayer-foil filler metal-superalloy, Fig. 1b) to two-component assemblies (C_f/SiC-integrated filler metal/superalloy, Fig. 1c). Here, cold sprayed CuTi (CS-CuTi) and cold sprayed CuTi + W (CS-CuTi + W) composite filler metals coatings were successfully prepared on the surface of GH3536 by cold spray additive manufacturing technology. The interfacial bonding mechanism of CuTi + W composite filler metals coating was clarified by microstructure characterization. The microstructure evolution and interfacial reaction mechanism of C_f/SiC-GH3536 joint brazed by CS-CuTi + W composite filler metals was investigated.

2. Experimental procedure

The C_f/SiC composite fabricated by polymer infiltration pyrolysis technology and commercial Ni-based superalloy GH3536 was used in this study. The spherical Cu, Ti and W powders with particle size of 15~53 μm were used to prepare filler metal. Before cold spraying, the GH3536 substrate was cut to 50 mm × 50 mm × 3 mm and then the surface was shot peened. During cold spraying process, nitrogen at 600 °C was used as the carrier gas with a gas pressure of 3 MPa. The standoff distance between the nozzle and the substrate was 15 mm, with a substrate traverse speed of 50 mm/s. The CS-CuTi filler metal was prepared by mixing the spherical Cu and Ti powders with atomic ratio of 85:15. The CS-CuTi + W composite filler metals were prepared by Cu, Ti and W powders with atomic ratio of 68:12:20. Before brazing, the CS-CuTi/GH3536 and CS-CuTi + W/GH3536 were cut to 10 mm × 10 mm × 3 mm, and then the surface was polished. As shown in Fig. 1c, the C_f/SiC composite and composite filler metals/GH3536 were assembled, then the workpieces were transferred to a vacuum furnace. The brazing process was performed at 1140 °C for 20 min. The microstructure characterization was analyzed by scanning electron microscopy (SEM). The element mapping distribution was performed by energy dispersive spectroscopy (EDS) in a transmission electron microscope (TEM). The focused ion beam (FIB) technology was used to prepare the TEM samples. Furthermore, the selected area electron diffraction (SAED) and high-resolution transmission electron microscope (HRTEM) were employed to analyze the phase composition. The shear strength test of the brazed joint was carried out using the INSTRON 3382 universal testing machine with a load speeding of 0.5 mm/min, as described in our previous work [15].

3. Results and discussion

As shown in Fig. 2a–c, the spherical Cu and Ti powders and irregular shape of W powder with a size of 300 mesh was employed. In Fig. 2d, the CS-CuTi filler metal coating with an area of 35 mm × 50 mm was successfully prepared on the GH3536 substrate by cold spray additive manufacturing. The SEM image of CS-CuTi/GH3536 indicates that the CS-CuTi coating has a high density. The gray phase and light gray phase are the Ti and Cu phases, respectively. The interface of CS-CuTi filler metal coating and GH3536 shows a partial interface combination. The digital photograph and SEM image of CS-CuTi + W/GH3536 are shown in Fig. 2e. The CS-CuTi + W composite filler metals coating exhibits a continuous dense microstructure. No obvious interface defects and cracks were observed at the interface. The white W phase and gray Ti phase were evenly distributed inside the Cu matrix. Due to the high impact velocity, the Cu and Ti particles underwent large plastic deformation. The Cu and Ti particles underwent significant shape changes after a strong impact, resulting in tighter interfacial connections between the particles. Compared to CS-CuTi, the interface of CS-CuTi + W and GH3536 shows a better interface combination, indicating that the W particles have a tamping effect on Cu matrix.

To investigate the interfacial microstructure of CS-CuTi + W composite filler metals coating, the sample at Cu, Ti and W interface was extracted by FIB technology. As shown in Fig. 2f–i, the W particle had a tight combination with Ti particle and a partial combination with Cu particle. As shown in Fig. 2j, the HRTEM image of Cu particle was exhibited. The lattice fringe of the selected area of Cu particle (Fig. 2j) obtained by inverse fast Fourier transformation (FFT) is shown in Fig. 2k. The interplanar spacing of Cu particles was 0.209 nm, corresponding to the (111) plane of Cu. Furthermore, there appear many dislocations marked with T symbol in Cu particles, indicating that the high impact velocity leads to large plastic deformation of Cu particles. As shown in Fig. 2j_1, the diffraction pattern corresponding to the [011] zone axis of Cu is shown. Fig. 2l. Fig. 2l_1 shows the HRTEM image and the diffraction pattern of the $[2\bar{4}2\bar{4}]$ zone axis of the Ti particle. The FFT image (Fig. 2m) of the Ti particle indicates the lattice fringe with an interplanar spacing of 0.253 nm, which belongs to the (100) plane of Ti. During the cold spraying, a large number of dislocations were also observed inside the Ti particles. However, the high hardness of W particle led to weak deformation ability. Therefore, the Cu and Ti particles had large plastic deformation under high-speed impact. Furthermore, the high density of W particles (19.35 g/cm^3) can obtain higher kinetic energy during cold spraying [16], resulting in a tamping effect on the Cu matrix, thus making a tighter interfacial bonding of particles. The tighter interfacial bonding between W particles and CuTi filler was beneficial to the dispersion of W particles in the brazing seam, improving the interfacial wettability and

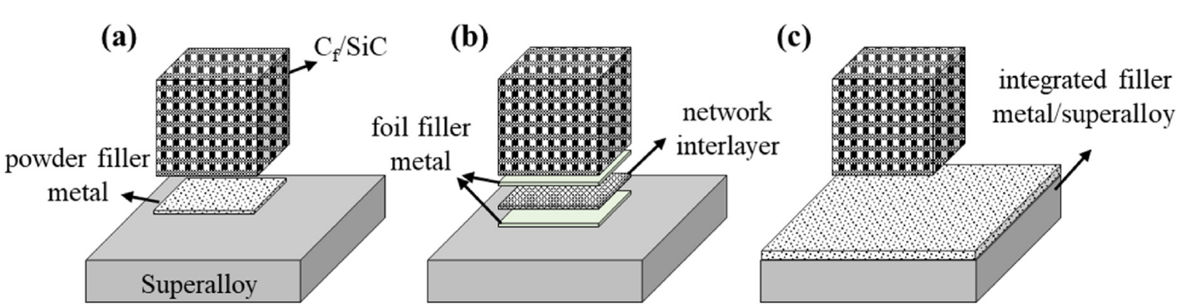

Fig. 1. The different composite filler metal manufacturing method: (a) powder metal filler, (b) foil filler metal, and (c) integrated filler metal/superalloy.

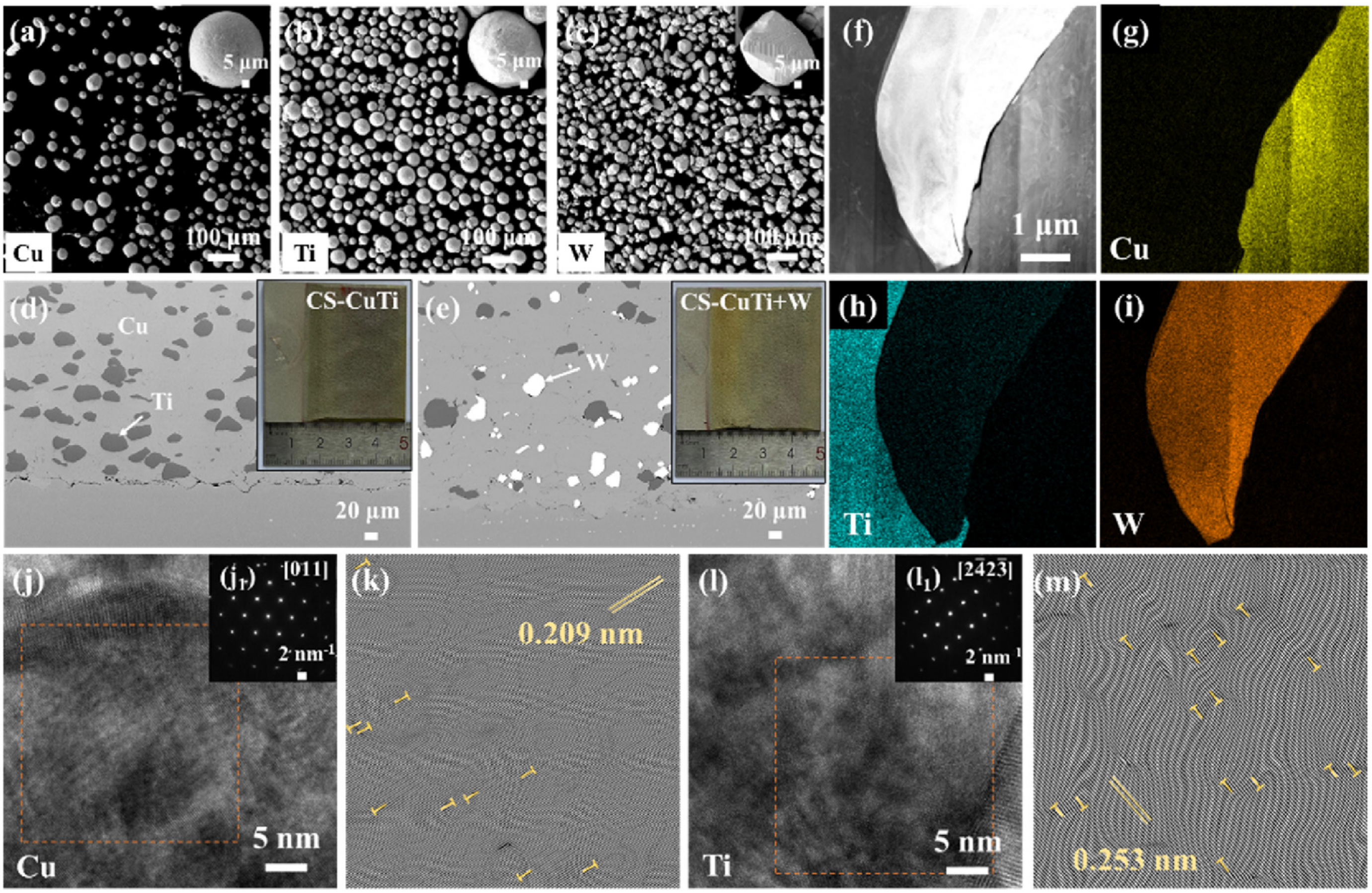

Fig. 2. SEM images of (a) Cu powder, (b) Ti powder and (c) W powder, (d) digital photograph and SEM image of CS-CuTi/GH3536, (e) digital photograph and SEM image of CS-CuTi + W/GH3536, (f) TEM image, (g–i) element mapping of Cu, Ti and W interface, (j) HRTEM image, (j_1) SAED pattern and (k) IFFT image of Cu particle, (l) HRTEM image, (l_1) SAED pattern and (m) IFFT image of Ti particle.

avoiding the formation of brazing defects, which effectively improves the weldability of CS-CuTi + W composite filler metals.

Fig. 3a shows the microstructure of the C_f/SiC-GH3536 joint brazed by the CS-CuTi filler metal. The C_f/SiC composite and GH3536 were successfully brazed by the CS-CuTi filler metal. No obvious defects and cracks were observed at the brazed joint. Fig. 3b exhibits the microstructure of the C_f/SiC-GH3536 joint brazed by the CS-CuTi + W composite filler metal. The W particles were evenly distributed in the brazing seam. As shown in Fig. 3c, the W particles reacted with CuTi filler metal, forming a gray phase around the W particles. As shown in Fig. 3d, the brazing seam was closely connected to the C_f/SiC composite by an interfacial reaction layer. An obvious diffusion zone was observed at the interface of GH3536 and brazing seam. In order to clarify the phase compositions of the interfacial reaction layer, the TEM sample of the interfacial reaction layer was extracted by FIB technology. Details of the microstructures of the interfacial reaction layer using bright-field TEM and HRTEM are exhibited in Fig. 3e and f. The interfacial reaction layer was composed of $Cr_{23}C_6$ and TiC, and the fine TiC phase was observed at the interface of $Cr_{23}C_6$ and TiC. The SAED result in Fig. 3g belongs to the [011] zone axis of TiC phase, and the SAED result in Fig. 3h is due to the [001] zone axis of $Cr_{23}C_6$ phase. The element distribution mapping of the interfacial reaction layer is shown in Fig. 3j-p. The zone I was mainly composed of Cu element, belonging to Cu (*s, s*) phase. The zone II was primarily composed of Cr, Ti and C elements, belonging to $Cr_{23}C_6$ and TiC phases. The zone III was mainly composed of Ni and Si elements. The SAED result of zone III is presented in Fig. 3i, and the diffraction pattern belongs to the Ni_3Si phase. According to the above results, the $Cr_{23}C_6$ and TiC phases were attributed to the reaction of active Ti elements and C_f/SiC composites. Furthermore, the formation of Ni_3Si phase was attributed to the reaction of Ni elements and C_f/SiC composites. Therefore, the interfacial reaction layer of C_f/SiC composite and brazing seam were composed of $Cr_{23}C_6$, TiC and Ni_3Si phases.

Fig. 4a shows the microstructure of the interfacial reaction layer around the W particle using bright-field TEM. The interfacial reaction layer around the W particle consists of three regions: zone IV, zone V, and zone VI. The zone IV was mainly composed of Cu element, belonging to the Cu (*s, s*) phase. The zone V was mainly composed of Cu, W, Fe, Cr and Ni elements, belonging to the Cu-W-Fe-Cr-Ni multi-principal element alloy, which was formed by the reaction of Cu, and W elements in CS-CuTi + W with Fe, Cr and Ni elements in GH3536. The diffraction patterns result of zone V using SAED are shown in Fig. 4h and i, belonging to [113] zone axis of face-centered cubic (FCC) Cu-W-Fe-Cr-Ni multi-principal element alloy and [012] zone axis of body-centered cubic (BCC) Cu-W-Fe-Cr-Ni multi-principal element alloy respectively. Therefore, the zone V was composed of Cu-W-Fe-Cr-Ni multi-principal element alloy with BCC and FCC crystal structure. The zone VI was mainly composed of Cu and W elements. Combining with the SAED analysis in Fig. 4j, the diffraction pattern at zone VI belonged to [111] zone axis of W. Furthermore, a solid solution can be formed between Cu and W elements [17], so zone VI consists of W(Cu) solid solution. Therefore, the interfacial reaction layer around W particles is composed of Cu-W-Fe-Cr-Ni multi-principal element alloy and W(Cu) solid solution. The W particles with a low-CTE help to reduce thermal mismatch between C_f/SiC composites and brazing seam, thereby relieving residual stress and improving mechanical properties of brazed joints.

In addition, the Cu-W-Fe-Cr-Ni multi-principal element alloy shows a superior mechanical property [18], which is helpful for preventing crack propagation and improving mechanical properties of brazed joints. To

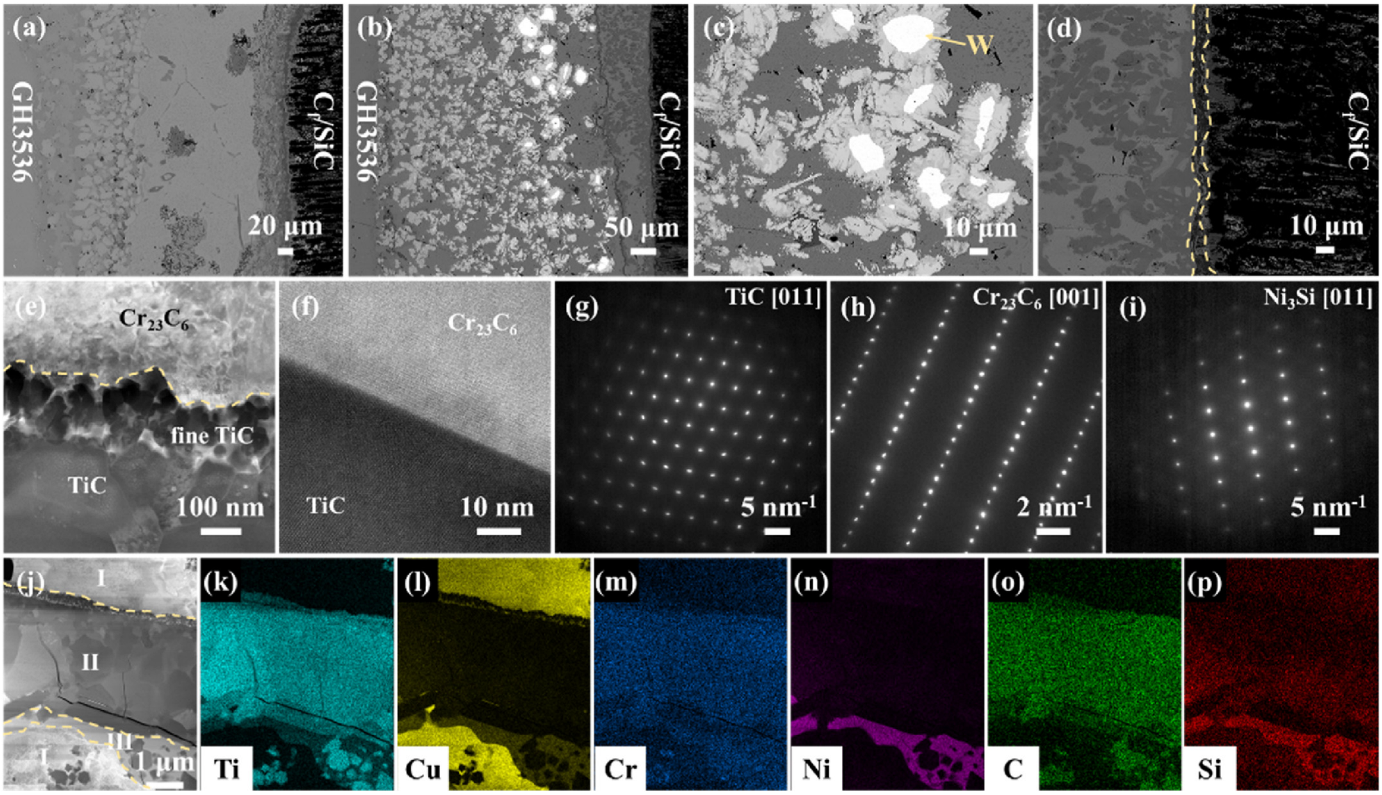

Fig. 3. (a) SEM image of C_f/SiC-GH3536 joint brazed by CS-CuTi, (b) SEM image of C_f/SiC-GH3536 joint brazed by CS-CuTi + W, (c) the enlarged image of W particles from Fig. 4b–(d) the enlarged image of interfacial reaction layer from Fig. 4b–(e) the bright-field TEM image and (f) HRTEM image of the interface of $Cr_{23}C_6$ and TiC, the SAED results of (g) TiC [011], (h) $Cr_{23}C_6$ [001] and (i) Ni_3Si, (j–p) the element mapping of the interfacial reaction layer.

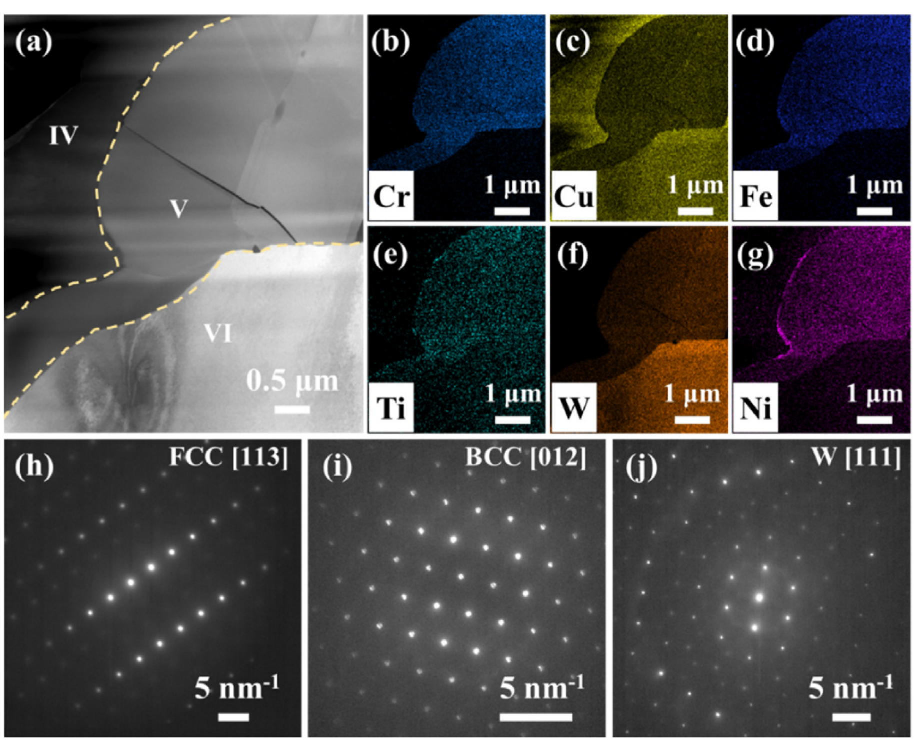

Fig. 4. (a) The bright-field TEM of the interfacial reaction layer around W particle, (b–g) the element distribution mapping, the SAED results of (h) FCC-MPEA [113], (i) BCC-MPEA [012] and (j) W [111].

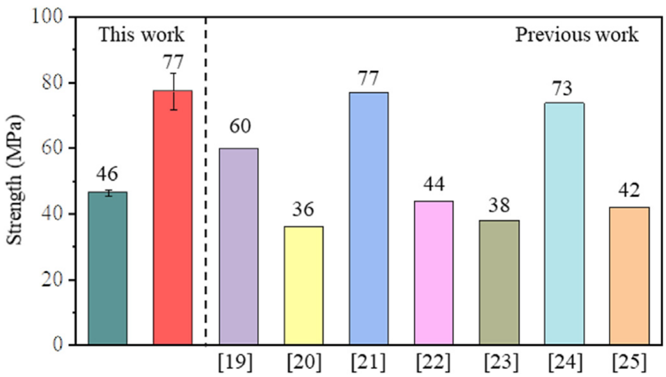

Fig. 5. The comparison of joint strength between this work and previous works [19–25].

evaluate the shear strength of the brazed joint, the shear strength of the C_f/SiC-GH3536 joint brazed by CS-CuTi and CS-CuTi + W was tested. As shown in Fig. 5, the shear strength of C_f/SiC-GH3536 joint brazed by CS-CuTi was 46 MPa. Furthermore, the maximum shear strength of C_f/SiC-GH3536 joint brazed by CS-CuTi + W was 77 MPa, which is 1.7 times higher than that of CS-CuTi composite filler metal. As discussed on the microstructure optimization, introducing W particles can promote the formation of Cu-W-Fe-Cr-Ni multi-principal element alloy. In addition, the low-CTE W particles are beneficial to reduce the thermal mismatch, relieving the high residual stress in C_f/SiC-GH3536 joint. For comparison, Fig. 5 exhibits the result of the previous works on the shear strength of ceramic-metal joint, including SiC-metal joint, C_f/SiC-metal joint, SiC_f/SiC-metal joint. Compared to the previous work about ceramic-metal joint, the joint brazed by CS-CuTi + W in this work exhibits a competitive joint strength.

4. Conclusions

In summary, we have proposed an integrated filler metal/base metal manufacturing by cold spray additive manufacturing. The CS-CuTi and CS-CuTi + W filler metal coatings were successfully fabricated on the surface of GH3536 substrate using cold spray additive manufacturing technology. Combined to the microstructure analysis, the large plastic deformation of Cu and Ti particles and the tamping effect of W particles promote the interfacial bonding, improving the wettability of CS-CuTi + W composite filler metals. Furthermore, the C_f/SiC composite and GH3536 were successfully brazed by CS-CuTi and CS-CuTi + W composite filler metals. The interfacial reaction layer of C_f/SiC composite and brazing seam were composed of $Cr_{23}C_6$, TiC and Ni_3Si. The W particles reacted with active Ti to form Cu-W-Fe-Cr-Ni multi-principal element alloy with BCC and FCC crystal structure and W(Cu) solid solution. Finally, the shear strength of C_f/SiC-GH3536 joint brazed by CS-CuTi + W composite filler metals reaches 77 MPa. This work proposes a novel strategy for integrated filler metal/base metal manufacturing by cold spray additive manufacturing, which broadens a new way for designs of composite filler metals.

CRediT authorship contribution statement

Pengcheng Wang: Writing – original draft, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Weihan Liu:** Methodology, Investigation, Data curation. **Lei Gu:** Methodology, Investigation. **Hao Ran:** Writing – review & editing, Methodology, Investigation. **Xiaoguo Song:** Writing – review & editing, Project administration. **Zhaoyi Pan:** Visualization, Resources. **Haiyan Chen:** Writing – review & editing, Validation, Project administration, Funding acquisition, Data curation. **Wenya Li:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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