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Strengthening the ZrC-SiC ceramic and TC4 alloy brazed joint using laser additive manufactured functionally graded material layers

J.M. Shi, L.X. Zhang*, Q. Chang, Z. Sun, J.C. Feng

State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Harbin 150001, China

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ABSTRACT

In order to improve the ZrC-SiC and TC4 brazed joint property, functionally graded material (FGM) layers (two SiC particles reinforced TC4-based composite layers) were designed to relieve the residual stress in the ZrC-SiC and TC4 brazed joint. The FGM layers were fabricated on the TC4 surface using laser additive manufacturing technology before the brazing. Then the TC4 coated with the FGM layers and ZrC-SiC ceramic were brazed using Ti-15Cu-15Ni (wt%) filler. According to the SEM and TEM results, the volume fractions of SiC particles in the FGM layers could reach 20% and 39% respectively. Ti from the braze filler and TC4 reacted with the ZrC-SiC ceramic to form TiC and Ti_5Si_3 adjacent to the ZrC-SiC ceramic. The shear test results indicate that the adoption of the FGM layers and the brazing temperature both affected the joint property significantly. The FGM layers could benefit the mitigation of coefficient of thermal expansion (CTE) mismatch between the ZrC-SiC and TC4, so that the residual stress caused by the CTE mismatch in the joint was relieved and the joint strength increased. The brazing temperature would affect the microstructure of the brazing seam and then control the joint strength. When the ZrC-SiC ceramic and TC4 coated with the FGM layers were brazed at 970 °C for 10 min, the maximum shear strength could reach 91 MPa, and cracks propagated in the ZrC-SiC ceramic substrate during the shear test.

1. Introduction

ZrC-SiC ceramic (named as ZrC-SiC for convenience) has attracted considerable interest in recent years primarily due to its combination of remarkable thermal and electrical conductivity, excellent oxidation resistance, and good high temperature performance [1–5]. This ceramic is an attractive candidate for high temperature thermo mechanical structural applications. However, the hardness and brittleness of the ceramic make it difficult to obtain large-sized or complex components. Therefore, joining the ZrC-SiC to itself or metallic materials is meaningful to satisfy its application. Ti6Al4V alloy (named as TC4 for convenience), which possesses super plasticity, low density and high specific strength, is widely used in aerospace industry [6,7]. Achieving the joining of the ZrC-SiC and TC4 can take advantage of respective superiorities of ceramic and metal, and extend their scopes of application.

Among the joining methods, brazing is suitable to obtain robust and high quality ceramic/metal joints due to its simplicity, lower cost investment and so on [8–11]. In the ceramic/metal joint, the residual stress caused by the CTE mismatch between the ceramic and metal would produce stress concentration in the ceramic and deteriorate the joint strength significantly [12,13]. Thus, designing the joint structure and relieving the residual stress in the joint are meaningful to the property and reliability of the joint. Yang et al. used Cu foil to relieve the residual stress of SiO_2-BN/Invar joint through the plastic deformation of Cu [14]. But, the joint cannot serve at high temperature due to the low strength of Cu based solid solution (named as Cu(s.s) for convenience) at high temperature. Shen et al. brazed C/C composite and Ni-based superalloy using Al_2O_3 interlayer to relieve the residual stress [15]. The Al_2O_3 interlayer benefited the mitigation of the CTE mismatch between C/C composite and Ni-based superalloy. The joint strength increased significantly, but the brittleness of Al_2O_3 layer and the complex assembly of the joint limited the use of this method. Song et al. used AgCuTi-Si_3N_4 composite filler to braze Si_3N_4 ceramic and TiAl [16]. The Si_3N_4 in the filler decreased the CTE of the filler and relieved the residual stress in the joint. However, the addition of Si_3N_4 into the braze filler would decrease the flowability of the filler and induced defects in the brazing seam because of the aggregation of Si_3N_4 particles.

The researches on the residual stress in the joint show that functionally graded material (FGM) layers can be used to relieve the residual stress in the joint [17,18]. The FGM layers could make the CTE change gradually, minimizing the thermal stresses generating during the brazing process. Comparing with the common methods, the adoption of the FGM layers to relieve the residual stress has three

* Corresponding author.
E-mail address: hitzhanglixia@163.com (L.X. Zhang).

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advantages. Firstly, the composition of the FGM layers could be designed and the joint could serve at high temperature. Secondly, the FGM layers are usually metal/ceramic composite materials and they can exhibit high toughness and strength. Finally, the FGM layers are usually fabricated before the brazing process, so the volume fraction of ceramic in the FGM layers could be high to relieve the residual stress maximally. However, in the reported researches [17,18], the FGM layers were usually fabricated by hot pressing method and the fabrication process was complex. Thus, developing the convenient fabrication method of the FGM layers is necessary to realize the use of the FGM layers in brazed joint.

In order to fabricate the FGM layers with high property on the TC4 surface efficiently, the fabrication technology and reinforcement in the FGM layers should be considered scientifically. Firstly, laser additive manufacturing technology is a new method to fabricate the FGM layers. Li et al. obtained functionally graded TiCp/Ti6Al4V composite using laser additive manufacturing technology [19]. Through controlling the additive composition, the volume fractions of TiCp in the composite are graded. Lin et al. deposited the 316L stainless steel and Rene88DT superalloy mixed powders on 316L stainless steel substrate [20]. The composition of the mixed powders changed linearly from 0% to 100% Rene88DT, making the composition and phases graded in the deposited specimen. Secondly, the investigation on the TC4 surface modification shows that SiC particles are usually injected into the TC4 surface to fabricate composite layers, and the bond between the SiC particles and TC4 is strong due to the perfect compatibility between the SiC particles and TC4 [21,22]. In addition, the CTE of SiC ($4.7 \times 10^{-6}\ K^{-1}$) is much lower than TC4, indicating that the SiC particles in the FGM layers could change its CTE significantly. Therefore, we plan to fabricate two kinds of SiC particles reinforced TC4 layers (FGM layers) on the TC4 surface using laser additive manufacturing technology before the brazing. Then the FGM layers could relieve the residual stress in the ZrC-SiC and TC4 coated with the FGM layers brazed joint during the brazing process.

In the present paper, the SiC particles reinforced TC4-based FGM layers were fabricated using laser additive manufacturing technology. To ensure that the joint could serve at high temperature, Ti-15Cu-15Ni was adopted to braze the ZrC-SiC and the TC4 coated with the FGM layers. The interfacial microstructure and the shear strength of the joint were investigated in details. Furthermore, fracture behavior of the joint and the strengthening mechanism of the joint by the FGM layers were studied.

2. Experimental procedures

ZrC-SiC (the volume fractions of ZrC and SiC were 80% and 20% respectively) used in the experiments was processed by hot pressing the ZrC (mean particle size 2 μm, > 99 wt% purity, Changsha Wing High High-Tech New Materials Co., Ltd., China) and SiC (β-SiC, mean particle size 2 μm, > 99 wt% purity, Central Iron & Steel Research Institute, China) mixed powders. The hot pressing was carried out at 2000 °C for 60 min under the pressure of 30 MPa. The relative density of the prepared ZrC-SiC was 97%. The microstructure of the ZrC-SiC is shown in Fig. 1. The ZrC-SiC was sliced into 5 mm × 3 mm × 3 mm pieces for the metallographic observation and shear test.

The experimental set-up for laser additive manufacturing process included a laser source, powder feeding system, robot system and cooling system. DL028Q semiconductor laser equipment bought from Rofin-Baasel China Co., Ltd. was used to produce laser beam in our study. KUKA60 robot system bought from KUKA Robotics China Co., Ltd. was integrated with laser system in experiment. The powder feeding process was achieved by a powder feeder produced by Xinsong company. The FGM layers are two-layer structure, the Layer I adjacent to the TC4 substrate and the Layer II away from the TC4 substrate. The laser additive manufacturing process is shown in Fig. 2. Before the laser additive manufacturing process, the TC4 plate was cleaned

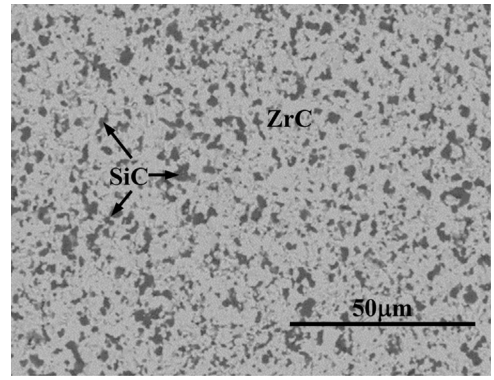

Fig. 1. Microstructure of the ZrC-SiC.

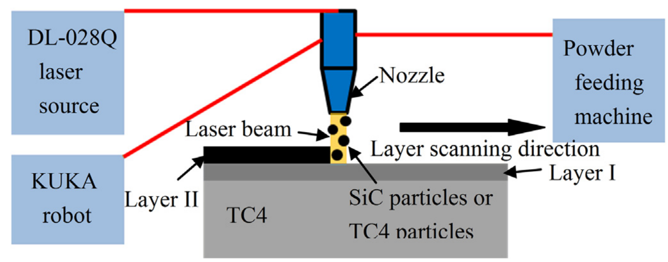

Fig. 2. The schematic of the laser additive manufacturing process.

ultrasonically in acetone for 20 min. Then the TC4 plate surface was melt to form a molten pool by semiconductor laser beam (laser wavelength: 808 nm and 940 nm, laser power: 1100 W, laser scanning speed: 6 mm/s), and the SiC particles (mean particle size 50 μm, > 99 wt% purity) were coaxially injected into the molten pool (powder feeding rate: 3 g/min). The Layer I would form after the solidification of the molten pool. In the end, the Layer I melt again under the laser beam (laser wavelength: 808 nm and 940 nm, laser power: 800 W, laser scanning speed: 3 mm/s) and the mixed particles of TC4-60 vol%SiC were coaxially injected into the molten pool (powder feeding rate: 5 g/min), then the Layer II could be obtained. In our experiment, the composite layers are single-pass additive manufacturing layers and the scanning pattern is straight line with the length of 70 mm.

In this study, the self-prepared Ti-15Cu-15Ni (wt%) filler (marked as TiCuNi for convenience in the following paragraphs) was used to braze the base materials. In the TiCuNi filler, TiH_2 powders were applied to replace Ti to avoid the oxidation during mechanical milling. The TiH_2 (mean particle size 50 μm, > 99.0 wt% purity), Cu (mean particle size 50 μm, > 99.0 wt% purity) and Ni (mean particle size 50 μm, > 99.0 wt% purity) mixed powders were milled for 1 h using planetary ball mill. Subsequently, the mixed powders weighted about 0.2 g were pressed at the pressure varying from 75 to 150 MPa into 10 mm diameter sheet specimens. Before brazing, the surface to be brazed were polished by 800 grit paper, and then ultrasonically cleaned in acetone for 5 min. Then the TiCuNi filler was placed between the ZrC-SiC and the TC4 coated with the FGM layers, as shown in Fig. 3(a). The research about the TiCuNi filler shows that its melting point was about 934 °C [23], so the brazing temperature should be higher than 934 °C. In addition, the β transus temperature of TC4 was about 1005 °C [24], and the TC4 property decreased significantly when the temperature was higher than 1005 °C. Therefore, the brazing experiments were conducted at 950–990 °C for 10 min in a vacuum furnace with the vacuum level up to 5×10^{-3} Pa. During the heating process, the temperature held at 650 °C for 120 min and 800 °C for 30 min to remove the residual stress in the FGM layers generated in the laser additive manufacturing process. A small pressure of 1.5 KPa was applied to prevent the specimen from moving when the braze filler melted.