

JOINING OF SiC CERAMIC TO Ni-BASED SUPERALLOY WITH Cu/Ta/Cu MULTIPLE INTERLAYER⁺

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ABSTRACT: Joining of ceramics to metals is of great interest from both technical and economical points of view. Welding of SiC ceramic to Ni-based superalloy has been realized using Cu/Ta/Cu multiple interlayer. The obtained bonding strength of the welded sample is 45.4% of that of SiC ceramic measured under the identical conditions. The effect of welding temperature, duration to keep the temperature, pressure for welding and thickness of Cu interlayer contacting with SiC ceramic on bending strength of the welded samples was studied. The mechanism of bonding was investigated based on observation of microstructure and determination of distribution of the elements at the welded area. The experimental results indicate that interdiffusions take place at the interfaces and contribute to bonding at the interfaces. The function of the multiple interlayer was discussed. Cu interlayer can effectively relax the welding thermal stress by plastic deformation. Ta interlayer can improve the distribution of residual thermal stress.

KEYWORDS: Ceramic/Metal Joining, Diffusion Welding, Multiple Interlayer, Joining Process

INTRODUCTION

Ni-based superalloys are widely used to manufacture the parts of aeronautical turbine serving at elevated temperatures. The working temperature of these materials can reach 950°C (type GH128) or higher. But, it is still not high enough for the next new turbines. In order to further increase the working temperature and decrease the weight of turbines, new high temperature structure materials are being expected and investigated. Among them, SiC ceramic and SiC matrix composite are promising ones. However, one problem arising with the application of SiC is joining of the ceramic to metals, particularly Ni-based superalloys. As indicated by Reference [1], the application of ceramic materials normally depends on the joining of ceramics to metallic structures, as we are living in a technological world based on

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the application of metals.

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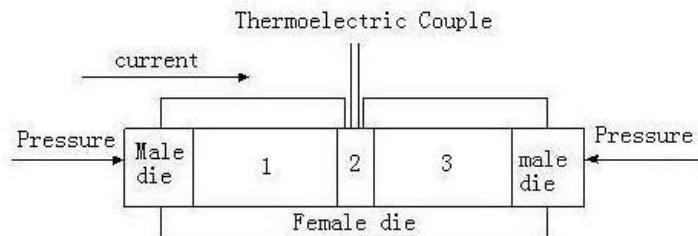
The structure and properties, especially the coefficient of linear thermal expansion (CTE) of SiC ceramic are quite different from those of Ni-based superalloy. The CTE of SiC ceramic within the temperature range of 0~1000°C is $4.7 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ [2], but the CTE of GH128 Ni-based superalloy within the temperature range of 18~1000°C is $16.29 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ [3]. Hence, the direct joining of these two materials will generate concentration of thermal stress during welding and cooling. The residual thermal stress may cause generation of microcracks leading to low welding strength and even failure of welding. In order to solve this problem, diffusion welding using Cu/Ta/Cu multiple interlayer was experimentally studied. Since copper is a soft metal with good plasticity, it is promising that the presence of Cu layers could relax the thermal stress by plastic deformation. As indicated in Reference [4], using tungsten intermediate layer, as a hard layer with low CTE, can effectively improve distribution of thermal stress of the joints and reduce concentration of residual thermal stress in the ceramic near welding seam. Taking into account that the density of tungsten (19.3 g/cm^3) is too high for aeronautical application, tantalum layer was used as a hard layer in this study. Since the CTE of tantalum is $6.5 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ [5], close to that of SiC, the presence of Ta layer could improve distribution of thermal stress of the joints. The process of welding and the microstructure of the joints have been investigated.

EXPERIMENTAL PROCEDURES

The commercial recrystallized SiC ceramic with the density of 2.65 g/cm^3 , the porosity of 15~16% and the purity of >99wt% was used as the starting ceramic to be welded. The forged Ni-based superalloy (type GH128) was used as the starting metallic material to be welded. This alloy contains Cr(19.0~22.0wt%), W(7.5~9.0wt%), Mo(7.5~9.0wt%), Ti(0.4~0.8wt%), Al(0.4~0.8wt%) and other elements with low contents.

The thickness of the original copper sheets was 0.2mm and 0.1mm. The soft layers with various thickness were made by piling up the original copper sheets. The thickness of tantalum sheet was 0.2mm. The copper sheet and tantalum sheet were ground and polished, then washed in alcohol with an ultrasonic bath for 15 minutes.

The SiC ceramic and the Ni-based superalloy were machined to cylindrical billets with the size of $\Phi 10 \text{ mm} \times 25 \text{ mm}$. The surfaces for welding were polished. The SiC billets were washed in NaOH solution with an ultrasonic bath for 15 minutes, then washed in pure water. The Ni-based superalloy billets were washed in acetone with the same ultrasonic bath for 15 minutes too. Then the dried and cleaned billets were put in a desiccator.



1. SiC ceramic 2. Cu/Ta/Cu multiple interlayer 3. Ni-based superalloy

Fig.1. Sketch map of diffusion welding of SiC ceramic to Ni-based superalloy with Cu/Ta/Cu multiple interlayer

The SiC ceramic billet, Cu/Ta/Cu multiple interlayer, and the Ni-based superalloy billet were loaded into a graphite die. Then, pressure-aided diffusion welding test was carried out by a thermo-mechanical testing machine, type Gleeble 1500, as schematically shown in Fig.1.

The thickness of the Cu layer contacting with SiC ceramic varied from 0.2mm to 0.8mm in order to investigate the effect of the thickness on the strength of welded samples. The thickness of the Ta layer and the Cu layer contacting with Ni-based superalloy kept constant (0.2mm). The samples were heated up by passing an electrical current. As SiC ceramic is an electrical insulator, the electrical current was transferred by the graphite die. The temperature of the sample was measured by thermocouple as shown in Fig.1 and controlled automatically.

The samples were heated up in vacuum of 1×10^{-3} torr at the rate of $3^\circ\text{C}/\text{second}$ to welding temperature, of which the range tested in this study is $975\sim 995^\circ\text{C}$. After keeping the welding temperature for 5~30 minutes, the samples were cooled down to 500°C at the rate of $3^\circ\text{C}/\text{second}$ and then to 100°C at the rate of $1^\circ\text{C}/\text{second}$. The welding experiment was conducted under an axial pressure, called as welding pressure, of which the range tested is 6.4~32.0MPa.

The strength of the welded samples was determined by Three Points Bending Test. The size of the cylindrical samples is $\Phi 10\text{mm} \times (50.4\sim 51.0)\text{mm}$. The bending strength was calculated using the formula given in Reference [6]. The strength of SiC ceramic was determined in the same way. Then the relative welding strength was calculated by dividing the strength of the welded sample by the strength of the SiC ceramic.

The microstructure and composition of the welded area were analyzed by scanning electron microscope (SEM) and energy dispersive x-ray microanalysis system (EDX).

RESULTS AND DISCUSSION

1. Effect of Welding Temperature on Bending Strength of Welded Samples

The effect of welding temperature on bending strength of welded samples is shown in Fig.2. The experimental conditions are as follows: thickness of the copper layer contacting with SiC is 0.2mm; time to keep the welding temperatures is 10 minutes; welding pressure is 19.1MPa. This figure indicates that the welding strength is lower at lower welding temperature; it increases with increasing the temperature and reaches a maximum at 985°C ; then it decreases with further increasing the temperature.

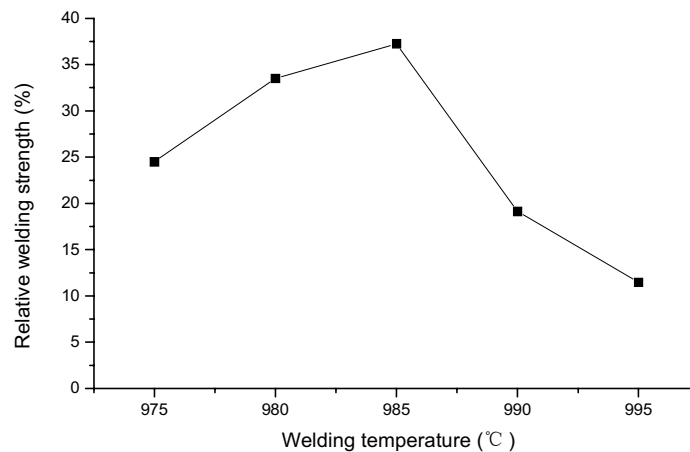


Fig.2 Effect of welding temperature on relative bending strength of welded samples

The process of diffusion welding can be approximately divided into two stages. In the first stage, the surfaces to be welded deform plastically leading to good contact between them. The distance between the surfaces is so small that chemical bonding can form. In the second stage, diffusion of atoms and vacancies as well as migration of dislocations take place, which can eliminate the boundaries between the surfaces. Both the stages are remarkably affected by welding temperature. If the welding temperature is too low, good contact between the surfaces cannot be obtained by plastic deformation, and this will hinder diffusion to take place in the second stage, besides the diffusion coefficient will be low, unfavorable to bonding at the interfaces. However, if the welding temperature is too high, the plastic deformability of copper will be much more improved. In the meantime, some impurity elements will diffuse fast into the copper interlayers, which may decrease the melting point of the interlayers, leading to the fact that the copper interlayers partially melt at the temperature lower than the melting point of copper, and flow out of the interface area. This will result in thinning of the copper interlayers and consequently reduce the ability of the interlayers to relax the welding stress. Hence, an optimum welding temperature exists, as can be seen in Fig.2.

2. Effect of Thickness of the Copper Interlayer Contacting with SiC Ceramic on Welding Strength

The experiment to study the effect of the thickness of the copper interlayer contacting with SiC ceramic on welding strength was carried out under the conditions that welding temperature was 985°C; time to keep the temperature was 10 minutes; and welding pressure was 19.1MPa. The results are shown in Fig.3. One can see that below 0.6mm welding strength increases with increasing thickness of the copper interlayer, it reaches a maximum at 0.6mm, and then the strength keeps stable approximately.

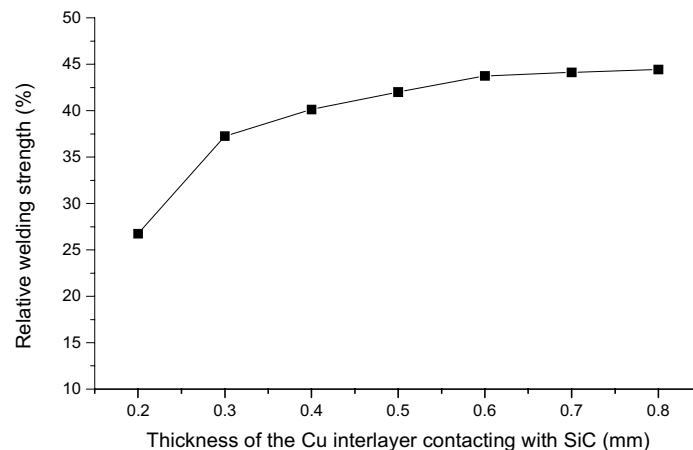


Fig. 3 Relative bending strength of welded samples as a function of thickness of the copper interlayer contacting with SiC ceramic.

The effect of thickness of the copper interlayer on welding strength can be attributed to the influence of the thickness on the capability of the interlayer to relax welding thermal stress. This capability is related to the plasticity, the CTE and other properties of the interlayer. Since the plasticity of Cu is good, the welding thermal stress can be relaxed by plastic

deformation of the interlayer. From this point of view, the thicker the Cu interlayer, the higher the capability to relax thermal stress. Considering that the CTE of Cu ($16.5 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ [7]) is much larger than that of SiC ($4.7 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ [2]), hence thicker Cu interlayer generates larger thermal mismatch between SiC ceramic and the Cu interlayer. The experimental results shown in Fig.3 can be explained by the joint influence of these two aspects.

3. Effect of Time to Keep Welding Temperature on Welding Strength

The effect of time to keep welding temperature on welding strength is presented in Fig.4. The experimental conditions are given below: thickness of the copper interlayer contacting with SiC ceramic was 0.6mm; welding temperature was $985 \text{ }^\circ\text{C}$; welding pressure was 19.1MPa. This figure shows that when the time is shorter than 15 minutes, the welding strength increases with prolonging the time and reaches a maximum 45.4% at 15 minutes; then it decreases with further prolonging the time.

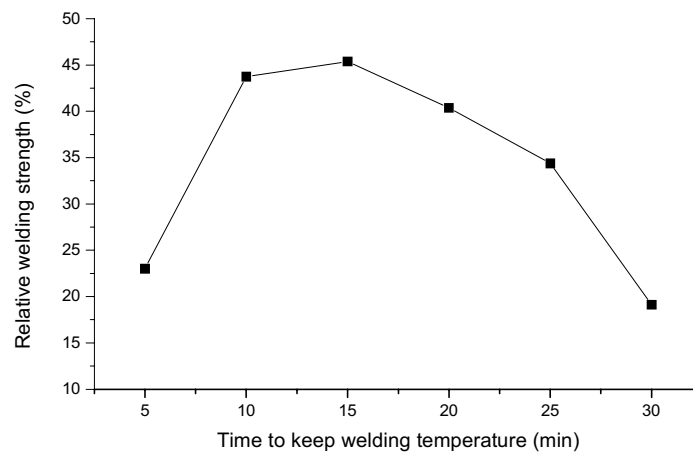


Fig. 4 Relative bending strength of welded samples as a function of time to keep the welding temperature ($985 \text{ }^\circ\text{C}$)

The effect of time to keep welding temperature on welding strength can be attributed to the influence of the time on interdiffusion at the interfaces. The experimental results shown in Fig. 4 indicate that when the time is shorter than 15 minutes, interdiffusions take place continuously at the interfaces with prolonging the time, resulting in increasing of the bonding strength at the interfaces. However, as indicated in Reference [8], with further prolonging the time, a great deal of impurity elements diffuse into the interlayers, leading to decreasing of the melting point of the interlayers. Thus, the interlayers melt at the temperature below the melting point of Cu and flow out of the interface area, resulting in thinning of the interlayers and consequently reducing of the capability of the interlayers to relax thermal stress.

4. Effect of Welding Pressure on Welding Strength

The experiment to investigate the effect of welding pressure on welding strength was conducted under the conditions that thickness of the Cu interlayer contacting with SiC was 0.6mm; welding temperature was $985 \text{ }^\circ\text{C}$ and time to keep the temperature was 15 minutes. The results are presented in Fig.5. The figure shows that the welding strength is lower at

lower welding pressure; it increases with increasing the pressure and reaches a maximum 45.4% at 19.1MPa; then it decreases with further increasing the pressure.

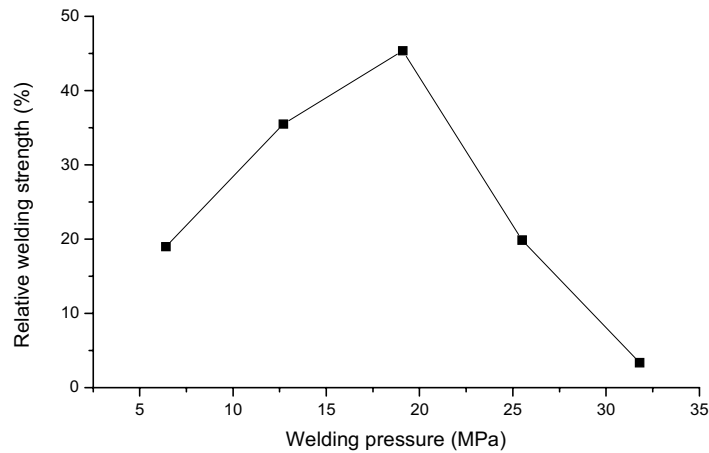


Fig. 5 Relative bending strength of welded samples as a function of welding pressure

Welding pressure directly affects the plastic deformation of the materials to be welded, the contact of the surfaces for welding, the consequently the interdiffusion at the interfaces. Moderate welding pressure not only contributes to a good contact of the surfaces and benefits the interdiffusion but also facilitates Cu to penetrate into the open pores of SiC ceramic, leading to mechanical bonding between SiC and the Cu interlayer after cooling down. However, when the welding pressure is too high, copper plastically deforms excessively and flows out of the interface area, resulting in thinning of the Cu interlayers. Consequently, the capability of the interlayers to relax thermal stress reduces. Besides, excessively high welding pressure may induce microcracks, causing decreasing of welding strength.

5. Microstructure at the welded interfaces

Fig.6 is the scanning electron microphotograph of the welded sample, of which the constitution is SiC/0.3mmCu/0.2mmTa/0.2mmCu/Ni-based superalloy. This sample was prepared under the conditions that welding temperature was 985 °C; time to keep the temperature was 15 minutes; and welding pressure was 19.1MPa. It is seen that there are four interfaces in the photograph, which are SiC/Cu interface, Cu/Ta interface, Ta/Cu interface and Cu/Ni-based superalloy interface. The figure shows that the contact at the interface is good. Also, at SiC/Cu interface, Cu penetrates into the open pores of SiC ceramic, leading to mechanical bonding. Obviously, the intensity of the mechanical bonding is affected by various technological parameters. Moderate welding temperature, proper time to keep the temperature and appropriate welding pressure are beneficial to the penetration of Cu into the open pores of SiC by plastic flow and contribute to the mechanical bonding at SiC/Cu interface.

As mentioned previously, Cu interlayer with thickness of 0.3 mm was made by piling up the Cu sheets with thickness of 0.2 mm and 0.1 mm. Fig. 6 indicates that Cu/Cu interface was eliminated during diffusion welding.

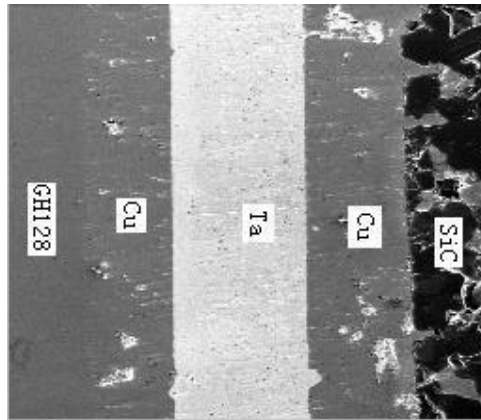


Fig.6 Microstructure at the welded interfaces (985°C, 15 minutes, 19.1MPa, 90×)

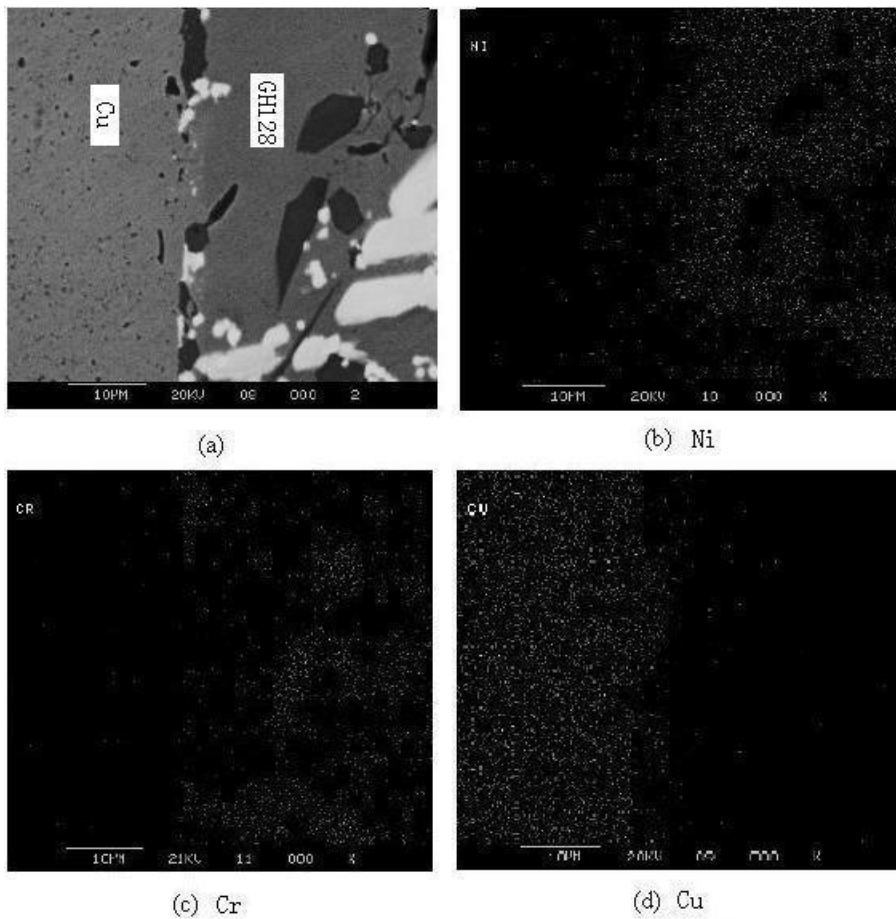


Fig. 7 Microstructure and distribution of the elements at Cu/Ni-based superalloy interface (985°C, 15 minutes, 19.1MPa)

- (a) Microstructure
- (b) Dot map of Ni
- (c) Dot map of Cr
- (d) Dot map of Cu

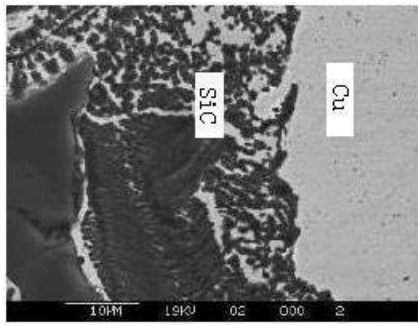
Fig.7 shows the microstructure and the distribution of Ni, Cr and Cu at Cu/Ni-based superalloy interface of the welded sample shown in Fig. 6 analyzed by EDX. The right side of the photographs is GH128 Ni-based superalloy, and the left side is Cu interlayer. This figure clearly indicates that in diffusion welding process both Ni and Cr diffuse from GH128 Ni-based superalloy into Cu interlayer, the diffusion of Ni is more remarkable than that of Cr; in the meantime, Cu diffuses from the interlayer into Ni-based superalloy. Due to these interdiffusions, a diffusion layer with the thickness of about 20 μ m forms at the interface area of which the composition changes continuously. Thus, the joining of Ni-based superalloy to Cu interlayer is realized by the interdiffusions.

Fig.8 shows the microstructure and the distribution of Si and Cu at SiC/Cu interface of the same sample analyzed in the same way. The left side of the photographs is SiC ceramic, and the right side is Cu interlayer. This figure clearly indicates that in diffusion welding process Si diffuses from SiC ceramic into Cu interlayer, in the meanwhile, Cu diffuses from the interlayer into SiC ceramic. Also, a diffusion layer, of which the composition changes continuously, forms at the interface area due to the interdiffusions. It is believed that the interdiffusions contribute to the joining of SiC ceramic to Cu interlayer.

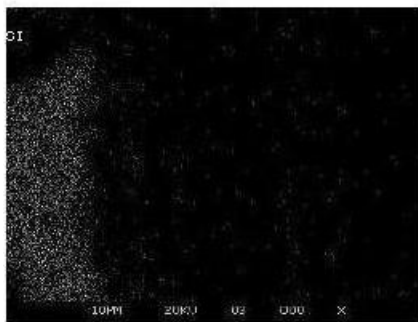
Fig.9 shows the microstructure and the distribution of Cu and Ta at Cu/Ta interface of the same sample analyzed in the same way. The left side of the photographs is Ta, and the right side is Cu. This figure clearly indicates that interdiffusions take place at the interface, forming a diffusion layer, of which the composition changes gradually. Taking into account that fracture never occurs at Cu/Ta interface in bending test to determine the bending strength of the welded samples, one can say that the interdiffusions lead to formation of strong bonding at the interface, and this interface has never been the weakest area of the welded samples.

CONCLUSION

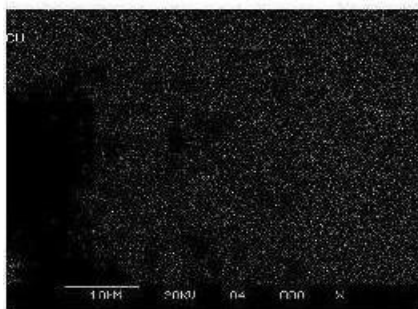
1. Joining of SiC ceramic to GH128 Ni-based superalloy has been successfully realized by diffusion welding with Cu/Ta/Cu multiple interlayer. The obtained maximum welding strength is 45.4% of the strength of the ceramic to be welded. Cu interlayer can effectively relax the thermal stress generated due to the thermal mismatch of the welded materials by plastic deformation. The presence of Ta interlayer can improve the distribution of residual thermal stress.
2. The effect of technological parameters, including welding temperature, time to keep the welding temperature, welding pressure and thickness of Cu interlayer contacting with SiC ceramic, on welding strength has been experimentally studied. The optimum ranges of the parameters have been achieved.
3. The joining of GH128/Cu interface mainly relies on interdiffusions at the interface, while the joining of SiC/Cu interface mainly relies on both interdiffusions at the interface and mechanical bonding formed due to penetrating of Cu into open pores of SiC ceramic. The joining of Cu/Ta interface mainly relies on interdiffusions at the interface. This interface has never been the weakest area of the welded samples.



(a)



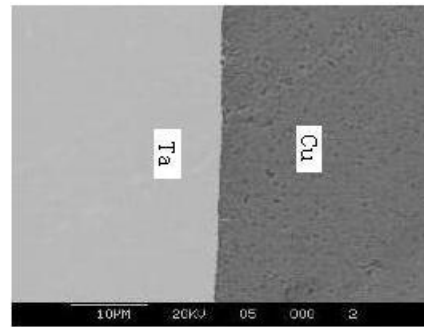
(b) Si



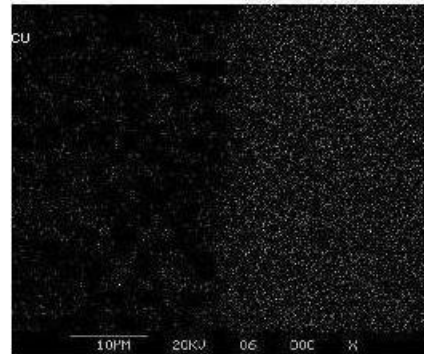
(c) Cu

Fig. 8 Microstructure and distribution of the elements at SiC/Cu interface (same sample as that in Fig.7)

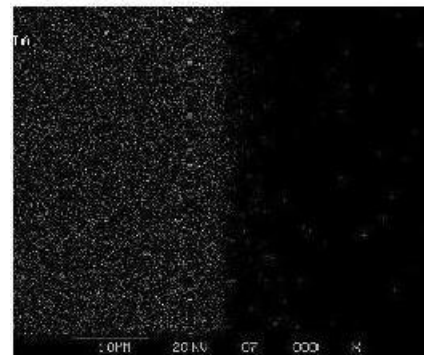
- (a) Microstructure
- (b) Dot map of Si
- (c) Dot map of Cu



(a)



(b) Cu



(c) Ta

Fig. 9 Microstructure and distribution of the elements at Cu/Ta interface (same sample as that in Fig.7)

- (a) Microstructure
- (b) Dot map of Cu
- (c) Dot map of Ta

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