

# Joining of SiC ceramic to Ni-based superalloy with functionally gradient material fillers and a tungsten intermediate layer

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Joining of ceramics to metals is of great interest from both technical and economical points of view. SHS welding of SiC ceramic to Ni-based superalloy has been achieved with TiC-Ni functionally gradient materials as filler and a tungsten sheet as an intermediate layer. The obtained welded strength of the welded sample is 60% of the strength of the SiC ceramic measured under the identical conditions. This value is 25% greater than that without the tungsten intermediate layer. Within the range tested in this study, the strength of welded samples increases with increasing thickness of the tungsten layer. The presence of the tungsten intermediate layer can effectively improve the distribution of process-induced thermal stresses in the joints and reduce the concentration of residual thermal stresses in the ceramic near the welding seam. Microstructural study reveals that diffusion takes place at the interfaces, which promotes bonding at the interfaces.

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## 1. Introduction

Ni-based superalloys are widely used to manufacture parts of aeronautical turbines, which must serve at elevated temperatures. The working temperature of these materials can reach 950°C or higher. But, next-generation turbines will operate at even higher temperatures. In order to further increase the working temperature and decrease the weight of next-generation turbines, new high temperature structural materials are needed and are being investigated. Among them, SiC ceramic and SiC-matrix composite are promising examples. However, one problem arising with the application of SiC is joining of the ceramic to metals, particularly Ni-based superalloys. As indicated by [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 the application of metals.

Self-Propagating High-Temperature Synthesis (SHS) of powder compacts is a novel processing technique being developed as a route for the production of engineering ceramics and other advanced materials [2].

This process seems to offer just the sort of innovative new method for welding of ceramics and intermetallics. SHS offers many attractive features such as energy efficiency, limitation of thermal disruption of heat-sensitive substrate microstructures, easy availability of chemical compatibility between the reaction products and substrates, the possibility to form a composite filler incorporating reinforcing phases such as particles, chopped fibers or whiskers, and the possibility to form functionally gradient material (FGM) joints that show promise for overcoming mismatches between the chemical composition, physical and mechanical properties of dissimilar materials [3–5]. Although SHS welding of ceramics to metals is currently almost exclusively the subject of laboratory investigation, it has shown broad potential and promise from both technical and economical points of view.

With regard to joining of SiC ceramic to Ni-based superalloy, the major problem is concentration of thermal stresses generated during welding and cooling due to the mismatch between the thermal expansion coefficients of the welded materials. The residual thermal

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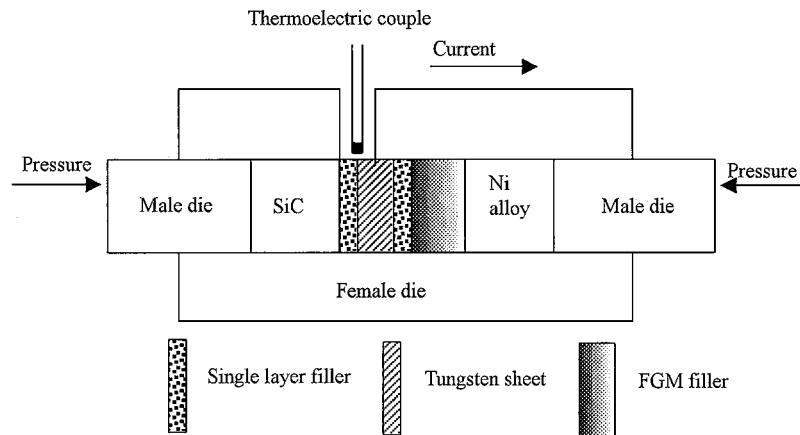


Figure 1 Schematic map of SHS welding of SiC ceramic to Ni-based superalloy with FGM fillers and a tungsten intermediate layer.

stresses may cause generation of microcracks leading to low welded strength and even failure of welding. In order to solve this problem, SHS welding using FGM fillers and a tungsten intermediate layer was experimentally studied. Since the coefficient of linear thermal expansion (CTE) of tungsten ( $4.43 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ , at  $20^\circ\text{C}$  [6]) is very close to that of SiC ceramic ( $4.7 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ , at  $0\text{--}1000^\circ\text{C}$  [7]), it was expected that the presence of this plastically deformable intermediate layer could improve the distribution of thermally-induced residual stresses and reduce stress concentration in the ceramic near the interface of the SiC ceramic/reaction products of the filler (called as the ceramic near the welding seam). The process of welding and the microstructure of the resulting joints were investigated.

## 2. Experimental procedures

Commercial recrystallized SiC ceramic with a density of  $2.65 \text{ g/cm}^3$ , a porosity level of 15–16% and a purity of >99 wt% was used as the starting ceramic to be welded. Forged Ni-based superalloy (type GH128) was used as the starting metallic material to be welded. This alloy contains Cr(19.0–22.0 wt%), W(7.5–9.0 wt%), Mo(7.5–9.0 wt%), Ti(0.4–0.8 wt%), Al(0.4–0.8 wt%), and other elements with very low contents. The CTE of the alloy within the temperature range of  $18\text{--}1000^\circ\text{C}$  is  $16.29 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$ . Its working temperature can reach  $950^\circ\text{C}$  [8]. The properties of this alloy are similar to but better than the properties of the Russian type ЭИ868.

The utilized welding filler materials and their purities as well as the particle sizes were as follows: Ti powder (99.23 wt%, –200 mesh), Ni powder (99.7 wt%, –200 mesh) and W powder (99.5 wt%, 0.5–10  $\mu\text{m}$ ). Amorphous carbon powder was also used as a component of the fillers. Its particle size was 0.1–1  $\mu\text{m}$ .

The SiC ceramic and the Ni-based superalloy were machined to cylindrical billets with the size  $\phi$  10 mm (diameter)  $\times$  5 mm (length). The surfaces for welding were polished. The SiC billets were washed in NaOH solution with an ultrasonic bath for 15 min, then, washed in deionized water. The Ni-based superalloy billets were washed in acetone with the same ultrasonic bath also for 15 min. Following cleaning, the billets were put in a desiccator.

The diameter of the tungsten sheet used as an intermediate material was 10 mm. Its thickness was 0.6 mm and 1.0 mm, respectively, in two different trials. The tungsten sheet was washed in the same way as the Ni-based superalloy.

The powders with designed composition, as mentioned below, were mixed homogeneously and then cold compacted to form compacts with the size  $\phi$  10 mm (diameter)  $\times$  (0.5–2) mm (thickness). The SiC ceramic billet, the powder compact of the single layer filler, the tungsten sheet, the powder compact of FGM filler, and the Ni-based superalloy billet were loaded into a graphite die in the proper order. Then, a pressure-aided SHS welding test was carried out by a thermo-mechanical testing machine, type Gleeble 1500, as shown schematically in Fig. 1. This is a completely computer-controlled apparatus. All of the parameters of the process including heating and pressurization can be controlled automatically. The samples were heated 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 a thermocouple, as shown in Fig. 1, and temperature was controlled automatically.

The size of the powder compact of the single layer filler was  $\phi$  10 mm (diameter)  $\times$  0.5 mm (thickness). This layer was composed of W, Ti, Ni and amorphous carbon powders. The composition of this layer was designed based on the consideration that sound bonding at the interface of the SiC ceramic/the single layer filler and at the interface of the tungsten sheet/the single layer filler can be achieved by SHS welding, and also that the CTE of the reaction products of the single layer filler is close to the CTE of SiC and to the CTE of tungsten in order to reduce the concentration of thermally-induced residual stresses in the ceramic near the welding seam.

The FGM filler was composed of multilayers, in which the composition of the layer contacting the tungsten sheet was the same as the composition of the single layer filler, and the rest layers consisted of Ti, Ni, and amorphous carbon powders, of which the CTE of the reaction products changed step by step between the reaction products of the single layer filler and the Ni-based superalloy. The difference of CTE between the neighbouring layer was less than 25%. The details about the FGM fillers will be reported in [9].

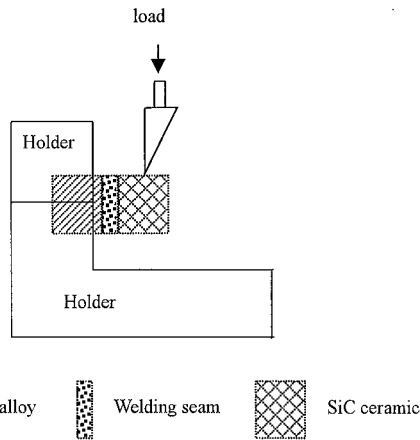


Figure 2 Schematic map of the instrument used to determine welded strength.

The samples were heated to 1190°C in a vacuum of  $1 \times 10^{-3}$  Torr. After holding the temperature constant at 1190°C for 10 min, the samples were cooled down. Both the heating rate and the cooling rate were 3°C/sec. The pressure for welding was 25.5 MPa, which was constant throughout the process.

The size of the welded samples obtained in this study was  $\phi$  10 mm (diameter)  $\times$  (11–12) mm (length). This size is not standard for bending strength measurement. Hence, a special instrument, as shown in Fig. 2, was made to determine the welded strength. The metallic part of the welded sample was inserted into the holder of the instrument for 3 mm, and then it was fixed in place. The load on the ceramic part was applied at a position 3 mm away from the free-end of the ceramic part of the sample. In this way, the entire welded area was included in the range to be tested. As can be seen in Fig. 2, the strength measured in this way is neither a real bending strength, nor a real shearing strength. It combines both of them, and is called as “welded strength” in this paper. In order to acquire data allowing comparison, every measurement has to be conducted under identical conditions. The strength of SiC ceramic was determined in the same way.

Eight specimens were manufactured and examined under identical conditions for each thickness of tungsten intermediate layer (i.e., 0.6 mm and 1.0 mm). The same experiments were carried out for eight specimens without the tungsten layer. The average of welded strength was calculated in each case.

The microstructure and composition of the welded area was analyzed by scanning electron microscope (SEM) and energy dispersive X-ray microanalysis system (EDX) respectively. The phases of the reaction products of the fillers were examined by X-ray diffraction phase analysis (XRD).

### 3. Results and discussion

#### 3.1. Welded strength and fracture position

The fracture loads and fracture positions of the SHS welded samples with tungsten intermediate layer with various thicknesses are shown in Table I. The fracture load of the SiC ceramic sample with the same size measured in the same way is 194 kg. The relative welded

TABLE I Experimental results of SHS welding

No. of sample	Thickness of tungsten sheet (mm)	Fracture load (kg)	Relative welded strength (%)	Fracture position
1	0	93	48	SiC near welding seam
2	0.6	109	56	SiC near welding seam
3	1.0	117	60	SiC near welding seam

strengths of the welded samples (the fracture load of the welded sample divided by the fracture load of the SiC ceramic sample with the same size) are also listed in this table. The experimental results indicate that the relative welded strength increases remarkably using a tungsten intermediate layer, moreover the welded strength increases with increasing thickness of tungsten intermediate layer within the range tested. The welded strength of Sample No. 3, for which the thickness of tungsten intermediate layer was 1.0 mm, is 25% more than that of the welded sample without the intermediate layer (Sample No.1).

Table I shows that the fracture positions of all the samples are located in the ceramic near the welding seam (i.e., in the SiC ceramic about 0.5–1.0 mm away from the interface of the SiC ceramic/reaction products of the filler.) These results indicate that for all the samples, the ceramic near the welding seam is the weakest area due to the concentration of residual thermal stresses.

Based on all the results listed in Table I, it can be seen that the presence of a tungsten intermediate layer remarkably reduces the concentration of residual thermal stresses in the ceramic near the welding seam, even though it has not been eliminated thoroughly.

As mentioned in Section 1, the CTE of tungsten is very close to that of SiC ceramic, and the elastic modulus of tungsten is 405 GPa at 20°C and 325 GPa at 1000°C [6]. This value is also very close to that of SiC ceramic, which is reported to be 412–441 GPa [10]. Taking into account that the CTE of the reaction products of the single layer filler, which was placed between the SiC ceramic and the tungsten intermediate layer, is also close to that of SiC, it is understandable that the thermal stresses-concentrated area of the joints shifts from the ceramic near the welding seam to the tungsten intermediate layer by the presence of the tungsten intermediate layer. This point has been proved by finite element analysis [11]. Since the ability of tungsten to bear stresses, especially tensile stress, is much better than that of SiC ceramic, the welded strength of the joints increases.

As reported in [12], the process-induced thermal stresses of the joints due to the mismatch of CTE of the welded materials can be estimated by the following formula:

$$\sigma = \frac{\Delta\alpha \cdot E_1 E_2}{E_1 + E_2} \cdot \Delta T$$

where  $\sigma$ : thermal stresses of the joints,  $\Delta\alpha$ : difference of the CTE of the welded materials,  $\Delta T$ : difference of temperature, and  $E_1$ ,  $E_2$ : elastic modulus of the welded material.

The formula indicates that the smaller the  $\Delta\alpha$ , the lower the  $\sigma$  will be. Since the CTE of tungsten is very close to that of SiC ceramic, the thermal stresses of the SiC/W joint is low. This can be the other reason for the fact that the welded strength of the samples increases by inserting a tungsten intermediate layer. Besides the consideration of elastic aspect, tungsten can plastically deform at elevated temperatures. The residual thermal stresses of the joints can be relaxed by the plastic deformation of the tungsten intermediate layer. Therefore, it can be concluded that using tungsten intermediate layer can effectively improve the distribution of thermal stresses and reduce the concentration of residual thermal stresses in the ceramic near the welding seam, leading to an increase of the welded strength. Other methods are required to further relax residual thermal stresses in the ceramic near the welding seam. This investigation is well underway.

### 3.2. Microstructure and diffusion at interfaces

Fig. 3 shows the microstructure at the interface of the SiC ceramic/the single layer filler of Sample No. 2. The right side is SiC ceramic. The photograph indicates that a reaction layer exists near the interface. The compositions of three points A, B, C, as shown in the figure, were analyzed by EDX (It should be mentioned that the utilized EDX instrument cannot analyze carbon). The results are listed below:

- A: Si (40.527 at.%), Ti (0.04 at.%), Ni (59.080 at.%)
- B: Si (3.444 at.%), Ti (80.431 at.%), Ni (2.937 at.%)
- C: Si (40.628 at.%), Ti (30.472 at.%), Ni (26.997 at.%)

These results show that a great quantity of Ti and Ni diffuse from the filler into SiC ceramic during welding.

As mentioned previously, the single layer filler contacting with the SiC ceramic was composed of Ti, Ni, W and C powders, among which Ti and Ni, especially Ti, were active elements. Obviously, the gradients of the chemical potentials of Ti and Ni existed at the interface of the SiC ceramic/the single layer filler, which led to the diffusion of Ti and Ni from the filler to the SiC ceramic. These experimental results are in accordance with that reported in [13].

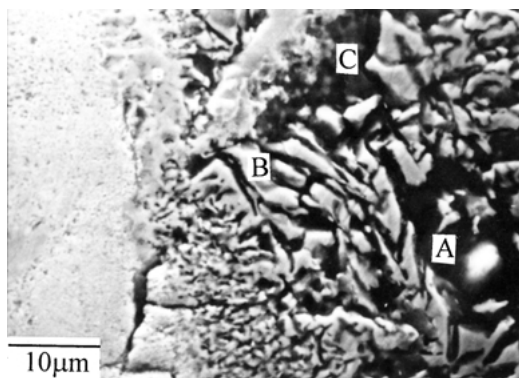


Figure 3 Microstructure at the interface of the SiC ceramic/the single layer filler of Sample No. 2.

It was reported in [13] that the liquid phase formed in SHS welding of SiC ceramic to itself with the Ni + Ti + C (carbon was less than 9 at.%) series fillers can wet the SiC ceramic, and reacts with the SiC ceramic at the interface. This interfacial reaction is realized mainly by the diffusion of Ti from the filler to the ceramic [13]. The wettability and interfacial reaction in SiC/Ni plus Ti system were reported in [14]. The reaction products of SiC/pure Ni system include Ni<sub>2</sub>Si, Ni<sub>5</sub>Si<sub>2</sub>, Ni<sub>3</sub>Si and graphite. Adding Ti to Ni improves the wettability of the system. Ti accumulates at the wetting interface and facilitates the interfacial reactions, resulting in an increase of the driving force of wetting [14]. Kurokawa and Nagasaki reported the formation of only TiC and Ti<sub>5</sub>Si<sub>3</sub> in reaction couples prepared using pressureless sintered SiC and Ti at temperatures between 1173 and 1373 K [15]. Gottselig *et al.* reported that above 1473 K, the primary product was Ti<sub>3</sub>SiC<sub>2</sub> [16]. Based on these reported experimental results, one can conclude that the mechanism of bonding at the interface of the SiC ceramic/ the single layer filler includes wetting and adhesion as well as compound formation due to chemical reactions.

The microstructure at the interface of the tungsten sheet/the filler of Sample No. 2 is shown in Fig. 4. The left side is tungsten sheet. (It should be noted that the compositions of the fillers directly contacting both faces of the tungsten sheet are the same). The compositions of four points A, B, C, D, as shown in the figure, were analyzed by EDX (Again, carbon cannot be analyzed). The results are as follows:

- A: W (4.868 at.%), Ti (21.070 at.%), Ni (74.062 at.%)
- B: W (68.338 at.%), Ti (3.608 at.%), Ni (28.054 at.%)
- C: W (3.012 at.%), Ti (69.445 at.%), Ni (27.543 at.%)
- D: W (69.06 at.%), Ti (7.383 at.%), Ni (23.557 at.%)

As can be seen from Fig. 4 and the analytical results, point D seems to be located within a diffusion layer. It is thinkable that this layer was formed due to the diffusion of Ti and Ni from the filler to the tungsten sheet. The photograph shows good contacting at the interface. No pores were found in the interfacial area. These evidences imply a sound bonding at the interface.

Based on the results of microstructural studies, it can be concluded that substantial diffusion takes place at both the SiC ceramic/filler interface and the tungsten

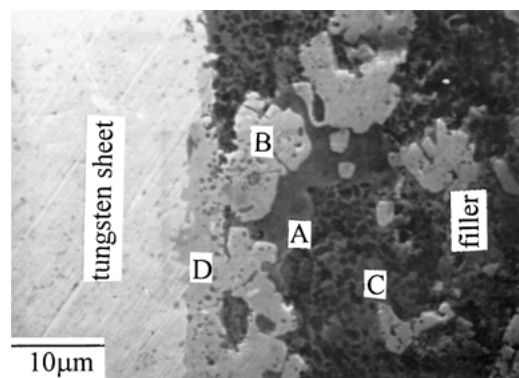


Figure 4 Microstructure at the interface of the tungsten sheet/the filler of Sample No. 2.

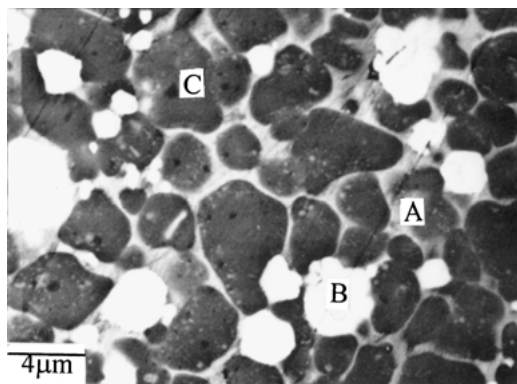


Figure 5 Microstructure of the reaction products of the single layer filler composed of W, Ti, Ni and C.

sheet/filler interface during SHS welding. This diffusion promotes bonding at the interfaces.

The microstructure of the reaction products of the single layer filler composed of W, Ti, Ni, C is shown in Fig. 5. As can be seen, the reaction products include a grey phase (e.g., point A), a white phase (e.g., point B), and a black phase (e.g., point C). The compositions of these three points, A, B, C, were analyzed by EDX. (Again, carbon cannot be analyzed). The results are listed below:

- A: W (12.641 at.%), Ti (10.187 at.%), Ni (77.172 at.%)  
 B: W (73.183 at.%), Ti (5.306 at.%), Ni (21.511 at.%)  
 C: W (4.073 at.%), Ti (88.673 at.%), Ni (7.254 at.%)

The phases of the reaction products of the single layer filler were analyzed by XRD. The X-ray diffraction pattern of the products indicates that the reaction products are composed of WC, W, TiC, Ni<sub>3</sub>Ti, NiC and other phases with very low contents. Taking the results of EDX analysis into account, one can infer that the major component of the black phase in Fig. 5 (e.g., point C) is TiC, the major components of white phase (e.g., point B) are WC and/or W; and the major components of the grey phase (e.g., point A) are Ni<sub>3</sub>Ti and/or NiC.

The reaction characteristics of the fillers composed of Ti, Ni and C powders for SHS welding of SiC ceramic to Ni-based superalloy were reported in [17]. The chemical reaction of the fillers was very vigorous. The main reaction product was TiC, while Ni remained [17]. The experiments for joining of SiC ceramic to Ni-based superalloy with functionally gradient material fillers composed of Ti, Ni and C powders using Gleeble 1500 have been conducted. The temperature of the fillers during SHS welding was recorded automatically. There was a peak on the line of the temperature against the time. The peak started when the temperature increased to 950–1050°C, indicating the occurrence of SHS in the fillers [9, 18]. Hence, it can be concluded that the mechanism for formation of the material comprising the joint is SHS.

#### 4. Conclusions

(1) SHS welding of SiC ceramic to Ni-based superalloy has been achieved utilizing FGM fillers and a

tungsten intermediate layer. Within the range tested in this study, welded strength increases with increasing thickness of the tungsten intermediate layer. When the thickness of the tungsten intermediate layer is 1 mm, the welded strength of the joint reaches 60% of the strength of the SiC ceramic. This value is 25% greater than that without the tungsten intermediate layer.

(2) Using a tungsten intermediate layer can effectively improve the distribution of process-induced thermal stresses in the joints and reduce the concentration of residual thermal stresses in the ceramic near the welding seam. But, the residual thermal stresses in the area were not eliminated thoroughly. This area still is the weakest range of the joints.

(3) Substantial diffusion takes place at the interfaces during SHS welding, which promotes bonding at the interfaces.

(4) The mechanism for formation of the material comprising the joints is SHS or reactive sintering. The reaction products of the single layer filler are mainly composed of WC, W, TiC, Ni<sub>3</sub>Ti and NiC.

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