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Tailoring the interfacial microstructure and mechanical strength of SiC ceramic joints using joining temperature and interlayer thickness

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ABSTRACT

Silicon carbide ceramic joints solid state diffusion bonded by spark plasma sintering with Ti/Ta-5W/Ti multi-interlayer in the temperature range from 1250 °C to 1500 °C were investigated in detail in terms of interfacial microstructure, elemental diffusion, phase constituents, shear strength, and fracture morphology. Layered reaction layers consisting of carbides (TiC, Ti_3SiC_2, (Ta,Ti,W)C), silicide ($(Ta,Ti,W)_5Si_3$) and Ta-rich solid solution (Ta,Ti,W) formed at the SiC/Ti/Ta-5W interfaces. It was found that joining temperature could be decreased from 1600 °C to 1300 °C by inserting the Ti foil between SiC and Ta-5W. Nevertheless, an attempt to further decrease the joining temperature to 1250 °C failed even using a thin Ti foil. The initial thickness interlayer and the number of interlayer had an influence on the interfacial diffusion bonding at SiC/Ti interface. Higher joining temperature was needed to achieve a tough interface joining for a thick interlayer compared with a thin interlayer. The increase of interlayer number also required higher joining temperature. The shear strength reached 123.5 ± 25.3 MPa for the joint diffusion bonded at 1300 °C.

1. Introduction

Silicon carbide (SiC) based materials, one of the most excellent engineering ceramic materials, have been considered as the ideal choice for applications in harsh environments due to their unique combination of their good thermal shock resistance, excellent high-temperature strength, and good chemical stability [1, 2]. Moreover, they also exhibit low cross-section neutron absorption and good irradiation resistance under high temperature neutron irradiation [3]. Hence, SiC based materials are proposed as a potential accident-tolerant fuel cladding candidate in light water reactors and a flow channel insert for advanced fusion reactors [4–6]. However, it is very difficult to manufacture a large component with complicated shapes. Thus, this is a critical issue to develop reliable joining technology to assemble them as large components in complex structures. Most of the works on joining of SiC based materials have been carried on SiC bulk substrate materials [7–9], owing to SiC bulk substrate is less expensive than SiC fiber reinforced SiC matrix composite (SiC_f/SiC). Because SiC_f/SiC composites are usually coated by a chemical vapour deposition SiC layer. Thus, joining technology suitable for SiC may be basically transferred to SiC_f/SiC [9].

Spark plasma sintering (SPS) is an attractive fabrication approach compared with conventional hot-pressing technology, since the electric filed can accelerate ion diffusion [10], thereby contributing to the migration of ions through the joining interface and integrating the materials in a relatively short time [11, 12]. SPS joining of SiC could be obtained by direct joining without joining materials. However, the severe conditions of direct joining, i.e. high temperature (1800 °C–2100 °C) and high pressure (35 MPa–60 MPa) [9, 13], may destroy the structure and damage the properties of SiC_f/SiC composites. Therefore, development of joining technology with relatively lower temperature or pressure is crucial for the application of SiC_f/SiC composites.

It has been demonstrated that the joining technology with appropriate joining materials could effectively decrease the joining temperature or pressure. Although residual stresses, resulting from the differences in coefficient of thermal expansion (CTE) between the interlayer and SiC based ceramics, are hard to eliminate, controlling interfacial microstructure and reaction is a promising strategy to obtain a robust joint that satisfies with the requirements of services environment. Therefore, materials with lower CTE, including Ti_3SiC_2 (TSC, CTE: $9.1 \times 10^{-6} K^{-1}$ [14]) ceramic [7, 11, 12, 15] and refractory

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metals such as Titanium (Ti, CTE: $8.4 \times 10^{-6} K^{-1}$ [16]) [17, 18], Tantalum (Ta, CTE: $6.62 \times 10^{-6} K^{-1}$ [19]) [20], Tungsten (W, CTE: $4.5 \times 10^{-6} K^{-1}$ [19]) [21] and Molybdenum (Mo, CTE: $5.1 \times 10^{-6} K^{-1}$ [19]) [22, 23], have been selected and interlayer to join SiC (CTE: $4.3 \times 10^{-6} K^{-1}$ [2]) based materials.

In our previous work [24], we selected tantalum (Ta) alloyed with 5 wt% tungsten (W) (hereinafter referred to as Ta-5W) foils as interlayer to join SiC ceramics. By controlling interfacial microstructure and reaction, the optimization of joining temperature still reached up to 1600 °C. It was still necessary to lower the joining temperature to prevent the high temperature damage to the materials. It has been reported that SiC could be successfully bonded with thin Ti coating as low as 900 °C [25]. Moreover, Ti can form a ductile solid solution with Ta [26], which was expected to form a good joining between Ti and Ta. Also, Comparing with using a single interlayer, using a multi-interlayer could significantly reduce the strain energy in ceramic joints [16, 23]. Therefore, in the present work, a multi-interlayer of Ti/Ta-5W/Ti was proposed to join SiC ceramics by SPS. The formation of brittle tantalum silicides ($TaSi_2$, Ta_5Si_3) that have high CTE (i.e. $TaSi_2$, CTE: $14 \times 10^{-6} K^{-1}$) at the SiC/Ta-5W interface [24] would be prevented by inserting Ti between SiC and Ta-5W. At the same time, TSC as a promising joining material for bonding of SiC [11, 17] was expected to form at the SiC/Ti interface. Therefore, in order to prevent the formation of silicides at SiC/Ta-5W interlayer and to lower the joining temperature, Ti was inserted between SiC and Ta-5W in this work. The interfacial microstructure, element diffusion behavior, phase constituent and fracture morphology were described in detail. In addition, the mechanical properties of SiC joints were evaluated by shear test.

2. Experimental

The commercial available pressureless sintered α-SiC (> 99.9%, Astek ceramic company, China) was machined in dish shaped samples with diameter of 9.8 mm and the height of 3 mm. The surfaces of these specimens were polished up to 1 μm finish and then these specimens were ultrasonically cleaned in acetone for 10 min dried in air before assembly. The cold-rolled Ta-5W foils (> 99.9%, 100 μm) and Ti foils (> 99.9%, 30 μm or 100 μm) were polished and then cleaned in acetone for 10 min.

The SiC/Ta-5W/SiC, SiC/Ti/SiC, and SiC/Ti/Ta-5W/Ti/SiC sandwiches were assembled in a graphite die for SPS joining (Labox-350 spark plasma sintering system, Japan). The thickness of Ta-5W was 100 μm. Two kinds of Ti thickness, 30 μm and 100 μm were used in this work. The SPS diffusion bonding was conducted with a heating rate of 50 °C/min in Ar atmosphere and pulse-mode direct current (pulse 40 ms, pause 7 ms) was applied for heating. The temperature during SPS process was measured by infrared pyrometer in the hole of graphite die. In order to reduce the residual stress caused by CTE mismatch in the joints, a low cooling rate was set when the holding time was finished. The cooling rate was 10 °C/min above 1000 °C, and 20 °C/min after that. A pressure of 50 MPa was applied on the sample during the heating step. The samples were diffusion bonded in the temperature range from 1100 °C to 1500 °C for 10 min.

After diffusion bonding by SPS, the specimens were cut and for polished cross-sectional microstructure analysis and shear test. The microstructure of the joints was analyzed in a field-emission scanning electron microscopy (FE-SEM, Sigma, Zeiss, Germany) with energy dispersive X-ray spectroscopy (EDS). The elemental intensity profiles of the chemical species across the interface were drawn from the electron probe X-ray microanalyzer (EPMA, JXA-8230, JEOL, Japan). The sample was prepared by focused ion beam (FIB, Helios Nanolab600i, FEI, United States) for transmission electron microscopy analysis (TEM, Tecnai G2 20, FEI, United States). Chemical solutions with a ratio of H_2SO_4: HF: HNO_3: H_2O = 5: 2: 2: 1 was used for sample etching. The etching time for slight corrosion was about 10 s and the etching time for heavy corrosion was about 30 s. The sample after slight corrosion was prepared for electron backscattered diffraction (EBSD) analysis. The EBSD microstructure investigation was obtained by TESCAN MIRI3 SEM equipped by HKL Channel 5 software in the cross-section of the specimens. Sub-size specimens (6 (L) × 2.5 (W) × 3 (T) mm^3) cut from the bonded samples were assembled for shear test with a cross-head speed of 0.5 mm/min and the schematic of shear test as our previous work [24]. To increase the reliability of the data, at least four samples were tested to calculate the shear strength for each bonding temperature.

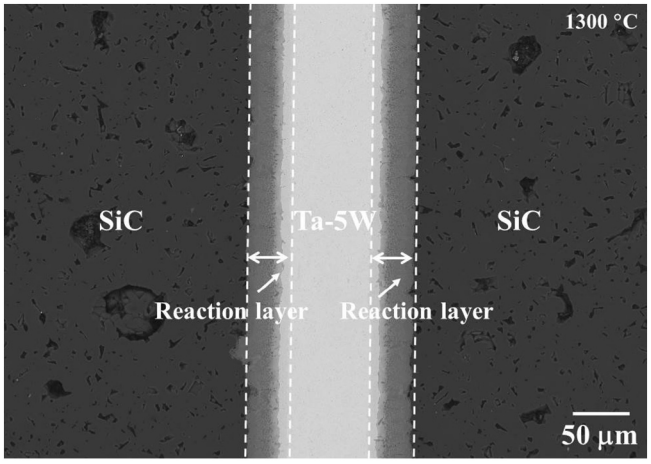

Fig. 1. Cross-section SEM image of SiC/Ti/Ta-5W/Ti/SiC joint diffusion bonded at 1300 °C.

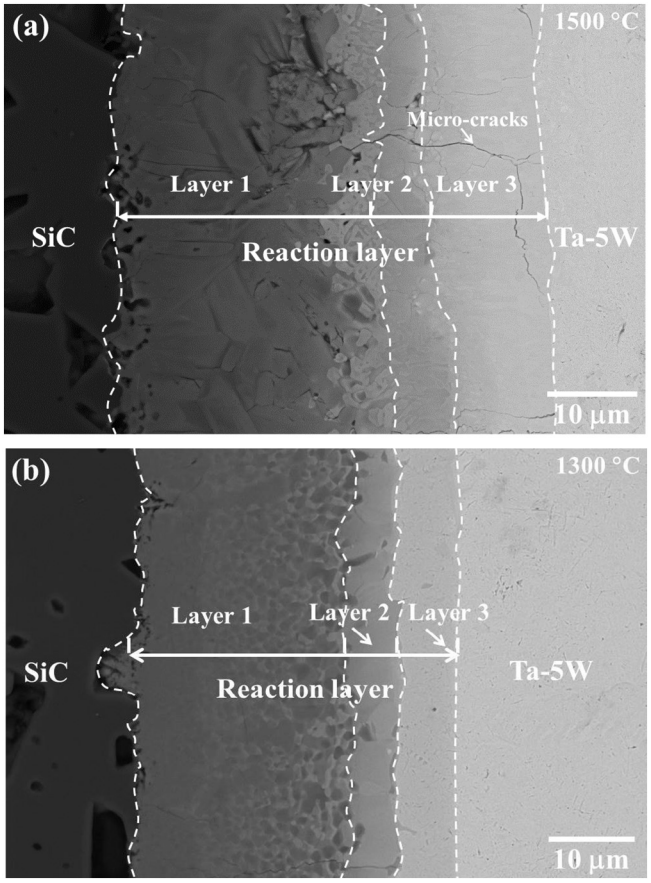

Fig. 2. SEM images of SiC/Ti/Ta-5W interfaces diffusion bonded at (a) 1500 °C and (b) 1300 °C.