



Improvement in mechanical/physical properties of TiC-based ceramics sintered at 1500 °C for inert matrix fuels



Jia-Xiang Xue^{a,b}, Ji-Xuan Liu^{c,d}, Guo-Jun Zhang^{c,d,*}, Hai-Bin Zhang^{a,**}, Tong Liu^b, Xiao-Song Zhou^a, Shu-Ming Peng^{a,**}

^a Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, Mianyang 621900, China

^b Nuclear Fuel Research and Development Center, China Nuclear Power Technology Research Institute, Shenzhen 518026, China

^c State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, Donghua University, Shanghai 201620, China

^d Research Institute of Functional Materials, Donghua University, Shanghai, 201620, China

ARTICLE INFO

Article history:

Received 21 August 2015

Received in revised form 9 November 2015

Accepted 21 November 2015

Available online 12 December 2015

Keywords:

Titanium carbide (TiC)

Titanium silicon carbide (Ti₃SiC₂)

Mechanical properties

Thermal conductivity

Nuclear materials

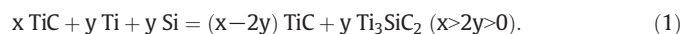
ABSTRACT

Dense TiC-based ceramics were obtained at 1500 °C by hot pressing, benefiting from Ti and Si additives in the TiC matrix. The in-situ formed Ti₃SiC₂ platelet grains refined TiC grains and reinforced the obtained ceramics. The TiC–20 vol.% Ti₃SiC₂ ceramic exhibits a much higher flexural strength of 1003 MPa, compared with 450 MPa of the pure TiC ceramic. Owing to the formation of continuous heat-conducting paths by the bridged Ti₃SiC₂ platelet, a high thermal conductivity over 33 W/m · k was measured in TiC–40 vol.% Ti₃SiC₂ at 500 °C, which is nearly 1.5 times than that in the pure TiC ceramics with a similar grain size.

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The developing of Generation-IV nuclear energy systems (Gen IV) is considered to be effective approaches to improve the nuclear energy efficiency and safety [1]. The inert matrix fuels (IMF) are required to optimize the burn-up of nuclear fuel in the system, which must bear fast neutron irradiation at very high temperature [2]. Titanium carbide (TiC) possesses a high melting temperature and a low neutron absorption cross-section. It also characterizes as a good thermal conductivity and exhibits high hardness and good corrosion resistance [3]. In addition, FCC lattice structure of TiC is isostructural with many actinide monocarbides, which facilitates TiC to form a solid solution with actinide carbides, such as PuC. Therefore, TiC is considered as a promising ceramic matrix to prepare IMF [4,5]. However, the sintering temperature of IMF should not be higher than 1500 °C to prevent vaporization of minor actinide materials [6]. Unfortunately, due to the covalent bonding characteristic of TiC, its densification temperature normally exceeds 1800 °C, even an external pressure is applied [7,8]. Thus, the challenge here is to get TiC with qualified properties sintered lower than 1500 °C.

Considering Ti₃SiC₂ is also a potential material for nuclear reactor applications [9,10], TiC, elemental Ti and Si powders as starting materials were used to prepare TiC–Ti₃SiC₂ ceramics by hot pressing. The stoichiometry reaction among these materials is as follows:



The key step of the technique is to add proper amounts of Ti and Si powders to the TiC matrix before sintering. The Ti–Si eutectic liquid appears around 1330 °C [11], which is benefit to enhance the rearrangement of the TiC grains and the densification process under a much lower temperature. The Ti–Si liquid phase reacted with TiC to form in-situ Ti₃SiC₂, while TiC grain size was reduced. Based on Eq. (1), 20 vol.% Ti₃SiC₂ consumes about 12.3% volume of TiC. During heating, the grain growth of TiC should be inhibited by the secondary isolated Ti₃SiC₂ grains, according to Zener pinning, which should result in a finer TiC grains with a narrow distribution in the final products. A schematic diagram of the proposed sintering process and microstructure evolution is shown in Fig. 1.

The starting materials used in this study were commercial TiC (Titanium Carbide, 2–4 μm, purity 99.9%), Ti (Titanium, 30 μm, purity 99.5%, Shaanxi Fengxiang Titanium Powder Co. Ltd., China) and Si (Silicon, 20 μm, purity 99.0%, Jinan Yinfeng Silicon Products Co. Ltd., China). According to the composition of the final TiC–Ti₃SiC₂ composites with different volume fraction of Ti₃SiC₂, the desired amounts of

* Correspondence to: G.-J. Zhang, State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, Donghua University, Shanghai 201620, China.

** Corresponding authors.

E-mail addresses: anoczhang@sina.com (G.-J. Zhang), hbzhang@caep.cn (H.-B. Zhang), pengshuming@caep.cn (S.-M. Peng).

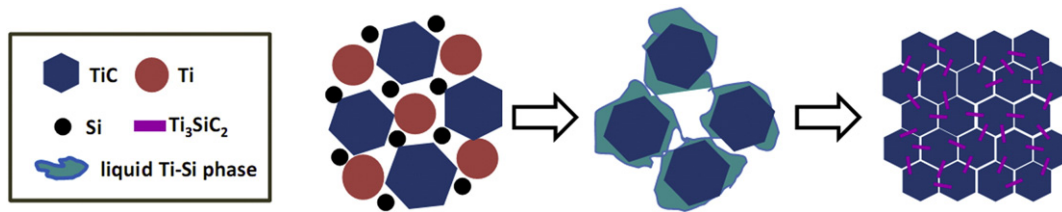


Fig. 1. Schematic diagram of a proposed sintering process and microstructure evolution for TiC-based ceramics introducing Ti_3SiC_2 by Ti, Si additives.

starting powders were mixed and planetary ball-milled in acetone at 560 rpm for 8 h in a polyethylene jar, using Si_3N_4 balls. After the ball-milling, the grain sizes of the starting powder mixture were reduced to around 1 μm . The powder slurry was then dried by rotary evaporation at 60 $^\circ\text{C}$. A sieving through a 200-mesh screen was followed to reduce the possible particle segregation and agglomeration. The as-treated powder mixtures were poured into a graphite die (30 mm \times 37 mm) and then were heated up at a speed of 10 $^\circ\text{C}/\text{min}$, the vacuum level in the furnace was lower than 10 Pa. When temperature reached to 1000 $^\circ\text{C}$, a pressure of 30 MPa was applied on the sample and the furnace was backfilled with argon gas. At 1300 $^\circ\text{C}$, the temperature was hold for 30 min. During the period, the TiC phase should react with Ti and Si to form Ti_3SiC_2 adequately. After that, the samples were heated to the final temperature (1500–2000 $^\circ\text{C}$) at a heating rate of 10 $^\circ\text{C}/\text{min}$ and held at the final temperature for 60 min for densification. Then the samples were cooled down naturally.

The bulk densities of the hot pressing samples were measured by the Archimedes method. The microstructures of the samples were analyzed using scanning electron microscopy (SEM, Hitachi TM-3000, Tokyo, Japan). Three-point bending strength was measured using Type A bars with dimensions of 2 mm \times 2.5 mm \times 30 mm. The span is 20 mm and the cross-head speed is 0.5 mm/min (Instron-5566, America). A total of five bars were fractured to determine the average strength and deviation. Young's modulus was determined by impulse excitation technique using specimens with dimensions 30 mm \times 37 mm \times 5 mm. Hardness and fracture toughness of the samples were determined using Vickers indentation at a load of 10 kg and a dwell time of 10 s. The fracture toughness can also be calculated according to the indentation crack lengths. Thermal conductivity (λ) was calculated as a function of temperature from the measured thermal diffusivity (α), heat capacity (C_p), and the true density (ρ) according to the equation: $\lambda = \rho\alpha C_p$.

According to Eq. (1), final TiC– Ti_3SiC_2 composites with various Ti_3SiC_2 volume fractions of 0, 5, 10, 20 and 40 vol.% were prepared. They were named as TiC, TiC-5TSC, TiC-10TSC, TiC-20TSC, and TiC-40TSC, respectively. Table 1 lists the characteristic of different TiC-based ceramics. Monolithic TiC was hot-pressed at 2000 $^\circ\text{C}$, but it was still not densified fully (relative density ~98.15%). The TiC-5TSC was sintered at 1700 $^\circ\text{C}$, and had a relative density of 98.54%. When the specimens contain more Ti_3SiC_2 , the sintering was performed at a lower temperature of 1500 $^\circ\text{C}$, and the relative densities were 99.21%, 99.58% and 99.66%, corresponding to TiC-10TSC, TiC-20TSC and TiC-40TSC. According to Table 1, lower sintering temperature and higher TiC consumption resulted in smaller average grain size of TiC with increasing of Ti_3SiC_2 content.

Fig. 2 compares SEM microstructure of the TiC and TiC-20TSC samples. A number of closed pores still could be observed in TiC grains after sintering at 2000 $^\circ\text{C}$ for pure TiC (Fig. 2(a) and (b)), and the average grain size is about 20 μm . The large grain size is attributed to the grain coarsening at high densification temperature, in such a case, the moving speed of grain boundary is expected to be higher than that for pores, therefore, closed pores were observed in TiC grains. As regards TiC-20TSC, owing to the low sintering temperature of 1500 $^\circ\text{C}$, the TiC grains can keep relative smaller grain size and nearly no closed pores (Fig. 2(c) and (d)). In TiC-20TSC, the smaller TiC grains are separated by platelet Ti_3SiC_2 grains.

As shown in the SEM images (Fig. 2), the Ti_3SiC_2 grains have platelet morphology and homogeneously distributed among the TiC matrix. The platelet Ti_3SiC_2 will improve the TiC mechanical properties, as shown in other platelet-reinforced structural ceramics. The mechanical properties of the TiC– Ti_3SiC_2 composites are shown in Fig. 3(a). Comparing to the TiC ceramic without Ti_3SiC_2 , the flexural strength of the TiC– Ti_3SiC_2 increases with the amount of Ti_3SiC_2 and reaches to the maximum value of 1003 MPa at 20 vol.% Ti_3SiC_2 , which is two times higher than the TiC ceramic (~450 MPa) and the Ti_3SiC_2 ceramic (~350 MPa). The enhancement of the flexural strength is not only attributed to the finer TiC grain size, but especially also to the strengthening effect between TiC and Ti_3SiC_2 phases caused by thermal residual strength. During the cooling process, due to the mismatch of the thermal expansion coefficients between the matrix TiC grains ($7.4 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$) and Ti_3SiC_2 grains ($8.6 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ and $9.7 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ along the a- and c-axes respectively), a compressive residual stress is supposed to be generated on the matrix of TiC, which improve its fractural strength.

As shown in Fig. 3(a), it should be noted that the platelet Ti_3SiC_2 can play as a strengthen phase at a certain content. A volume fraction of 20% is enough for the composites with high flexural strength, while more Ti_3SiC_2 is detrimental to the strength (TiC-40TSC). With increasing of Ti_3SiC_2 content, the average grain size of TiC from 20 μm for zero Ti_3SiC_2 decreased to 1.91 μm for 20 vol.% Ti_3SiC_2 . However, when the volume fraction of Ti_3SiC_2 was 40 vol.%, the strength dropped down to 420 MPa, which is almost the same with the strength of “pure” TiC with average grain size of 20 μm . It may be caused by a higher volume fraction of Ti_3SiC_2 with large grain size and possible contact between the Ti_3SiC_2 grains. The hypothesis is acceptable because of higher thermal conductivity and electric conductivity of TiC-40TSC (see Fig. 3(b) and (c)). The fracture toughness increased with the amount of Ti_3SiC_2 . The results are obvious and foreseeable because more platelet grains in the matrix will hinder the crack propagation and/or cause crack deflection. This fractural behavior enhanced the fracture toughness remarkably. More Ti_3SiC_2 phase with a low Yong's modulus of 326 GPa lowers Young's modulus

Table 1
The characteristic of different TiC-based ceramics.

Samples	Compositions	Relative density (%)	Open porosity (%)	Mean grain size of TiC (μm)
TiC	TiC	98.15	0.19	19.62
TiC-5TSC	TiC-5 vol.% Ti_3SiC_2	98.54	0.66	3.31
TiC-10TSC	TiC-10 vol.% Ti_3SiC_2	99.21	0.37	2.77
TiC-20TSC	TiC-20 vol.% Ti_3SiC_2	99.58	0.18	1.91
TiC-40TSC	TiC-40 vol.% Ti_3SiC_2	99.66	0.13	1.83

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