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Effects of ZrC content on the synthesis of MAX phase and mechanical properties of C_f-C-SiC-Ti_3SiC_2-ZrC composites

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ABSTRACT

In this research synthesis of Ti_3SiC_2 nano-laminate, effects of Al and ZrC on the amount and morphology of the synthesized MAX phase and mechanical properties of the C_f-C-SiC, C_f-C-SiC-Ti_3SiC_2 and C_f-C-SiC-Ti_3SiC_2-ZrC composites, fabricated by LSI method, were investigated. The infiltration process was conducted at 1500 °C for 30 min and then the samples were annealed at 1350 °C. X-ray diffraction (XRD) technique and scanning electron microscopy (SEM) were utilized in order to investigate the phase composition and microstructure of the samples, respectively. The results showed that the sample containing Al, had the largest amount of synthesized MAX phase and also addition of ZrC led to the decrease of intensities of MAX phase peaks. Among the samples, C_f-C-SiC-$Ti_3Si(Al)C_2$ had the best mechanical properties compared to the others. Bending strength, interlaminar shear strength and fracture toughness of this sample were 505 MPa, 34 MPa and 19.1 MPa $m^{1/2}$ respectively. The results confirmed that the mechanical properties were decreased by addition of ZrC. Among ZrC-containing samples, the sample containing 10 vol% ZrC has shown the least decrease properties including the bending strength of 369.11 MPa, interlaminar shear strength of 26 MPa and fracture toughness of 16.9 MPa $m^{1/2}$. Addition of ZrC phase caused pseudo-plastic behavior appearance in the force-displacement curve and led to fibers pull-out and also displacement enhancement. Microstructural observations confirmed the plate-like morphology of synthesized MAX phases. Furthermore, the distance between layers decreased and MAX phase size increased respectively by addition of Al. Also MAX phase size decreased by increasing the ZrC content. It was confirmed that the MAX phase-containing samples can tolerate various micro-deformation mechanisms including: crack deflection, bending and delamination of lamellae, kink boundary and laminate fracture. These mechanisms led to the toughening of the composites.

1. Introduction

The rapid development of advanced aerospace applications makes an urgent need for high-performance thermal protection system (TPS) materials [1]. Although the C_f-C composites have been introduced as suitable candidates for high temperature applications, but due to rapid ablation failure at high temperature, they cannot meet the requirements [2]. Therefore, it is necessary to improve ablation resistance of the C_f-C composites at high temperatures.

C_f-C-SiC composite is one of the appropriate candidates for high temperature applications. The main reasons behind this choice are low density, high fracture toughness, good thermal shock resistance and acceptable mechanical properties at elevated temperatures [3]. However, due to the activation of silica protection layer above 1700 °C [4,5], it is not able to tolerate the ablation caused by high temperature and high-pressure gas flux.

MAX phases have very interesting properties due to their special structure, for example, they have the properties of ceramics and metals, simultaneously [6]. Laminated structure, low density and excellent resistance against thermal shock are some of their unique properties. Regarding these properties, it can be predicted that addition of these phases to the mentioned composites, leads to the improvement of their properties. Among MAX phases, Ti_3SiC_2 is one of the suitable candidates for carbon-base composites. Ti_3SiC_2 is a ternary carbide with nano-layered structure that includes periodic planar stacking of edge-sharing Ti_6C octahedra and close-packed silicon atoms along the c-axis (Fig. 1).

In addition to easy machinability, this phase has a melting point above 3000 °C which improves ablation resistance of the sample. Weak bond of Ti_3SiC_2 grains causes this phase to show R-curve behavior with fracture toughness equaled with 16 MPa $m^{1/2}$ [7–10]. This phase reveals various deformation modes including delamination, kinking,

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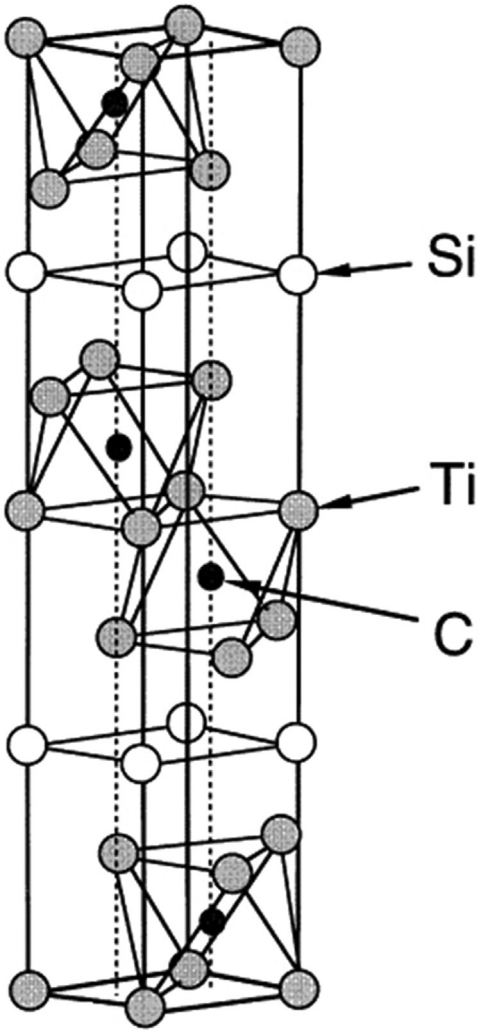

Fig. 1. Ti_3SiC_2 crystal structure.

buckling and bending when exposed to stress. These mechanisms consume some energy, leading to the improvement of the final mechanical properties. The proposed solutions can increase ablation resistance and mechanical properties of the C_f-C composites but reduction in their erosion is negligible due to low refractory carbide content in these composites [11].

On the other hand, reinforcement of C_f-C-SiC composites by ultra-high temperature-resistant ceramics like ZrC, H_fC, T_aC, etc. is one of the well-known solutions which is proposed for improvement of such composite properties [11–15]. Recently, in order to generate a perfect ablation resistant material within a wide range of temperature for prolonged durations, two or more of the mentioned ceramics have been introduced to the C_f-C composites to modify their properties. Among the mentioned ceramics, ZrC and SiC are superior candidates due to good anti-oxidation and anti-ablation properties at high temperature [16–19].

As mentioned, ZrC has attracted significant attention due to its high melting temperature (3540 °C) and also high melting temperature of its oxide ZrO_2 (2770 °C), good chemical inertness, relative low density and good ablation resistance [20,21]. Furthermore, in C_f-C-SiC-ZrC composites, generation of the molten binary oxides ZrC-SiC can seal the cracks and protect the internal matrix and fibers [22,23].

According to the literature survey, effects of Ti_3SiC_2, ZrC and SiC phases on the properties of C_f-C composites have not been investigated yet. In this research, C_f-C-SiC, C_f-C-SiC-Ti_3SiC_2, C_f-C -SiC-Ti_3SiC_2-ZrC composites were fabricated by liquid silicon infiltration (LSI) method and the effects of ZrC content on the amount of the synthesized Ti_3SiC_2, its morphology and eventually the mechanical properties of composites have been investigated.

2. Experimental procedure

2.1. Samples preparation

In order to fabricate C-TiC and C-TiC-ZrC preforms, plain-3k fabric (3k, fiber density: 1.78 g/cm^3, tensile modulus: 230 GPa, tensile strength: 3800 MPa), phenolic resin (Novolak 6109, Hexion, Germany), TiC particles (Luoyang Zhengjie Science & Technology Industry Trade Co. Ltd., china) with the mean particle size of 2–3 μm and ZrC particles (Alfa Aesar, 99.5%) with the mean particle size of 5 μm were utilized. TiC/polymer precursor mass ratio was adjusted to 4.

In order to evaluate the effect of ZrC amount, 0, 5, 10 and 12 vol% ZrC powders were added to the samples. Also Al powders (0.035 wt% of applied TiC) were added as an additive. The raw materials were mixed by planetary ball mill (Retsch PM200, 150 rpm) at 180 rpm for 1 h, under high purity argon atmosphere where the ball:powder ratio was adjusted to 1:10. Hot press was conducted at 180 °C, under a pressure of 1 MPa for 30 min. Then, the samples were post cured at 180 °C for 7 h, then pyrolyzed at 1000 °C for 1 h in order to convert polymer precursor into porous carbon matrix. Silicon infiltration process was carried out at 1500 °C for 30 min under vacuum (10^{-5} Torr). After that, annealing process was conducted at 1350 °C for 1.5 h, under vacuum (10^{-5} Torr) in order to accomplish the MAX phase formation.

2.2. Characterization

In this study, X-ray diffraction analysis (MPD 3000-GNR) was employed for determination of phase composition and scanning electron microscopy (VEGA3-TESCAN) was employed for characterization of microstructure and fracture surface of the samples. Image analysis software was employed in order to measure the distance between layers.

The 3-point bending strength test was carried out according to ASTM C-1341. The dimensions of the obtained samples, loading rate and support span were adjusted to 40 × 5 × 3 mm^3, 0.5 mm/min and 30 mm, respectively. In order to calculate the fracture toughness, single-edge-notched beam (SENB) method was utilized and the interlaminar shear strength test was conducted according to ASTM C1292-00. Loading rates for the fracture toughness and the interlaminar shear strength tests were adjusted to 0.05 mm/min and 0.5 mm/min, respectively and the dimension of the samples for the mentioned tests are illustrated in Fig. 2. The fracture toughness was calculated of Eqs. (1) and (2).

$$K_{1C} = \frac{P}{B}\frac{S}{W^{3/2}} f\left(\frac{C}{W}\right) \tag{1}$$

$$f\left(\frac{C}{W}\right) = 2.9\left(\frac{C}{W}\right)^{1/2} - 4.6\left(\frac{C}{W}\right)^{3/2} + 21.8\left(\frac{C}{W}\right)^{5/2} - 37.6\left(\frac{C}{W}\right)^{7/2} + 38.7\left(\frac{C}{W}\right)^{9/2} \tag{2}$$

Where P is the maximum load, B is the width of the rectangular bar-shaped samples, W is the height of the rectangular bar-shaped sample, S is the support span and C is the notch length.

It worth to note that the reported properties are the average data obtained from 5 samples. The average size of at least 50 Ti_3SiC_2 particles have been measured.