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Suitability of layered Ti_3SiC_2 and $Zr_3[Al(si)]_4C_6$ ceramics as high temperature solar absorbers for solar energy applications

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ABSTRACT

Layered Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ ceramics were prepared by hot-pressed sintering. The spectrally selective absorption properties of these layered carbides have been investigated and compared with those of TiC and ZrC. It is found that the Ti_3SiC_2 ceramic exhibits a solar absorbance of 0.70 and a thermal emissivity of 0.19, yielding spectral selectivity (absorbance divide by emissivity) of 3.7, while the spectral selectivity of $Zr_3[Al(Si)]_4C_6$ is 2.8 (0.71/0.25). To evaluate the suitability of these ceramics as high-temperature absorbers for solar radiation, the investigation on the thermal stability and temperature dependent emittance was performed in air. Compared with TiC and ZrC, both Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ show better thermal stability. Meanwhile, Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ exhibit a low emissivity in the investigated temperature range, benefiting from their metal-like electrical conductivity.

1. Introduction

Harvesting energy directly from the sun is becoming increasingly important in the current energy landscape. There are two well-known technologies available to directly capture the solar energy: solar photovoltaic for direct power generation and solar thermal collection for providing heat. For the application of solar thermal energy, solar hot-water (or solar cooking stove) systems, concentrating solar power (CSP) and solar thermoelectric generators (STEGs) are three distinct ways [1–4]. Both the CSP plants and STEGs rely on the high temperature thermal energy (relative to environmental temperature) to generate electric power, and the energy conversion efficiency increases with the increase of the highest available temperature [3,4]. To convert solar radiation into thermal energy with high efficiency, materials with high adsorption and low emissivity are needed. For spectrally selective absorbers, the characteristics of high absorptance (α_S) in the Ultraviolet-visible-Near Infrared (UV–vis-NIR) region and low thermal emission (ε_T) in the infrared (IR) region have been known as the ideal requirement [5].

So far, a variety of selective absorbing surfaces have been successfully developed [6–10]. In order to achieve better spectral selectivity, most of these selective absorbing surfaces consist of multilayer stacks, such as anti-reflection layer, absorption layer, and IR reflection layer. However, the high temperature application scope of commercial spectrally selective absorber materials is usually restricted because of the oxidation and interfacial diffusion [11,12].

Recently, Sani and Sciti et al. [12–16] investigated the room temperature spectral reflectance and high temperature emissivity characteristics of different carbide and boride ultra-high temperature ceramics. The high temperature emissivity of carbide and boride ceramics were compared with that of silicon carbide (SiC) ceramic, which has been used as volumetric solar absorber [17]. Based on the results, they found that carbide and boride ceramics showed a lower emissivity than that of SiC ceramic at high temperature. However, their high temperature emissivity measurements were carried out in the vacuum environment, rather than in the actual working environment with the presence of air. Actually, poor oxidation resistance is one of the main concerns that reduce the reliability of the transition metal carbide ceramics [18,19].

Previous studies have reported that the oxidation resistance and fracture toughness of the transition metal carbides, such as ZrC and TiC, could be greatly improved by incorporating Al and/or Si into the binary carbides [20–22]. For the quarternary Zr-Al(Si)-C system, the oxidation products are ZrO_2, Al_2O_3 and mullite [20]. Here, Al_2O_3 and mullite act as the oxidation resistance material because the diffusion coefficient of oxygen ions in Al_2O_3 and

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mullite is much lower than that in ZrO_2 at 1273–1573 K [23]. Inspired by this fact, it is reasonable to assume that the high temperature selective absorption properties of Ti-Si-C and Zr-Al (Si)-C ceramics in air environment are better than those of the corresponding binary carbides, TiC and ZrC.

The Ti-Si-C ternary system is developed by adding Si into TiC. These ternary titanium silicon carbides are part of a class of new materials, namely the $M_{n+1}AX_n$ phases (where M is an early transition metal, A is an A-group element, X is C and/or N, and $n=1$, 2 or 3) [24]. Ti_3SiC_2 is a typical member of the MAX phases. Its crystal structure can be described as two layers of Ti_6C octahedra (edge sharing) interleaved with a layer of Si-element [22]. This unique structure provides Ti_3SiC_2 with the characteristics of both metals and ceramics. Like metals, it possesses good electrical and thermal conductivity, excellent thermal shock resistance and machinability. Meanwhile, like ceramics, it has high elastic moduli, high melting points and good thermal stability [25–28]. Up to now, a lot of research works on Ti_3SiC_2 have been conducted, and are mainly focused on the mechanical properties [24,25], thermal properties [29,30], absolute thermopower [31], dielectric permittivity and electromagnetic interference shielding effectiveness [32].

On the other hand, the Zr-Al(Si)-C quaternary system is developed by adding Al(Si) into ZrC. $Zr_3[Al(Si)]_4C_6$ is one member of the Zr-Al(Si)-C system with the general formula of $(TC)_n[Al(Si)]_4C_3$ (where T is Zr or Hf, $n=1, 2, 3\ldots$) [18], and the crystal structure is ZrC in the NaCl-type structure being interleaved by $[Al(Si)]_4C_3$ unit [20]. Similar to MAX phases, it also shows good physical and mechanical properties. Furthermore, $Zr_3[Al(Si)]_4C_6$ exhibits excellent oxidation resistance besides other merits shared by ZrC ceramics [20,33].

To the best of our knowledge, there has been no investigation on the spectrally selective absorption properties of Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ yet. In this work, the layered Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ ceramics were prepared and investigated as high-temperature solar absorbers. The spectral properties and thermal stability of Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ samples were investigated. The high temperature emittances of Ti_3SiC_2 and $Zr_3[Al(Si)]_4C_6$ samples are also measured in air atmosphere to evaluate their potential as high-temperature solar absorbers. For comparison, TiC and ZrC ceramics were also prepared and investigated.

2. Experimental

2.1. Samples preparation

To prepare the bulk Ti_3SiC_2 (TSC) ceramics, commercially available Ti_3SiC_2 powder (4 μm, 98% purity, Yuehuan New Material Technology Co. Ltd, Shanghai, China) was used as the raw material. Firstly, the Ti_3SiC_2 powder was loaded into a graphite mold with an inner diameter of 55 mm, which was pre-sprayed with a layer of boron nitride, and then pre-pressed under a pressure of 5 MPa. Subsequently, the compacted powder was hot-pressed at 1773 K for 0.5 h under the pressure of 30 MPa in Ar atmosphere.

$Zr_3[Al(Si)]_4C_6$ (ZA(S)C) samples were prepared by in situ reaction hot-pressing process, as has been described elsewhere [34].

Table 1
Main features of the as prepared and polished samples.

Samples	Abbreviations	Relative density (%)	Surface roughness (μm)
Ti_3SiC_2	TSC	98	~ 0.08
$Zr_3[Al(Si)]_4C_6$	ZA(S)C	98	~ 0.09
TiC	TC	95	~ 0.06
ZrC	ZC	88	~ 0.04

Bulk TiC (TC) and ZrC (ZC) ceramics were also prepared through hot-pressing method using TiC powder and ZrC powder (both with purity of 99.99%, New Metal Materials Tech. Co., Ltd, Beijing, China) as starting materials. The sintering process is similar to that of Ti_3SiC_2 ceramics, except that the TiC powder and ZrC powder were hot-pressed at 2273 K for 1 h, respectively.

All the as-sintered samples were cut into disks with 27 mm in diameter and 2 mm in thickness. And the disks were ground, polished with the emery paper and diamond pastes. The main features of the samples for subsequent investigation are listed in Table 1.

As to the investigation on the long-term thermal stability of these samples in air atmosphere, the ceramic samples were heated up at 700 K, 800 K, and 900 K for 9 h, respectively. Samples of TSC heated at 700 K, 800 K, and 900 K are denoted as TSC-700 K, TSC-800 K and TSC-900 K, respectively, and so for ZA(S)C.

2.2. Characterization

The relative density was determined by the Archimedes method. The mean surface roughness was analyzed with a Laser Scanning Microscope (LEXT OLS4000, OLYMPUS). X-ray diffraction (XRD) measurements were carried out on an ARL X'TRA system with Cu Kα radiation source ($\lambda=$ 0.15406 nm). The surface and

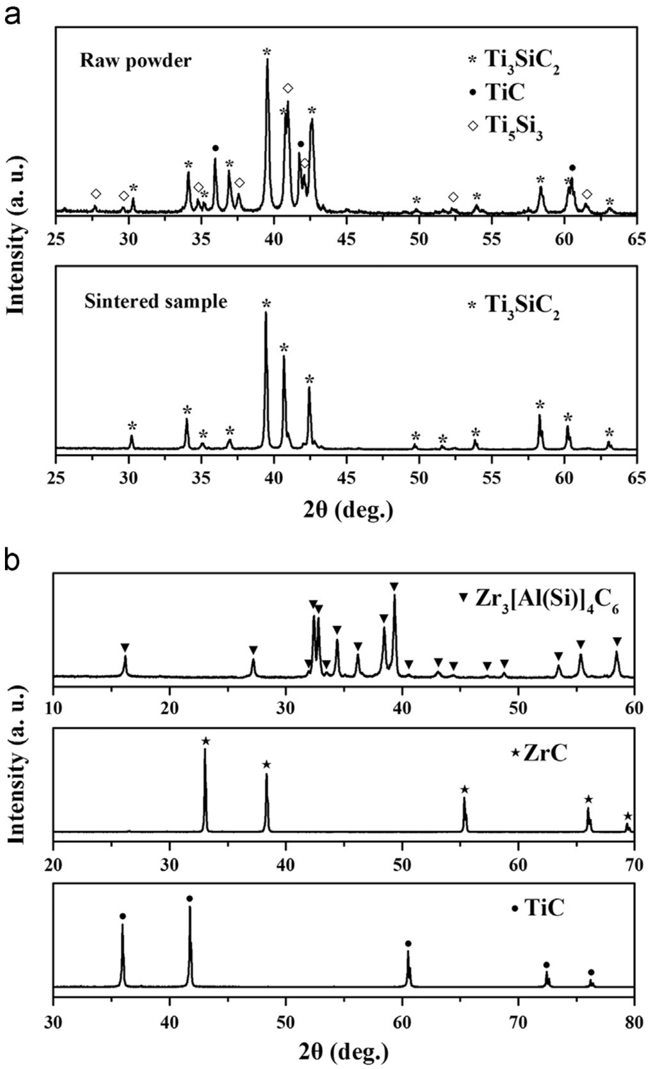

Fig. 1. XRD patterns of (a) the raw powder and sintered bulk sample for Ti_3SiC_2; (b) the as-sintered $Zr_3[Al(Si)]_4C_6$, TiC and ZrC samples.