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Synthesis and characterization of Al_2O_3/SiC composite ceramics via carbothermal reduction of aluminosilicate precursor for solar sensible thermal storage

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ABSTRACT

Al_2O_3 material with high heat capacity and high thermal conductivity was in situ formed to bond with SiC for synthesizing Al_2O_3/SiC composite ceramics by the removal of silica in aluminosilicate precursor using carbothermal reduction method. Phase transformation, microstructural evolution, and the relevant variations in properties were studied in the temperature range 1460–1580 °C. Results indicated that the addition of Y_2O_3 improved the carbothermal reaction rate significantly and decreased the finish temperature of carbothermal reaction to 1500 °C. Incorporation of the in situ formed Al_2O_3 could lower the sintering temperature of SiC ceramic without decreasing the heat capacity. By sintering at the optimal temperature as 1540 °C, Al_2O_3/SiC composite ceramics with the heat capacity of 1.16 J/(g·K) and the high thermal conductivity of 13.73 W/(m·K) were obtained. The relatively high thermal conductivity endowed the composites with a good thermal shock resistance. This study was intended to identify the reaction conditions for obtaining sensible thermal storage materials with favorable morphology and properties by using a simple in situ synthesis method.

1. Introduction

CO_2-induced global warming has become a pressing issue, and needs to be tackled urgently. Efficient utilization of solar energy is thereby proposed and being considered increasingly as a way to achieve a sustainable and clean energy supply for human beings [1,2]. Solar thermal power generation is one of the prevalent power generation technologies using solar energy, and the conversion of solar energy into thermal energy is the easiest and the most extensively accepted method [3]. The main problem of solar thermal power generation is the mismatch between the solar energy and the energy demand, since solar energy is an intermittent energy resource. To remove the fluctuations caused by the intermittent nature of solar energy, a thermal storage system composed of thermal storage materials is required to be attached with solar collectors to store energy and ensure the continuous power supply [4,5]. The thermal storage materials are classified into three main categories according to different storage technologies: sensible heat storage (SHS), latent heat storage, thermo-chemical storage. Among these thermal storage technologies, sensible heat storage is the most simple and inexpensive way for thermal storage system with technological and economical superiority, although there are few advantages of latent storage and thermo-chemical storage over sensible heat storage [6,7].

Excellent properties of the thermal storage materials are the key factors to guarantee the operation of solar sensible thermal storage system. A high thermal storage capacity (i.e. sensible heat capacity) is essential to reduce the volume of sensible thermal storage system and increase the thermal storage efficiency, whilst a good thermal shock resistance allows the thermal storage materials to avoid mechanical degradation after thermal cycles [2]. Numerous oxide ceramics, such as alumina, zirconia, mullite, cordierite, were discovered to be suitable for SHS due to the high thermal storage capacity, the good thermo-chemical stability and the good corrosion resistance against hot heat transfer fluid (HTF, usually air) [8]. Owing to the poor thermal conductivity, however, oxide ceramics were susceptible to thermal shock, and the drastic degradation in mechanical properties after the thermal shock had limited their wide applications at high temperature (≥850 °C), especially the thermal storage application [9,10]. Also the poor thermal conductivity of the oxide ceramics led to the poor heat transfer capability,

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which might slow down thermal storage-retrieval process of the thermal storage system [11,12]. SiC ceramics consequently attracted much attention as candidate materials for solar sensible thermal storage due to the high thermal conductivity as well as the high thermal storage capacity [13,14]. The high sintering temperature (≥2100 °C) of covalent compound SiC [15,16] and the high cost of SiC starting materials [17], however, rendered the difficulties of producing SiC ceramics with low manufacturing cost. Due to the high sintering temperature, the complex sintering processes of SiC ceramics, such as the hot-pressing sintering [18], the spark plasma sintering [19,20], and the vacuum sintering [21], etc, were generally employed for the production of SiC ceramics, which imposed strict requirement on sintering equipment and made the fabrication of products with complex shapes extremely difficult [22], thus hampering the comprehensive utilization of SiC ceramics in solar sensible thermal storage. As known, pressureless liquid phase sintering of SiC by introducing oxide materials as sintering additives did not suffer from these shortcomings, and the introduced oxide materials (e.g. Al_2O_3, silica, and mullite, etc.) not only promoted the sintering of SiC, but also substituted partial SiC to synthesize SiC composite ceramics with lower cost [23–26]. Among the commonly-used oxide materials, Al_2O_3 possesses optimal thermophysical properties. For example, in the temperature range 25–600 °C, the heat capacity of Al_2O_3 is 0.77–1.19 J/(g·K), while those of silica and mullite are 0.44–1.13 J/(g·K), 0.76–1.18 J/(g·K), respectively [27,28]; the thermal conductivity of Al_2O_3 is 17.5 W/(m·K) at room temperature, which exceeds the thermal conductivity levels of silica and mullite (i.e. 1.4 W/(m·K) for silica, 6.1 W/(m·K) for mullite) [29–31].

In our previous works [13,31,32], Al_2O_3–SiC composite ceramics with the high thermal storage capacity, such as Al_2O_3–SiC and Al_2O_3–SiC–ZrO_2 composites, were obtained by using aluminosilicate minerals (e.g. kaolin, andalusite, talc, etc.) as additives to promote the pressureless liquid phase sintering process, whilst the properties were characterized for elevating the suitability for solar sensible thermal storage. The composite ceramics suffered from the poor thermal shock resistance as the bending strength decreased significantly after 30 cycles of thermal shock (instantaneous temperature up to 1000 °C), although the high thermal storage capacity and the low sintering temperature were accomplished. The poor thermal shock resistance of the composites was attributed to the poor thermal conductivity of 1.5–2.3 W/(m·K), which mainly resulted from the introduction of silica from the aluminosilicate additives. Hence there was an urgent need to remove the negative effect of silica on the thermal conductivity when using aluminosilicate minerals of low cost as raw materials for fabricating thermal storage materials. It is noteworthy that, as reported by R. Naghizadeh and F. Golestani-fard [33], silica in the aluminosilicates could be reduced in the form of gaseous silicon monoxide (SiO) by carbothermal reduction, whose principle was described in Eq. (1). This method is thereby considered to be an effective way to cope with the thermal conductivity degradation caused by the introduced silica.

$$3Al_2O_3 \cdot 2SiO_2(s) + 2CO(g) \rightarrow 3Al_2O_3(s) + 2SiO(g) + 2CO_2(g) \quad (1)$$

In this study, the carbothermal reduction method was employed to remove the negative effects of silica on the thermal conductivity as well as the heat capacity of Al_2O_3/SiC composite ceramics, which were fabricated using andalusite, an abundant aluminosilicate mineral with high alumina content, thus avoiding the conventional costly, energy-consuming and complicated sintering processes of SiC ceramics. The main factors affecting the carbothermal reaction rate, the phase transformation and the microstructural evolution of the composite ceramics were investigated, whilst the relevant variations in properties were studied in detail. The Al_2O_3/SiC composite ceramics were characterized by the high thermal storage capacity, the high thermal conductivity as well as the good thermal shock resistance. The present work was aimed at studying the reaction conditions for obtaining Al_2O_3/SiC composite thermal storage ceramics with favorable properties by a less complex and low-cost in situ synthesis method.

2. Material and methods

2.1. Starting materials

Starting materials for experiments were SiC powder with two particle sizes of 106 μm and 58 μm (Dan Jiangkou Hongyuan Silicon Carbide Co., Ltd, Hubei, China), andalusite (under 250-mesh, Xinjiang Baoan New Energy-Mineral Co.,Ltd, Xinjiang, China), Al powder (under 250-mesh, Sinopharm Chemical Reagent Co.,Ltd, Shanghai, China) and Y_2O_3 (chemical pure, Sinopharm Chemical Reagent Co.,Ltd, Shanghai, China). Based on our previous study [31], the weight ratio of SiC to Al_2O_3 in the final products were taken as 65:35, respectively (i.e. 35 wt.% SiC was substituted by Al_2O_3). According to Eq. (1), the proportions of the starting materials for formula AS were calculated to be: 63.5 wt.% SiC, 22.7 wt.% andalusite, 13.8 wt.% Al. 3 wt.% Y_2O_3 was added as the catalyst for the decomposition of mullite as well as the sintering additive for densification, since the finish temperature for carbothermal reaction reported in Ref. [33] was as high as 1650 °C. Al powder was added to react with the excess silica of andalusite to produce the intermediate mullite. The chemical compositions of starting materials and formula AS were listed in Table 1, in which the compositions of formula AS were calculated including Al_2O_3 derived from the transformation of Al. The other two formulae, i.e. AS1 and AS2, were designed to study the effects of Y_2O_3 and the added SiC on the carbothermal reaction rate and the microstructural evolution by performing XRD and SEM analysis of as-sintered samples. Both formulae excluded the addition of SiC, and the ratio of andalusite:Al was consistent with that of formula AS. Formulae AS1 and AS2 were differentiated by 8 wt.% Y_2O_3 addition and no Y_2O_3 addition, respectively.

2.2. Processing

The starting materials were mixed thoroughly with 5 wt.% PVA binder addition by ball milling, and then pressed by a uniaxial pressure of 20 MPa into rectangular samples of 6.8 mm × 6.8 mm × 38 mm and cylindrical samples of Φ10 × 2 mm. The formed powder compacts were dried at 100 °C for 24 h in an electric oven and pressureless sintered by the carbon-buried sintering method (Fig. 1) from 1440 °C to 1580 °C in a seggar full of carbon at molybdenum disiliciade furnace with a heating rate of 5 °C/min and a holding time of 3 h at the maximum temperatures.

2.3. Characterization

The water absorption (Wa), the open porosity (Pa) and the bulk density (D) were measured by with the rectangular samples by AUY120 electronic analytical balance (Japanese Shimazu) through static weighing method. The bending strength was measured with the rectangular samples by Computer Control Electronic Universal Test Machine (Shenzhen Reger Instrument Co., Ltd., Guangdong, China). The heat capacity, the thermal diffusivity as well as the thermal conductivity were measured using the cylindrical samples by TC-7000H laser flash thermal constant analyzer(ULVAC SINKU-RIKO. Inc., Yokohama, Japan). The thermal shock resistance of the