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Corrosion behavior of porous silicon nitride ceramics in different atmospheres

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ABSTRACT

The corrosion behavior of silicon nitride (Si_3N_4) ceramics with a porosity of 46% at 1200–1500 °C under different conditions including dry O_2, O_2 containing 20 vol% H_2O and Ar containing 20 vol% H_2O is compared. The results show that porous Si_3N_4 ceramics exhibit good oxidation resistance up to 1200 °C. Their corrosion behavior varies depending on the temperature and atmosphere. Water vapor can obviously affect the morphology of the reaction product and thus accelerate the corrosion rate due to its specific inward diffusion mechanism and devitrified effect at high temperature. In view of the reaction kinetics, it proceeds in a diffusion-controlled manner in dry O_2 while follows the parabolic-linear law at water-containing atmosphere. Furthermore, a new model considering both oxidation and volatilization reactions is established. These provide a baseline for expanding the application fields of non-oxide porous ceramics such as Si_3N_4 and silicon carbide (SiC) etc.

1. Introduction

Porous ceramics as a kind of promising candidates are widely applied in various industrial fields, for instance gas filters, catalyst supports, separation membranes and high-temperature insulations [1–3]. Porous Si_3N_4 ceramics have acquired great interests recently because they possess such outstanding properties as high hardness and fracture toughness, excellent thermal-chemical corrosion resistance, low thermal expansion and acceptable dielectric property etc. [4–6]. Mesoporous Si_3N_4 ceramics have been suggested as a good candidate for base-catalysed reactions [7,8]. In the area of biomaterials, their outstanding mechanical properties, fracture toughness, hardness and wear resistance have pin-pointed Si_3N_4-bioglass composites as a kind of promising candidates for various high-load medical applications [9]. Many methods including alginate template [10], gel casting method [11,12] and slip casting [13] etc. have been put forward to fabricate porous Si_3N_4 ceramics. However the tendency to be oxidized at high temperature damages their properties. Especially when exposed to combustion environment, Si_3N_4 ceramics are oxidized and corroded simultaneously according to the following equations [14–21]:

$$Si_3N_4(s) + 3O_2(g) = 3SiO_2(s) + 2N_2(g) \quad (1)$$

$$Si_3N_4(s) + 6H_2O(g) = 3SiO_2(s) + 2N_2(g) + 6H_2(g) \quad (2)$$

$$SiO_2(s) + 2H_2O(g) = Si(OH)_4(g) \quad (3)$$

Several studies have been carried out on the corrosion behavior of porous Si_3N_4 ceramics [22–25]. Porz et al. [22] pointed out that the pore channel radius affected the oxidation of reaction-bonded silicon nitride (RBSN) with the porosity of 21% and the typical oxidation was represented by an asymptotic law. Fox [23] investigated on the oxidation of porous RBSN at 900–1000 °C in flowing dry oxygen. The results showed that an amorphous Si-C-O coating prevented the further oxidation and a small linear weight loss was observed. Zhang et al. [24] systematically studied the effect of pre-oxidation on the microstructure, mechanical and dielectric properties of porous Si_3N_4 ceramics with the porosity of 62% at 1150–1400 °C for 2–3 h in dry air. Cai et al. [25] combined freeze drying and oxidation sintering techniques to fabricate the porous Si_3N_4 ceramics with the porosity of 35% or so. Their reaction behavior at 1150–1400 °C for 2–3 h in dry air on the microstructure, mechanical and dielectric properties was investigated. Compared with the work on the reaction behavior of porous Si_3N_4 ceramics in dry air, their reaction behavior at water-containing atmosphere is seldom investigated. Considering water-containing atmosphere is also a kind of common working conditions, it is necessary to clarify the effect of water vapor on the corrosion behavior of porous Si_3N_4 ceramics because water vapor can aggravate the reaction process [17–21]. As for the reaction kinetics under water-containing condition, it has been reported that both the paralinear

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model [26,27] and the dual-reaction model [28] can be adopted. However, both the above models are only suitable to deal with the dense sample. A new model is required to be developed to treat the reaction kinetics of porous Si_3N_4 ceramics under water-containing condition.

In this work, Si_3N_4 ceramics with the porosity of 46% were fabricated in large scale through a new flash combustion method. The corrosion behavior of the porous Si_3N_4 ceramics was investigated at 1200–1500 °C for 10 h at different atmospheres including dry O_2, O_2 containing 20 vol% H_2O and Ar containing 20 vol% H_2O. The aggravated mechanism of water vapor on the reaction is further clarified based on the morphological development of the oxide product. The reaction kinetics under different conditions is discussed. Especially for the reaction at water-containing atmospheres, the model considering both solid product layer and hypothetically volatile product layer is adopted and gets a good agreement with the experimental results.

2. Material and methods

2.1. Preparation of porous Si_3N_4 ceramics

The porous Si_3N_4 ceramics were obtained by a new flash combustion method using industrial FeSi75 powder (Si > 75 mass %) with an average particle size of less than 74 μm as raw material [29]. The reaction was carried out in an industrial flashing furnace with a size of Φ2.5 m×7.5 m. The preparation procedure is as following: Industrial FeSi75 powder was charged into the flash combustion furnace from the top while the nitrogen with the purity of 99.999% was input in the furnace from the bottom. FeSi75 powders were combusted rapidly to produce Fe-Si_3N_4 at 1450 °C, which fell down to the bottom of the furnace due to the gravity [29]. The Fe-Si_3N_4 was further treated by acid to obtain porous Si_3N_4 ceramics.

2.2. Corrosion behavior of porous Si_3N_4 ceramics under different conditions

The as-received porous Si_3N_4 ceramics were cut into cuboids with the size of 5 mm×5 mm ×20 mm. The corrosion behavior of porous Si_3N_4 ceramics under different conditions was investigated using online thermogravimetric (TG) method [30]. The heating unit was a molybdenum wire furnace with a maximum temperature of 1600 °C. As for the water-containing atmosphere, the water vapor was generated by the peristaltic pump. It was mixed with the carrier gas (O_2 or Ar) with the required ratio in volume in a 300 °C-tank at the flow rate of 100 ml/min. The mass change data were collected every 30 s using a balance with a precision of 0.0001 g.

The experiments were carried out in both non-isothermal and isothermal modes. In the non-isothermal experiment, the furnace was firstly heated to 600 °C. Then the sample placed in a Φ 20×25 mm high purity alumina crucible (99.5 mass% Al_2O_3) was put at the hot zone of the furnace and heated from 600 to 1500 °C at the heating rate of 4 °C/min. During the whole experiment, the gas flow was adjusted by the flow meter to keep at 100 ml/min. In view of the isothermal experiments, the following steps were adopted: the furnace was heated to the required temperature as quickly as possible and then the reaction tube was flushed with the reaction gas for 3 h in advance. After establishing the thermal equilibrium, the sample placed in the high purity alumina crucible was put at the hot zone of the furnace and reacted for a certain time. At the end of the experiment, the sample was cooled quickly in argon. According to previous reports, the impurity elements including Al and Ca can enhance the oxidation behavior of ceramic materials [31–36]. In our experiment, the spallation phenomenon caused by the oxide product peeled off from the porous Si_3N_4 ceramics is likely to exist at high temperature. Therefore high purity Al_2O_3 crucible is adopted. To minimize the influence of Al_2O_3 crucible, two kinds of steps have been adopted. First is to adopt stabilizing treatment of Al_2O_3 crucible before each experiment. Second, we also perform blank trials as a base line under the same conditions to correct the experimental data and to exclude the possible influence caused by the reaction of water with alumina crucible [32,33].

2.3. Characterization

The crystalline phases before and after reaction were analyzed using X-ray diffraction (XRD: M21XVHF22; MAC Science Co. Ltd., Japan) on a Rigaku DMax-RB anode diffraction instrument equipped with a θ/θ goniometer using Cu Kα radiation (40 kV, 150 mA) from 10° to 90° at a step of 0.02°. The surface and cross-section morphology of the sample were characterized using scanning electron microscopy (SEM: JSM-840A, JEOL, Japan). The element distributions on both the surface and the cross section of the samples after reaction were measured via energy dispersive X-ray spectrometry (EDS). The porosity was characterized by Archimedes method according to ASTMC-20 standard.

3. Results and discussion

3.1. Phase and morphology characterization of porous Si_3N_4 ceramics

XRD pattern of as-received porous Si_3N_4 ceramics is shown in Fig. 1a. It can be seen that all the characteristic peaks correspond to both β-Si_3N_4 and α-Si_3N_4 phases. No other phases exist, indicating the prepared Si_3N_4 ceramics are with high purity. The microstructure of the porous Si_3N_4 ceramics illustrated in Fig. 1b indicates that two kinds of typical morphologies exist. One is characterized as the prismatic whiskers, which possess smooth surface and cross together (area P_1 in Fig. 1b). Combing the TEM analysis, the prismatic whiskers belong to β-Si_3N_4[29]. The other is characterized as the relatively denser areas located at the bottom of the prismatic whiskers (area P_2 in Fig. 1b). At higher magnification (inset in Fig. 1b), these relatively denser areas are composed of shorter interconnected whiskers. The structure consists of both α and β-Si_3N_4[29]. According to the EDS analysis (Table 1), the chemical compositions of the prismatic whiskers are pure β-Si_3N_4. The relatively denser area contains less amount of O, Al and Ca besides Si_3N_4. These impurity elements should originate from the raw materials, i.e. FeSi75. As for the fraction of α and β phases in the obtained ceramics, the microstructure of the porous Si_3N_4 ceramics is analyzed using SEM at different fields. The proportion of the prismatic whisker, i.e. β-Si_3N_4, is calculated to be 90% by statistical method. From above results, the obtained Si_3N_4 ceramics should be porous due to the large amount of the prismatic β-Si_3N_4 whiskers. The porosity is measured to be 46% according to Archimedes method.

3.2. Corrosion behavior

Fig. 2 shows the non-isothermal reaction behavior of porous Si_3N_4 ceramics from 600 to 1500 °C at the heating rate of 4 °C/min in dry O_2 and O_2 containing 20 vol% H_2O respectively. It can be seen that under the two conditions, both the reactions exactly begin from approximate 1000 °C and the reaction rates obviously increase from 1200 °C until 1500 °C. By comparison, the total mass gain per unit area of the sample in O_2 containing 20 vol% H_2O (8.64 mg/cm^2) is higher than that in dry O_2 (6.72 mg/cm^2), suggesting that water vapor enhances the reaction rate.

Based on the non-isothermal reaction result, the temperature range of the isothermal reaction is set at 1200–1500 °C for 10 h with the interval of 100 °C. The corresponding curves are shown in Fig. 3, in which x and y axes represent the reaction time (h) and the mass gain per unit area (mg/cm^2) respectively. Fig. 3a represents the reaction behavior of the porous Si_3N_4 ceramics in dry O_2. It can be seen that the total mass gain per unit area increases with reaction temperature

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