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Short communication

Oxidation protection of SiC in porcelain tile ceramics by adding Si powder

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ABSTRACT

Polishing porcelain tile residues (PPR) have accumulated to millions of tons and increases every year. It has become important to recycle the PPR due to the economic gains and environmental sustainability of doing so. However, its direct recycling in the production of porcelain tile has been limited by the fact that SiC particles oxidise during sintering which prevents the processing of dense ceramics. In this study, Si powder was introduced into the porcelain ceramics matrix containing SiC. The effect of Si powder additive on inhibiting the oxidation of SiC was evaluated using the relative density and the microstructure of the sintered samples. Results showed that the porcelain body containing SiC could be fully densitified when the suitable content of Si powder addition is introduced. These results indicate that it is a promising method to effectively protect the oxidation of SiC particles in the porcelain ceramics matrix during sintering.

1. Introduction

Porcelain stoneware tile is an advancement in the sector of ceramic tile development [1]. Its low porosity is an essential feature which gives it excellent mechanical and chemical properties and makes the material stain and frost resistant [2,3]. In recent years, there has been an increase in porcelain stoneware tile manufacturing. In China, 3000 million m^2 of porcelain stoneware tile has been produced annually and made up more than half of the total ceramic tile production [4–6].

Porcelain stoneware tile is usually surface-polished to improve its aesthetic quality and increase its competitiveness with natural stone [7,8]. During the polishing process, around 0.4–0.8 mm of the porcelain tile surface is removed to create a gloss surface level of 65–70%. This results in the generation of large amounts of polishing porcelain stoneware tile residues (PPR). It has been reported that the output of PPR has reached about 6 million tons per year in China and increases every year [5,6].

Not only does disposal of PPR cause serious environmental pollution but it also wastes mineral resources which increases the production cost. Therefore; the search for new recycling technologies is of great technological, economic, and environmental importance [9–13]. A valid alternative to landfill confinement of PPR could be the direct recycling back into the production of porcelain stoneware tile. This could form a closed process that, following a logical chain of events, allows the consumption of the waste in the same production line as the starting material. However, previous investigations reported that the relevant amount of silicon carbide prevented the processing of dense ceramics [5,6,14]. At the usual firing temperatures adopted in the production of ceramic tiles, >1100 °C, silicon carbide (SiC) decomposes in the presence of oxygen and releases gases such as SiO, CO and CO_2, giving rise to porous microstructures [5,6,11,13]. The related reactions are shown in Eqs. (1)–(3).

$$SiC + 2O_2 \rightarrow SiO_2(s) + CO_2(g) \quad (1)$$

$$SiC + O_2 \rightarrow SiO(g) + CO(g) \quad (2)$$

$$2SiC + O_2 \rightarrow 2SiO(g) + C(s) \quad (3)$$

PPR were mostly recycled when incorporated into low density ceramic products, which still cannot essentially solve the landfill problem from the large amount of PPR yet. Therefore, a method that could avoid the oxidation of SiC particles in the porcelain ceramics matrix is urgently needed.

In this study, Si powder was introduced into the porcelain tile ceramics to avoid the oxidation of SiC particles. It was expected that SiC would be protected by the oxygen consumption of Si. In addition, SiO_2 was a type of silicate network which formed oxides, so it was possible that the oxidation resultant of Si would enter the silicate network and promote the bonding strength of the sili-

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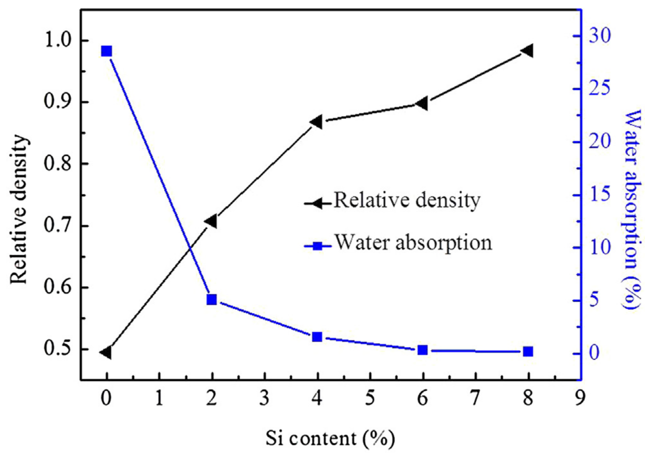

Fig. 1. Effect of Si additive content on the relative density and water absorption of the porcelain body containing 2 wt% SiC fired at 1200 °C for 20 min.

cates. With this in mind, the oxidation behavior of SiC in porcelain ceramics matrix was studied by adding Si powder. The effect of Si additive on inhibiting the oxidation of SiC was evaluated by the relative density and the microstructure of the resultant samples with different amounts of Si additive.

2. Experimental procedures

A porcelain stoneware tile powder (Newpearl Ceramics Co. Ltd., Guangdong, China) as the main raw material was obtained by the shattering and planetary-milling of porcelain stoneware tile for 45 min. The particle size range of the powder was between 0.1 and 45 μm, and the median diameter was 6.2 μm, which was measured using laser diffraction/scattering (Mastersizer 2000, Malvern) with distilled water as dispersant. The composition of the porcelain tile was as follows: (SiO_2 69.01 wt%, Al_2O_3 22.29 wt%, Fe_2O_3 0.44 wt%, TiO_2 0.17 wt%, CaO 0.54 wt%, MgO 0.48 wt%, K_2O 1.35 wt%, and Na_2O 4.93 wt%), which was measured by silicate chemical composition analysis instrument (DHF86, Xiangtan Songshan instrument Co.,Ltd., China). Silicon carbide powder (Haixu abrasive Co. Ltd., Zhengzhou, China) was used as an additive. The purity was more than 99.8 mass%, and the particle size was defined using the standard mesh-size designation 600 mesh corresponding to an average particle size 23 μm (as measured from SEM images). The SiC powder was a typical abrasive used in the porcelain tile polishing process. Si powder was commercially available (Jinan Tianqin Silicon Industry Corp. Ltd, Jinan, China), and the average particle size was about 6.5 μm, which was estimated using the following equation: $D_{BET} = 6/(\rho \times S_{BET})$. The specific surface area (S_{BET}) was determined by the nitrogen gas adsorption based on the BET multipoint method (BET, ASAP-2010, Shimadzu, Tokyo, Japan), and ρ is the theoretical density of Si powder. The slurries of the porcelain tile powder, 2 wt% SiC and different contents of Si powder, were prepared with distilled water and 0.5 mass% of polyethylene glycol (PEG-400 binder, Guangzhou Taiqi Chemical Technology Co., Ltd, China) in the water: mixture mass ratio of 0.8: 1, respectively. The slurries were planetary-milled for 30 min. The raw material powders were prepared via drying, granulating and sieving (in a sieve with 80 mesh, i.e., 180 μm). The porcelain green bodies with a 30 mm diameter and a 5 mm thickness were formed by dry-pressing the raw material powders at 15 MPa. The green bodies were then fired in an electric furnace with an air temperature of 1200 °C for 20 min with a heating rate of 10 °C/min.

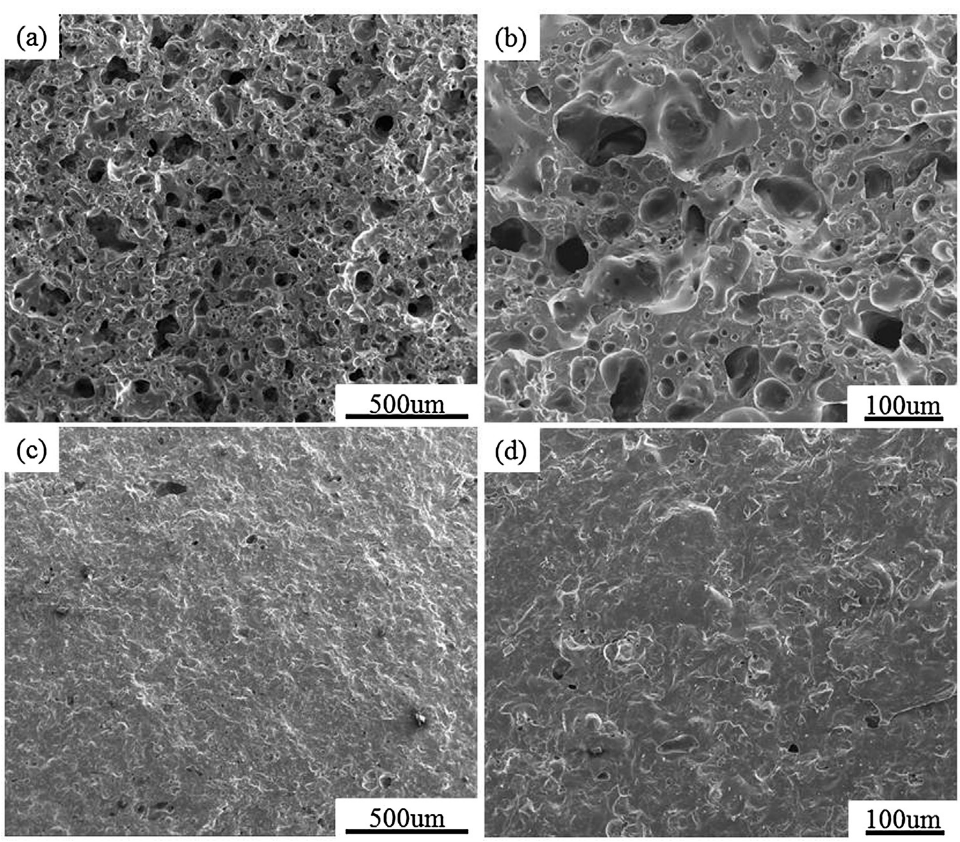

Fig. 2. SEM images of porcelain body containing 2 wt% SiC without Si powder addition (a) and (b); and with 8 wt% Si powder addition (c) and (d) at the firing temperature of 1200 °C for 20 min.

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