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Effect of sintering additives on the densification, crystallization and flexural strength of sintered glass-ceramics from waste granite powder

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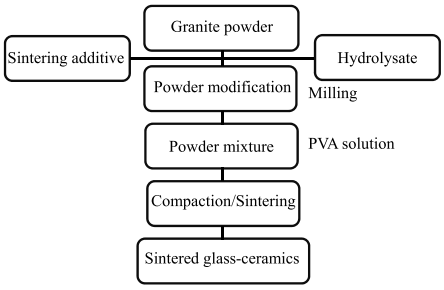
HIGHLIGHTS

- The boehmite was preferable for the densification and strengthening.
- The crystallinity decreased with increasing the sintering temperature.
- The crystallinity increased with increasing the boehmite content.
- The flexural strength depended on densification and crystallinity.

GRAPHICAL ABSTRACT



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ABSTRACT

Large amounts of granite wastes in the stone processing industry should be recycled to prevent the environmental pollution and health hazards. In this work, the sintered glass-ceramics have been prepared from granite powder by the sintering-crystallization method. The sintering additives were investigated to reveal their influence on the densification, crystallization and flexural strength of glass-ceramics. Compared to colloidal silica and float glass powder, the boehmite sol was more effective in the strengthening of glass-ceramics. The crystallinity of glass-ceramics decreased with increasing the sintering temperature due to the melting of microcline phase, and the microstructure was typical of liquid phase sintering. The crystal phases included anorthite, quartz and hematite. As the boehmite content was increased, the flexural strength firstly increased and then decreased, showing an optimal content of 3 wt%. The maximum flexural strength (125 MPa) was far superior to that of natural granite. The crystallinity continuously increased with the increase in the boehmite content. The sintered glass-ceramics with decorative patterns and high flexural strength promise the practical reutilization of granite wastes in the ornamental tiles.

1. Introduction

For the granite production and consumption, China has been one of the main countries in the world. The gross production of granite slabs amounts to 730 million square meters in 2016 [1]. Being ~60% of the granite blocks, granite wastes should be properly disposed to prevent the environmental pollution and health hazards [2,3]. The reutilization of granite wastes not only gets rid of the environmental problems, but also ensures the sustainable granite exploitation [4].

The granite wastes have been usually recycled into the construction and building products to reduce the consumption of natural minerals [5,6]. They were incorporated in the cements and concretes as the aggregate, filler and mineral additive without the detrimental effects on the workability, water absorption and durability. With the feldspar

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constituents, granite powder was used as the fluxing agent in the porcelain tiles and ceramics [7,8]. In addition, granite wastes were applied in the treatment of wastewater sludge, acid soil and radioactive waste [9–11].

Regarding the profitable reutilization, granite wastes are promising in such applications as glaze and glass-ceramics. When the diopside glaze was prepared from glass-ceramic frit and granite waste, the iron content played a crucial role in controlling the crystalline composition, crystallization and glaze appearance [12]. Transparent and opaque glazes from the combination of float glass, granite and lime shale exhibited the comparable hardness and corrosion/abrasion resistance to that of commercial glazes [13]. With the addition of other chemicals/minerals, the glass-ceramics could be prepared from granite wastes by the melting-crystallization method, exhibiting the excellent mechanical properties [14,15].

Unlike the common melting-crystallization process, the sintered glass-ceramics were simply prepared by the simultaneous sintering-crystallization process from the powder mixtures [16]. In this work, the modified granite powders were directly sintered into high strength glass-ceramics. The sintering additives were investigated to reveal their influence on the densification, crystallinity and flexural strength of glass-ceramics. The microstructural evolution was carefully examined to clarify the densification process. The strengthening mechanism of glass-ceramics was discussed in terms of the densification and crystallinity. The decorative glass-ceramic was preliminarily prepared to demonstrate its commercial feasibility in ornamental tiles.

2. Materials and methods

2.1. Materials

To remove the soluble impurities, grey granite powder from a stone processing factory was washed with distilled water, filtered and dried, and passed through a 120-mesh sieve. The silicane coupling agent of γ-glycidoxy-propyltrimethoxysilane (Silquest A-187) was selected as the surface modifier of granite powder. The sintering additives included aqueous boehmite sol (γ-AlO(OH), 20 wt%, pH = 4.5), colloidal silica (30 wt%, pH = 9.0) and self-made float glass powder (200 mesh). Analytically pure methanol was purchased from Sinopharm Chemical Reagent Co. An aqueous polyvinyl alcohol solution (PVA, 2 wt%) was used in the powder compaction. Zirconia beads (NanorZr-93, ∅ 2–5 mm) as the milling media were from Guangzhou Pleased Grinding Media Co.

2.2. Preparation of sintered glass-ceramics

The processing route of sintered glass-ceramics was schematically illustrated in Fig. 1. In the powder modification process, the modifying solution was prepared with methanol (97 wt%), A-187 (2 wt%) and distilled water (1 wt%). After magnetically stirred for 25 min, the hydrolytic solution was mixed with 60 wt% of granite powder in a polypropylene bottle. Afterwards, the sintering additives and zirconia beads ten times the mass of granite powder were added sequentially. The sintering additives were 3 wt% relative to the granite powder, and the boehmite could be adjusted at 0.5–5 wt%. The powder suspension was ground in a planetary ball mill at 300 rpm for 1 h. Finally, the milled slurry was separated from the zirconia beads, dried at 80 °C for 1 h, and passed through a 200-mesh sieve.

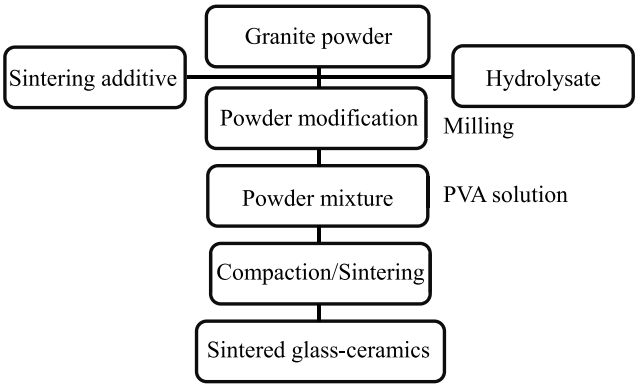

Fig. 1. Schematic illustration of the processing route of sintered glass-ceramics.

The powder mixtures were uniformly blended with 20 wt% of PVA solution, and uniaxially pressed in a rectangular mould at 100 MPa for 5 min. The green compacts were thermally treated at 500 °C for 1 h to remove the PVA binder, and sintered in an electrical furnace with a heating rate of 10 °C/min at 1050–1150 °C for 2 h. The sintered glass-ceramics were ground with SiC papers and polished with diamond paste.

2.3. Characterization and physical properties of glass-ceramics

The chemical composition of granite powder was analyzed by the wave length dispersive X-ray fluorescence (S4 PIONEER, Bruker AXS, Germany). The crystal structure of granite powder and sintered glass-ceramics was examined by X-ray powder diffraction (XRD, D8 Advance, Bruker AXS, Germany) using Cu K_α radiation. The volume percentage of different phases and crystallinity of glass-ceramics were estimated from the XRD pattern simulation using the MDI Jade 6 software [17,18]. The thermal behavior of granite powder was evaluated by the differential thermal analysis and thermogravimetry (DTA/TG, STA 449C, Netzsch GmbH, Germany) with a heating rate of 10 °C/min in an air flow (50 ml/min).

The bulk density of glass-ceramics was measured by using the Archimedes' method. The densification of glass-ceramics was observed with an environmental scanning electron microscope (SEM, Quanta 200, FEI, USA). The phase distribution of glass-ceramics was analyzed with the backscattered electron imaging using a field-emission scanning electron microscope (FESEM, Nova NanoSEM 450, FEI, USA), which was equipped with an energy dispersive spectrum analyzer (EDS, Inca 250 X-Max 50, Oxford Instruments, UK). Prior to the SEM and FESEM observations, the sample surfaces were deposited with gold films to improve the electrical conductivity. The flexural strength of glass-ceramics was measured in the three-point bending tests with a universal testing machine (WDW-50, Jinan Shijin, China). The cross-head speed was kept at 0.5 mm/min. All rectangular samples were 45 mm × 5 mm × 4 mm in size, and were chamfered to eliminate the stress concentration. Each data point represents an average value of at least five individual tests.

3. Results and discussion

3.1. Characterization and thermal behavior of granite powder

The granite powder was characterized by the particle morphology, crystal structure and chemical composition. As shown in Fig. 2, microsized particles exhibited sharp edges and cleaved surfaces, characteristic of the brittle and stiff fracture of granite stone. The crystal structure of granite powder consisted of quartz (PDF 46–1045), anorthite ($Na_{0.45}Ca_{0.55}Al_{1.55}Si_{2.45}O_8$, PDF 85–1415) and microcline ($KAlSi_3O_8$, PDF 72–1114) phases. The volume percentage of quartz and microcline was estimated as 66% and 11%, respectively. Owing to high similarity in the XRD patterns, the albite phase ($K_{0.2}Na_{0.8}AlSi_3O_8$, PDF 83–2215) was hardly discernible from the anorthite phase. An approximate estimation by the difference of minor diffraction peaks resulted in 14% anorthite and 9% albite. Practically, the albite and anorthite phases formed the solid solutions of plagioclase with different proportions [19].

The chemical composition of granite powder is shown in Table 1.