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Influence of different reinforcements on toughening and strengthening of sintered stoneware from modified marble powder

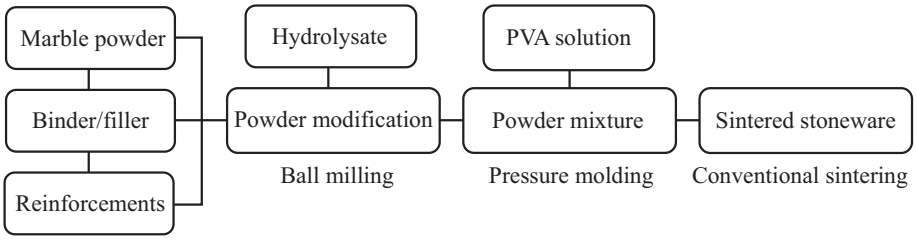
Jinshan Lu [a,b,*], Yingde Li [a], Xinquan Cong [a], Yong Hao [b], Chunlei Wang [b]

[a] School of Materials Science and Engineering, Nanchang Hangkong University, Nanchang 330063, China
[b] College of Engineering and Computing, Florida International University, FL 33174, USA

HIGHLIGHTS

- Mullite fibers and boehmite platelets were used as the reinforcements.
- The reinforced stoneware was greatly improved in the mechanical properties.
- The toughening mechanisms involved the crack deflection and bridging.
- The strengthening mechanisms included multiple factors.
- The synergetic toughening derived from the platelet-induced strengthening.

GRAPHICAL ABSTRACT



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ABSTRACT

In the stone processing industry, huge amounts of marble wastes should be properly disposed to prevent the environmental pollutions and health hazards. In this work, waste marble powder was recycled into artificial stoneware by means of surface modification and low temperature sintering. Mullite fibers and boehmite platelets were used as the reinforcements to improve the mechanical properties of sintered stoneware. For the single reinforcements, the flexural strength and fracture toughness firstly increased and then decreased with increasing the reinforcement content. There exists a percolation threshold of reinforcements to form the uniform reinforcement networks with respect to the optimization of mechanical properties. The increase of compaction pressure for the powder compacts promoted the densification and strengthening of stoneware. The composite reinforcements with fibers and platelets exerted the synergetic effect on the toughening of stoneware. As for the mechanical properties, the reinforced stoneware was high superior to the unreinforced stoneware and natural marble. The toughening mechanisms involved crack deflection and crack bridging, while the strengthening mechanisms included the reduction of flaw size, enhanced interface adhesion and toughening of stoneware. The reinforced stoneware with excellent mechanical properties was promising for the reutilization of marble wastes in construction tiles.

1. Introduction

Marble wastes are the by-products from the marble quarrying, cutting and polishing. With the booming demand of marble products, huge amounts of marble wastes should be properly disposed for the environmental protection and healthcare [1,2]. Besides the simple landfill in the abandoned quarries, various attempts have

* Corresponding author at: School of Materials Science and Engineering, Nanchang Hangkong University, Nanchang 330063, China.
E-mail address: 1474202488@qq.com (J. Lu).

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been made to explore the eco-efficient, profitable and sustainable reutilizations of marble wastes [3,4].

Marble wastes have been used to replace the natural aggregates in the concretes [5,6]. The river sands could be completely substituted with marble scraps to improve the workability and mechanical properties of concretes. There was insignificant difference in the concrete durability between coarse marble aggregates and conventional primary aggregates. As a mineral additive, waste marble powder was used to reduce the amount of cements in the concretes without the noticeable effect on the hydration process [7]. In the environmental management, marble wastes were applied in the neutralization of acid mine drainage, immobilization of heavy metals and removal of pollutants from industrial wastewater [8,9]. Furthermore, marble powders were used as renewable CO_2 adsorbent, feedstock of high purity $CaCO_3$ and fluxing agent [10–12].

Despite the extensive researches, practical recycling of marble wastes is limited relative to huge amounts of annual production. Considering the profitable and massive reutilization, the sintered stoneware prepared from marble wastes is preferable to the aggregates. This disposal could not only solve the pollution problems, but also extend the industrial chain of construction materials. In a previous work, the authors proposed a facile and scalable approach to prepare sintered stoneware from waste marble powder [13]. The combination of surface modification, sintering binder and inert filler significantly improved the densification and flexural strength of stoneware. Confined by the thermal decomposition of marble powder, the sintering process was conducted at low temperatures ($\leq$590 °C). Thus, the mechanical properties were not satisfactory, and should be further improved to meet the technical requirements of construction tiles.

It is well known that the incorporation of extrinsic reinforcements (e.g. fibers, whiskers and platelets) greatly increase the mechanical properties of brittle materials [14,15]. The present work described the toughening and strengthening of sintered stoneware with the reinforcements of mullite fibers and boehmite platelets. The effects of reinforcements and compaction pressure on the densification, microstructure and mechanical properties were systematically investigated to clarify the toughening and strengthening mechanisms. The combination of fibers and platelets was also evaluated in terms of the microstructure and fracture toughness.

2. Materials and methods

2.1. Materials

White marble powder (sawing dust) was collected from a stone processing plant. The surface modifier of γ-glycidoxypropyl trimethoxysilane (Silquest A-187) was provided by Foshan Daoning Chemical Co. Analytically pure methanol was purchased from Sinopharm Chemical Reagent Co. The sintering binder was an aqueous boehmite sol (20%, pH = 4.5) with an average particle size of 20–30 nm. The ground $CaCO_3$ powder (2000 mesh) was used as the inert filler. An aqueous polyvinyl alcohol (PVA) solution (5%) was used as the room temperature binder in the powder compaction. The chopped mullite fibers were obtained from Deqing Nengcheng Crystal Fiber Co. Boehmite platelets were supplied by Guangzhou Xinxi Chemical Co. Zirconia beads (NanorZr-93, ∅ 2–5 mm) were used as the milling media in the powder modification.

2.2. Preparation of sintered stoneware

The preparation process of sintered stoneware is schematically illustrated in Fig. 1. Prior to the surface modification of marble powder, a milling solution was prepared with methanol (97%), A-187 (2%) and distilled water (1%). After magnetically stirred for 25 min, the hydrolysate was mixed with marble powder in a polypropylene bottle, with a weight ratio of 3:4. Afterward, the sintering binder (3%) and filler (2%) were added successively, and followed by the addition of different reinforcements including mullite fibers (MF, 1–8%) and boehmite platelets (BP, 0.5–4%) with the predetermined proportion relative to the mass of marble powder. Finally, zirconia beads ten times the mass of marble powder were added in the mixed solution. The powder mixtures were mechanically milled in a planetary ball mill at 380 rpm for 1 h. In the milling process, the marble powder was modified with hydrolytic silanol groups, and uniformly mixed with other components. The milled slurry was separated from zirconia beads, dried at 100 °C for 1 h, and passed through a 120-mesh sieve.

The as-processed powder mixtures were blended with 10% of aqueous PVA solution to improve the powder formability, and uniaxially pressed at 422 MPa in a rectangular mould. The compaction pressure could be changed from 211 MPa to 581 MPa. In the sintering process, green compacts were thermally treated at 500 °C for 1 h to remove the PVA binder, and sintered at 590 °C for 90 min with a heating rate of 5 °C/min. The sintered stoneware was ground with SiC papers and polished with diamond paste.

2.3. Characterization and mechanical properties of sintered stoneware

The particle size distribution of marble powder was measured using a laser scattering particle size analyzer (LA-950, Horiba, Japan). The crystal structure of powders and stoneware was identified by X-ray powder diffraction (XRD, D8 Advance, Bruker AXS, Germany) using Cu K_γ radiation. The phase content and crystallinity were estimated from the XRD pattern simulation using the MDI Jade 6.0 software. The bulk density and water absorption of stoneware were measured by using the Archimedes' method. The densification of stoneware was examined with a polarizing microscope (PM, XP-600E, Shanghai Bimu, China). To clearly distinguish the surface pores, the polished samples were stained with black ink.

The microstructure of powders and stoneware was observed with a scanning electron microscope (SEM, S-3400N, Hitachi, Japan), which was equipped with an energy dispersive spectrum analyzer (EDS, Inca 250 X-Max 50, Oxford Instruments, UK). The samples were deposited with gold films to improve the electrical conductivity. The flexural strength of stoneware was determined 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.

The Vickers hardness and fracture toughness of sintered stoneware were measured by the indentation method with a digital Vickers hardness tester (HVS-1000, Jinan Fangyuan, China), and the indentation procedure was performed with a load of 9.807 N for 20 s. The Vickers hardness H_V (MPa) and fracture toughness K_{IC} (MPa·$m^{1/2}$) were calculated by using the following equations [16]:

$$H_V = 1.854\frac{P}{(2a)^2} \quad (1)$$

$$K_{IC} = 0.0181E^{0.4}H_V^{0.6}a(c-a)^{-0.5} \quad (2)$$

where P is the indentation load (N), $2a$ is the indentation diagonal size (mm), E is the elastic modulus of marble (55 GPa), c is the radius of the surface crack (mm). As a control, the natural marble was measured under the same conditions.

3. Results and discussion

3.1. Characterization of marble powder and reinforcements

Fig. 2 shows the SEM images of raw materials used in these experiments. The marble particles were mostly 1–4 μm in size. The average particle size was 10.8 μm, implying the slight agglomeration of primary particles. The sharp edges and layered structure of marble particles are indicative of the brittle fracture of marble during the cutting and polishing processes. As shown in Fig. 3(a), the XRD pattern of marble powder indicates a single calcite phase (PDF 05-0586) without any other phases.

In Fig. 2(b), the chopped fibers had a length of 100–500 μm and a diameter of 5–10 μm. Ultrafine particles adhered to the fiber surface in the inset image, and a few coarse grains (yellow circles) distributed along with the fibers, which should be ascribed to the detached slags in the solution processing [17]. As shown in Fig. 3 (b), the MF powder consisted of mullite ($Al_{2.3}Si_{0.7}O_{4.85}$, PDF 74-2419), corundum (Al_2O_3, 71-1124) and amorphous phases. The phase content of mullite and corundum was estimated to be 70.9% and 7.7% respectively, thereby the crystallinity amounted to 78.6%. The phase composition was consistent with that of commercial fibers, and the corundum and glassy phases were associated with the incomplete crystallization of fibers during the thermal treatment [18].