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High strength artificial stoneware from marble waste via surface modification and low temperature sintering

Jinshan Lu [a, b, *], Xinquan Cong [a], Yingde Li [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

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ABSTRACT

Huge amounts of marble wastes pose serious hazards to the environment, water resource, vegetation growth as well as public health. In this work, high strength artificial stoneware was prepared from waste marble powder by the combination of surface modification and low temperature sintering. The marble powder was initially modified with silicane coupling agent, and then sintered into stoneware with the sintering binders (boehmite and silica) and filler ($CaCO_3$). The processing parameters were investigated to clarify their effects on the bulk density and flexural strength of sintered stoneware. The elevated sintering temperatures and prolonged sintering times resulted in the thermal decomposition of calcite phase, so that the optimum sintering process was achieved at 590 °C for 90 min. Compared to silica sol, boehmite sol was more effective to increase the flexural strength. The addition of ground $CaCO_3$ powder facilitated the densification and strengthening, while the excessive filler caused the agglomeration of filler particles and reduction of bulk density and flexural strength. The maximum flexural strength of stoneware (42.2 MPa) is high superior to that of original marble slabs. The strengthening mechanism was qualitatively discussed in terms of the densification and interface adhesion. The facile and scalable approach and high strength stoneware pave the way to the massive recycling of marble wastes in the ornamental construction tiles.

1. Introduction

In the past decade, the annual growth rate of marble production is about 15% in China, and the marble products amounts to 350 million square meters in 2016 (Secretariat of the China Stone Association, 2017). It has been reported that 70% of marble resource was converted into marble wastes after the processes of quarrying, cutting and polishing (Hebhoub et al., 2011), which should be properly disposed to eliminate the ecological and public health hazards (Simsek et al., 2005; Rizzo et al., 2008).

Despite the widespread reutilization of solid wastes, marble wastes were primarily dumped in the landfills due to the limited economic profit (Gencel et al., 2012). The effective recycling of marble wastes could be realized by the development of high value-added products (Careddu et al., 2014). So far, marble wastes were generally recycled into the construction materials as coarse and fine aggregates in the concrete and a mineral admixture of cement (Alyamac et al., 2017; Arel, 2016). In the case of self-compacting concrete, marble powder exhibited excellent performance as sand replacement without the adverse effect on the hydration process (Sadek et al., 2016).

As an alkaline mineral, waste marble powder was used to neutralize the acidic wastewater, and remove harmful metals from aqueous solutions (Mehta et al., 2016; Mlayah and Jellali, 2015). For instance, heavy metals in the industrial wastewater could be removed with an efficiency of >85% by the zeolite adsorbent from marble powder (Javed et al., 2016). Marble wastes were suitable to the remediation of acidic and hazardous mine soils for their suitability of native vegetation growth and agricultural cultivation (Kaba et al., 2012; Tozsin et al., 2015). Moreover, there were various reutilizations in other fields, including inexpensive CO_2 sorbent (Pinheiro et al., 2016), feedstock of $CaCO_3$ powder (El-Sherbiny et al., 2015), adhesive filler (Özkaya et al., 2015) and fluxing agent of ceramics (Yeşilaya et al., 2017).

Regarding the massive and profitable reutilization, the preparation of artificial stoneware from marble wastes is preferable, which helps to extend the industrial chain of stone processing. In

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

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this work, high strength stoneware was prepared from waste marble powder by low temperature sintering. The marble powder was chemically modified with silicane coupling agent to increase its sintering activity, and sintered with sintering binders to enhance the interface adhesion of sintered stoneware. The ground $CaCO_3$ powder was added as an inert filler to increase the densification and flexural strength. The relationships between the processing parameters and bulk density/flexural strength were studied in detail to reveal the strengthening mechanism of sintered stoneware.

2. Materials and methods

2.1. Materials

The white marble powder (sawing dust) was collected from a stone processing plant. To remove the soluble impurities, the marble powder was mechanically washed with distilled water at a stirring speed of 800 rpm for 1 h. After filtered and dried at 120 °C for 2 h, the as-processed powder was passed through a 120-mesh sieve.

The surface modifier of γ-glycidoxypropyltrimethoxysilane (Silquest A-187) was supplied by Foshan Daoning Chemical Co. The sintering binders included aqueous boehmite (γ-AlOOH) sol (20%, pH = 4.5) and neutral silica sol (30%, pH = 7.0), both of which had an average particle size of 20–30 nm. Analytically pure methanol was purchased from Sinopharm Chemical Reagent Co. The ground $CaCO_3$ powder (2000 mesh) was used as the inert filler. The aqueous polyvinyl alcohol (PVA) solution (2%) was used as the room temperature binder in the powder compaction. Zirconia beads (NanorZr-93, ∅ 2–5 mm) as the milling media were purchased from Guangzhou Pleased Grinding Media Co.

2.2. Preparation of sintered stoneware

The 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 (300 ml) at a weight ratio of 0.75. The sintering binder and filler were added successively, followed by the addition of zirconia beads ten times the mass of marble powder. Afterwards, the mixture was mechanically blended by using a planetary ball mill (XQM, Changsha Tianchuang Technology Co.) at 380 rpm for 1 h. Eventually, the milled slurry was separated from zirconia beads, dried at 80 °C for 1 h, and passed through a 120-mesh sieve.

The modified powder was uniformly mixed with the aqueous PVA solution to improve the powder formability. It was uniaxially pressed in a rectangular mould at 200 MPa, and dried at 200 °C for 2 h. In the sintering process, 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 5 °C/min. The sintering process was conducted at 530–630 °C for 10–240 min. The sintered stoneware was ground with SiC papers, and polished with diamond paste.

2.3. Characterization of marble powder and properties of sintered stoneware

The chemical composition of marble powder was analyzed by the wave length dispersive X-ray fluorescence technique (XRF, S4 PIONEER, Bruker AXS, Germany). The size distribution of marble powder was detected by using a laser scattering particle size analyzer (LA-950, Horiba, Japan). The crystal structure of marble powder and stoneware was determined by X-ray powder diffraction (XRD, D8 Advance, Bruker AXS, Germany). The thermal behavior of marble powder was evaluated by the differential thermal/thermogravimetric analysis (DTA/TG, STA 449C, Netzsch GmbH, Germany) with a heating rate of 10 °C/min in air flow (100 ml/min). The structural variation of calcined marble powders was monitored by Fourier transform infrared spectroscopy (FTIR, Spectrum One, Perkin Elmer, USA) with a resolution of 0.5 cm^{-1}.

The bulk density of sintered stoneware was measured by using the Archimedes' method. The densification of stoneware was examined with a polarizing microscope (PM, XP-600E, Shanghai Bimu, China). The microstructure was observed with a scanning electron microscope (SEM, S-3400N, Hitachi, Japan), and the surface mapping of elemental distribution performed with an attached energy dispersive spectrum analyzer (EDS, EMAX Energy, Horiba, Japan). Prior to the SEM observation, the sample surface was deposited with a gold film to improve the electrical conductivity. The flexural strength of sintered 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 remove the stress concentration. Each data point represents an average value of at least five individual tests.

3. Results and discussion

3.1. Characterization of marble powder

Fig. 1 shows the SEM image and XRD pattern of marble powder. The irregular shaped particles are mostly 2–5 μm in size, and exhibit the layered structure in the inset image. The crystal structure of marble powder was determined to be a single calcite phase (PDF 05-0586). In Fig. 2, the size distribution of marble powder

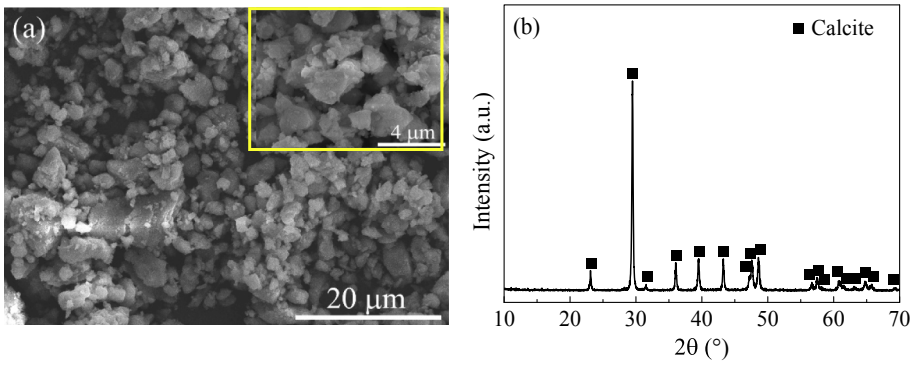

Fig. 1. Morphology and structure of marble powder. (a) SEM image, (b) XRD pattern.