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Mechanical behaviour and microstructure of an artificial stone slab prepared using a SiO_2 waste crucible and quartz sand

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HIGHLIGHTS

- We prepared a high-strength artificial stone slab using a SiO_2 waste crucible.
- The mineral and chemical characteristics of the SiO_2 waste crucible were studied.
- The effects of the UPR, coupling agent, compaction and cure time were investigated.

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ABSTRACT

This paper presents an experimental investigation of the production of a high-quality artificial stone slab using a SiO_2 waste crucible. A fine SiO_2 waste crucible powder and quartz sand were mixed with unsaturated polymer resin (UPR) as a binder, and all samples of artificial stone slabs were prepared using vibratory compaction in a vacuum environment. The properties of the artificial stone slabs as a function of different coupling agent contents, UPR contents, compaction times and curing time were studied using compression, three-point bending, bulk density and water absorption tests. The structural features of the samples were also characterized. Compaction served to prevent the formation of pores between particles; however, excessive compaction crushed the particles. The compressive and flexural strength of the samples increased as the curing time increased. Optimal proportions of the contents of the UPR and coupling agent achieved the most cost-effective production parameters. The artificial stone slabs obtained in this work have high compressive and flexural strengths of 170.9 and 73.5 MPa under a compaction time of 3 min and a curing time of 60 min, which are superior to natural construction slab and most artificial stone slabs.

1. Introduction

Artificial stone slabs have recently become an increasingly important construction material for covering walls or paving floors because of their high bending strength, low water absorption, low porosity and superior abrasion performance [1]. As a high-quality aggregate of artificial stones, quartz sand is now used in massive quantities [2]; however, its preparation requires significant resources. Economic and ecologic considerations point to one demand: sustainability [3–5]. Thus, significant research efforts have focused on finding alternative materials to replace conventional aggregate [6–18].

There are two types of artificial stone: resin-based and cement-based stone. Regarding resin-based artificial stone, the major alternative material is the direct waste generated during stone processing, especially marble waste [1,6–8], waste stone sludge [9–11], and glass fibre-reinforced plastic (FRP) [12]. Another type of alternative material is the waste produced in industrial production such as waste glass [1], fly ash [13,14], and steel slag [15]. For the recycling of marble waste, the best properties of artificial stone slabs have been reported by Lee et al. [1]. In that study, waste glass and stone fragments were recycled as raw materials, and artificial stone slabs with a high compressive strength of 148.8 MPa were obtained. To reduce the porosity and obtain a higher mechanical strength, the vibratory compaction method in a vacuum environment has been commonly adopted, whereas other techniques have been attempted. For example, Ribeiro et al. [6] fabricated artificial ornamental stone (AOS) using marble residue via a resin transfer molding (RTM) process. Although AOS has a lower mechanical, it could be used as interior wall lining tiles with both acoustic and thermal insulation characteristics.

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Some waste materials cannot be used to prepare resin-based artificial stone with a higher strength. They are usually made into cement-based artificial stone and have a lower mechanical strength and economic value, such as stone slurry [16] and limestone dust [17]. Marble residues have also been used to prepare cement-based artificial stone [3–4,18]. Experimental results show that both resin-based and cement-based artificial stones have lower mechanical properties when raw materials are used, such as limestone, marble or travertine stone, which are mainly composed of $CaCO_3$. However, artificial stone slabs synthesized with SiO_2 usually have a higher flexural strength and lower water absorption.

The alternative raw material used in this research is a SiO_2 waste crucible. A fused silica crucible is a container for preparing polycrystalline silicon, and it can be used only once because of the change of phase and volume that occurs during the casting process [19,20]. The amount of crucible waste in China is estimated to be approximately 100.000 tons per year, and the crucible residue is left in the open air and increases environmental risks. Compared with waste glass, which is usually used as an aggregate in concrete [21–24], a SiO_2 waste crucible has better thermal stability and chemical stability. Therefore, research on construction materials based on SiO_2 waste crucibles is valuable. However, to our knowledge, there have been few reports on the fabrication of high-value construction slabs using recycled SiO_2 waste crucibles. This research, which focused on producing a high-quality artificial stone slab using a waste SiO_2 crucible in the laboratory, aimed to offer a solution for recycling huge amounts of SiO_2 waste crucibles that are accumulating in the environment surrounding the related industries.

Parameters that characterize the properties of artificial stone usually include the type and content of coupling agents [25], the type and content of resins [9], curing methods [8], the curing time [8,26] and compaction conditions [1]. Therefore, the effects of the coupling agent content, unsaturated polymer resin (UPR) content, compaction time and curing time on the physical properties of an artificial stone slab made from a SiO_2 waste crucible were experimentally studied in this work. Because of the depleting supply of natural resources, the recycling of SiO_2 waste crucibles into construction material not only creates new products but also prevents environmental pollution.

2. Materials and methods

2.1. Materials

In this study, quartz sand was used as the coarse aggregate, and SiO_2 waste crucible powder was used as the fine aggregate. The UPR, coupling agent and hardener were pre-mixed to produce a binder. Details of the materials used are provided below.

Chemical analysis was conducted on the quartz sand and SiO_2 waste crucible using inductively coupled plasma optical emission spectroscopy (ICP-OES, Spectro, Blue Sop), and the results are presented in Table 1. Both raw materials are comprised of mainly SiO_2 and a small amount of other elements, and the SiO_2 waste crucible contained approximately 0.1% Si_3N_4, which originated from the crucible coating of Si_3N_4. The similar chemical composition to quartz sand causes the SiO_2 waste crucible to react with the silane coupling agent, which increases the interfacial strength of the sample. Additionally, the SiO_2 waste crucible has a good chemical stability because of its low amount of impurities.

The mineralogy of the SiO_2 waste crucible and quartz sand was assessed via X-ray diffraction (XRD) using a PANalytical X-Pert3 Powder X-ray Diffractometer equipped with Cu X-ray radiation at 40 kV and 40 mA. The Powder XRD was conducted at a reflection angle range of 2θ = 5–50° with a counting step of 0.013° and a scanning speed of 2.25 2θ/min. Fig. 1 shows the XRD patterns of the SiO_2 waste crucible and quartz sand. These patterns were used to identify the mineral phases present in the raw materials. The crystalline phase identified in quartz sand is α-quartz, and the SiO_2 waste crucible is mainly composed of fused silica and α-cristobalite (Fig. 1).

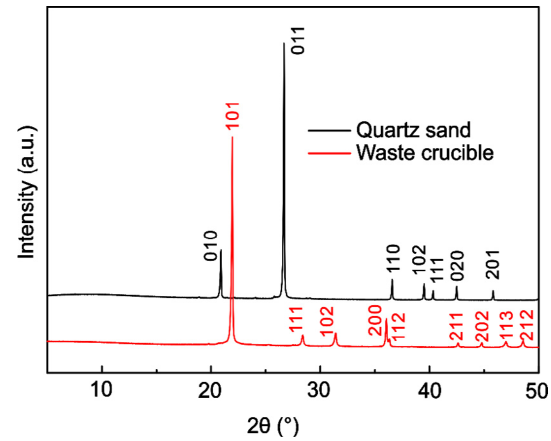

Fig. 1. XRD patterns of the SiO_2 waste crucible and quartz sand.

Table 2
Formulation of aggregates for artificial stone slab.

Raw material	Quartz sand	Quartz sand	SiO_2 waste crucible powder
Mesh number	40–70	70–100	≥200
Mass ratio (%)	50	17	33

The crystalline content of α-cristobalite in the fused silica crucible was measured according to a previous standard method [20] in which we proposed an empirical equation based on XRD analysis for the quantitative calculation of the α-cristobalite content in a SiO_2 waste crucible. The calculation for the crystalline content of α-cristobalite was carried out by using Eq. (1):

$$W_c = 4.3 \times 10^{-4} I_c - 3.53 \quad (1)$$

where W_C is the crystalline content of α-cristobalite, and I_C is the intensity of the 101 diffraction peak in the XRD pattern.

The quantitative phase analysis result shows that the SiO_2 waste crucible contains approximately 57% α-cristobalite and 43% fused silica. The crucible contained only fused quartz before use, and the α-cristobalite was crystallized from fused silica at a high temperature. There are many tiny cracks in the SiO_2 waste crucible because of the change of phase and volume during use. Therefore, the SiO_2 waste crucible is more suitable as a fine aggregate instead of a coarse aggregate.

Quartz sand was used as the coarse aggregate in this work, and it was graded into two categories: (1) between 70 and 40 mesh (0.212 and 0.380 mm) in diameter and (2) between 100 and 70 mesh (0.150 and 0.212 mm) in diameter (Table 2). The recycled SiO_2 waste crucible was crushed and ground into powder and then passed through a 200-mesh sieve, and it was used as a replacement for the fine aggregate. The particle size distribution of the SiO_2 waste crucible powder was evaluated using a laser particle size analyser (MASTERSIZER 2000, Malvern UK) as shown in Fig. 2. The refractive indices of the particles and dispersant are 1.544 and 1.000, respectively. D75 (75% passing size), D50, and D25 are approximately 41, 29, and 21 μm in size, respectively.

The binder used includes UPR (JH-860), a coupling agent (KH-570, γ-Methacryloxypropyl trimethoxy Silane) and a hardener (Methyl ethyl ketone peroxide). The UPR contains 66–72% ortho-phthalic unsaturated polyester and 28–34% styrene, and its viscosity ranges between 0.75 and 1.15 Pas when spindled at 60 rpm at 25 °C; it becomes gelated within 4.2–6.8 min at 25 °C.

Table 1
Chemical composition of the SiO_2 waste crucible and quartz sand.

Element (wt%)	SiO_2	Si_3N_4	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	L.o.I.
SiO_2 waste crucible	99.60	0.11	0.15	0.08	0.03	–	0.01	–	0.01	–	–
Quartz sand	99.29	–	0.43	0.06	0.04	0.02	0.01	0.09	0.03	0.01	0.01