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Effect of glass powder on sulfuric acid resistance of cementitious materials

Hocine Siad ^{a,b,*}, Mohamed Lachemi^a, Mustafa Sahmaran^c, Khandaker M. Anwar Hossain^a

^a Department of Civil Engineering, Ryerson University, Toronto, ON, Canada

^b Laboratory of Materials and Sustainable Development, Bouira University, Bouira, Algeria

^c Department of Civil Engineering, Gazi University, Ankara, Turkey

HIGHLIGHTS

• Effects of glass powder content on the sulfuric acid resistance of mortars were studied.

• Mechanical, physical, and microstructural investigations were carried.

• Sulfuric acid resistance of mortars improved with increased glass powder content.

• Incorporating limestone powder with glass powder improved resistance against sulfuric acid.

• Si/Al-rich residue in glass powder mortars acted as a barrier to acid ions.

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ABSTRACT

In an attempt to enhance the durability of cementitious materials subjected to acid environments, this paper outlines an investigation into the effect of incorporating glass powder (GP) as a cement replacement on mortar resistance against sulfuric acid attack. The study examined mass, compressive strength, ultrasonic pulse velocity (UPV) and electrical resistivity changes of mortars based on 15, 30 and 45% GP replacement rates and 12 weeks immersion in fresh water and 5% acid solutions. The effects of binary binders based on GP and limestone powder (LP), GP and slag (SG) and GP and fly ash (FA) were also investigated. Experimental results showed improved sulfuric acid resistance with increased GP content, and binary binder results confirmed the beneficial effect of incorporating limestone powder with GP. Mortars with 45% GP and binary binder based on 20% GP and 20% LP showed a loss that was significantly lower in terms of physical and mechanical characteristics. Microstructural analysis showed that Si/Al-rich residue, which was surface generated from pozzolanic reaction of GP, has the potential to inhibit further corrosion by acting as a barrier to acid ions. Therefore, incorporating glass powder up to 45% replacement of cement can enhance mortar resistance to aggressive sulfuric acid attack.

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1. Introduction

Concrete structures are regularly subjected to aggressive environmental conditions from a variety of naturally-occurring and industrial chemicals. Sulphuric acid (H_2SO_4) is one of the most destructive to concrete structures due to its presence in ground water, industrial waste and sewage systems [1]. Interaction between concretes and sulfuric acid causes substantial and rapid degradation and damage. Although sulfuric acid is always hazardous to cementitious material [2], cement type and content are

E-mail addresses: hcine_siad@yahoo.fr, hsiad@ryerson.ca (H. Siad).

important factors affecting performance in sulfuric acid environments [3]. According to Turkel et al. [4], the alkalinity of hardened cement binders, which is responsible for cementitious properties, may be partially or completely neutralized when sulfuric acid reacts with hydration products such as calcium hydroxide (CH) or calcium silicate hydrates (C-S-H).

Among the various methods proposed in literature to improve the performance of concretes against sulfuric acid attack [5–7], the incorporation of supplementary cementitious material (SCM) is the most common method, and it has gained great interest. However, despite its popularity, there is still debate over the efficiency of some kinds of SCM in terms of resistance to sulfuric acid attack. Contradictory results have been reported with the replacement of cement by silica fume [8,9], blast furnace slag [10,11], metakaolin





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 $[\]ast\,$ Corresponding author at: Department of Civil Engineering, Ryerson University, Toronto, ON, Canada.

[12,13], natural pozzolan [3,8,14], fly ash [3,15,16] and limestone powder [3,8,12,17]. Limited studies have successfully used PET particles as aggregate [18] or geopolymer as cement [19] to improve resistance against sulfuric acid attack. However, improvement of physical resistance has not exceeded 10%.

Using waste or recycled materials in concrete has attracted a lot of interest worldwide due to concerns about environmental protection and sustainable construction. Waste glasses have therefore been studied extensively as aggregate or cement replacement in concrete [20–24]. The particle size of glass has been found to have major impact on its valorization in concrete mixes. Alkali-silica reaction (ASR) problems were noticed in concretes containing larger particle sizes [20,21]. This concern was addressed by using finely-ground Glass Powder (GP) [25,26]. Moreover, finely ground glass is an effective ASR suppressant that can reduce ASR expansion in concrete by up to 50% [27]. In addition, it has been proven that GP exhibits very high pozzolanic activity [27] equal to or greater than that of fly ash or silica fume [28]. Better strength and transport properties with the use of fine glass particles were also reported for cement replacement rates of up to 30% [28]. Recently, Du and Tan [29] found that replacement of Portland cement with up to 60% GP in concrete production results in better chloride diffusion and migration coefficient, water penetration depth and sorptivity performance. However, a higher cement replacement ratio exceeding 30% [29] reduced strength, which is consistent with the better transport properties. Several studies related to chemical resistance performance suggest that appropriate GP replacement level leads to the refinement of the concrete pore structure through pozzolanic activity, which can improve carbonation and sulfate attack resistance of concretes and mortars [24,30,31].

Very limited studies have been conducted on acid resistance of concretes or mortars incorporating glass aggregates or GP as a cement replacement. Ling and Poon [32] studied self-compacting architectural mortar using recycled blue glass as aggregate replacement and metakaolin as cement replacement. One focus was the effect of different particle sizes of glass aggregates on the mass loss of self-compacting mortar subjected to 3% sulfuric acid attack. Results showed that including glass aggregates in mortar significantly reduced mass loss, up to 100% replacement rate. However, according to Ling and Poon [32], the metakaolin used as an admixture in the cement matrix can be also an important cause of the lower rate of acid attack in glass aggregate mortars. Wang [30] studied the effect of LCD glass powder proportions on cement mortar performance, looking at mass loss under five cycles of one day drying and one day immersion in concentrated H₂SO₄ solution. According to the results of this study, 10% replacement content was optimum for lower mass loss from 10 to 50% substitution; increments in mass loss were also registered in specimens with glass powder replacement exceeding 20%. However, limited exposure time to H₂SO₄ attack (five days) resulted in negligible mass loss differences (between 1 and 2.5%). This, in addition to the single parameter evaluated (mass loss), may have significantly limited the consistency of study results. A review of the aforementioned literature demonstrates the need to study mortars or concretes incorporating glass powder and their response to sulfuric acid attack.

In an attempt to enhance the resistance of cementitious materials against H_2SO_4 attack, a detailed study was undertaken to investigate the effects of different GP contents as cement replacement on the behavior of mortars in a sulfuric acid environment. The mechanical, physical and microstructural performances of mortars were investigated based on 15%, 30% and 45% GP substitution levels. The effects of binary binders when using GP with limestone filler (LF), fly ash (FA) or slag (SG) were also investigated. Resistance against H_2SO_4 attack was characterized by measuring mass loss, compressive strength change, ultrasonic pulse velocity (UPV) and the electrical resistivity change of mortar specimens immersed in the reference medium (fresh water) and in 5% H_2SO_4 solution for twelve weeks. Microstructural changes in the exterior and in the inner layers of degraded samples were also analyzed using scanning electron microscopy (SEM) and X-ray diffraction (XRD).

2. Experimental program

2.1. Materials and mixture proportions

Ordinary Portland cement complying with ASTM C150 Type I cement [33] was used as part of the cementitious materials in the production of different mortars. The finely-ground glass powder was provided by a sorting center and obtained by milling mixed recycled bottle glass. In addition to the glass powder, three other types of mineral admixtures were used to prepare the binary binders: extremely fine limestone powder (LP), Class F fly ash (conforming to ASTM C618 [34]) and ground granulated blast furnace slag (SG) type 100 (conforming to ASTM C989 [35]). The particle size distributions of cement, glass powder and mineral admixtures are shown in Fig. 1, and physical and chemical characteristics are provided in Table 1. Standard sand conforming to ASTM C778 [36] specifications was used for preparing mortars in the testing of the pozzolanic activity index. The natural fine aggregate used in the preparation of test mortar mixtures was river sand with a maximum particle size of 4.75 mm, specific gravity of 2.66 and water absorption of 1.08%.

The pozzolanic activity index of GP, slag and FA was evaluated as a preliminary characterization of these admixtures. 20% of the cement was replaced by GP, slag or FA in accordance with ASTM C 311 [37]. The water-to-cement ratio for the control mixture was 0.485, and for test mortars including the mineral admixture, it was ±5 of control mixture, as required for flow.

In the rest of study, cementitious material and sand amounts were kept constant at 500 kg/m³ and 1375 kg/m³ respectively, which corresponds to one part total binder and 2.75 parts sand, proportioned by mass in accordance with ASTM C 109-13 [38]. In the reference mortar (M0), 100% PC was used as a binder. For GP mortars (MGP15, MGP30 and MGP45), cement was systemically replaced by GP at 15%, 30%, and 45% by weight. To investigate the influence of the binary use of GP and the common mineral admixture, mortars were prepared with 20% GP-20% LP (MGPLP), 20% GP-20% Slag (MGPSG) and 20%GP-20% FA (MGPFA) by mass of binder. To avoid the probable effect of superplasticizer on the hydration of cement or the chemical resistance of mortars, all mixtures were prepared without superplasticizer. The water-to-cement ratio for pure Portland cement (reference) and all GP mortars was fixed at 0.45. The solid materials and water were mixed together according to ASTM C305-14 [39]; mixture proportions are summarized in Table 2. After mixing, specimens were stored in 50 mm cubes and $Ø100 \times 200$ mm cylindrical molds and covered with plastic sheets for 24 h. After demoulding, they were placed in the curing room at 21 °C and 95 ± 5% relative humidity for the chosen durations.

2.2. Testing methods

The pozzolanic activity index of GP and other pozzolanic admixtures was determined in accordance with ASTM C311 [37]. The ratio between the strength of 20% admixtures containing mortar and the strength of the equivalent control mortar was tested at 7, 28 and 90 days of age. Compressive strength data was obtained using $50 \times 50 \text{ xm}$ cubic specimens in accordance with ASTM C109 [38]. Mass change, UPV and electrical *resistivity* were monitored on $\emptyset100 \times 200 \text{ mm}$ cylindrical specimens.



Fig. 1. Particle size distributions of Portland cement, glass powder, limestone powder, fly ash and slag.

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