Materials Characterization 131 (2017) 98–107

Contents lists available at ScienceDirect

Materials Characterization

journal homepage: www.elsevier.com/locate/matchar

Microstructure characterization of alkali-glass particle and alkali-glass powder reacted gels with neutron scattering and imaging techniques

Xiao Sun[a], Shuaicheng Guo[b], Qingli Dai[c,*], Xianghui Xiao[d]

[a] Department of Civil and Environmental Engineering, Michigan Technological University, 1400 Townsend Dr., Houghton, MI 49931-1295, USA
[b] Department of Civil and Environmental Engineering, Michigan Technological University, 1400 Townsend Dr., Houghton, MI 49931-1295, USA
[c] Department of Civil and Environmental Engineering, Michigan Technological University, 1400 Townsend Dr., Houghton, MI 49931-1295, USA
[d] Advanced Photon Source, Argonne National Laboratory, 9700 S Cass Avenue, Argonne, IL, 60439, USA

ARTICLE INFO

Keywords:
Alkali-glass particle reacted gel
Alkali-glass powder reacted gel
(ultra) small angle neutron scattering
Scanning electron microscope
X-ray computed tomography

ABSTRACT

The major obstacles of the incorporation of recycled waste glass in concrete infrastructures are the deteriorations caused by alkali-silica reaction (ASR). Previous research found the concrete containing glass powder does not suffer from ASR. This indicates the size-effect, which is the main focus of this study, is crucial for the incorporation of waste glass in concrete. In order to gather the information for the proposed analytical models, the microstructure and morphology of the synthetic alkali-glass particle and alkali-glass powder reacted gels were characterized by using ultra small angle/small angle neutron scattering (USANS/SANS) and scanning electron microscope (SEM) techniques. The radius of gyration and the power-law constant of different gel samples were obtained by fitting the scattering intensity with a unified Guinier/power-law approach. The power-law constants were in the range from 3 to 4 for the condensed gel sample, indicating the gel particles are surface fractals with different roughness. The SEM images show that the gel particles were spherical-like with sizes around 2 to 3 μm, which is close to the characteristic size (3.4 μm) calculated from the radius of gyration. The dynamic alkali-glass particle reaction was captured with the synchrotron X-ray micro-computed tomography (X-ray micro-CT). The reacted gel particle distributions at different reaction stages were analyzed (up to 64 h). At earlier reaction stage, the majority of gel particles has a size ranged from 2 to 3 μm. At later reaction stages, more large-sized particles were observed in the reaction solution, indicating the volume change of the glass-particle reaction gel.

1. Introduction

The waste glass induces an urgent environmental problem because they cannot be easily reused economically and efficiently. The incorporation of waste glass aggregates into concrete has been attempted to resolve this issue in the past, unfortunately with unsatisfactory results [1,2]. The major obstacle is due to the alkali-silica reaction (ASR) between the high alkaline concrete pore solution and the active silica content from the recycled glass particles. The generated alkali-silica gel could expand due to water absorption. Under strong confinement, it generates internal hydrostatic pressure, which causes the expansion and cracking of the concrete. Understanding the mechanism of the ASR damage is crucial to facilitate the utilization of recycled glass in concrete industry.

A considerable amount of experimental research has been performed on ASR in concrete with natural aggregates, as summarized in [3,4]. Recently, the ASR in concrete with glass aggregates gains attentions due to the large amount of waste glass generated annually. Meyer and his collaborators studied ASR in concrete masonry blocks and precast concrete products [5,6] and reported that there is a pessimum particle size when the maximum expansion occurs. This pessimum size reduces as the reactivity of the glass particle increases. They suggested grinding glass into particles finer than 0.297 mm (passing a U.S. standard No. 50 sieve) to suppress the ASR damage [5]. This observed size-effect phenomenon contradicts to the previous surface reaction assumption of ASR. Three analytical models have been proposed to explain the existence of the pessimum size. Bazant and Steffen established a mathematical model to describe the chemical kinetics of ASR and the corresponding diffusion process [7]. The numerical solution of the model proved that there exists a pessimum size below which the hydrostatic pressure of ASR gel drops, resulting in a less expansion. In addition to the kinetic effects, Bazant et al. [8] simultaneously developed another mathematic model to explain the fracture mechanics of ASR. For sufficiently small particles, the glass volume that results from ASR is independent of particle size since the whole particle has reacted. The tensile strength increases with the decrease of the glass particle

* Corresponding author.
E-mail addresses: xiaos@mtu.edu (X. Sun), sguo3@mtu.edu (S. Guo), qingdai@mtu.edu (Q. Dai), xhxiao@aps.anl.gov (X. Xiao).

http://dx.doi.org/10.1016/j.matchar.2017.07.006
Received 18 January 2017; Received in revised form 30 June 2017; Accepted 1 July 2017
Available online 02 July 2017
1044-5803/

size. At the same time, the stress intensity factor reduces with the decrease of the glass particle size. When these two effects balance with each other, the pessimum size can be determined. Suwito et al. [9] proposed a coupled chemo-mechanical model, arguing that the large interfacial pore space around the small glass particles can accommodate the generated ASR gel, thereby relieving the gel pressure and mitigating the expansion. These three models all provide reasonable explanations for the ASR size-effect on the damage development.

The fine glass powder has been proven as an effective ASR mitigator [10]. For example, Shaya and Xu conducted both laboratory and field studies [11,12] to investigate the performance of glass powders in concrete. No deleterious ASR expansion was noted due to the beneficial pozzolanic reaction between glass powders and cement. Acceptable strength was achieved at the same time. They suggested that the glass powder can be incorporated with a dosage rate of 20–30% to replace cement without adverse effects. This indicates a great potential of utilization of waste glass as supplemental cementitious material [13] in concrete. If the particle size of glass powder is sufficiently small, the amorphous silica content dissolves quickly and reacts with the free portlandite, causing the pozzolanic reaction [14] and making the alkali content unavailable to react with the silica content in the aggregates. This shows that the size-effect of glass powders is crucial for the alleviation of ASR expansion hence the improvement of the concrete durability. The pozzolanic characteristics of glass are first appreciable at particle size below 300 μm [13]. Through compressive strength tests, Meyer et al. [15] proposed that the glass may become pozzolanic when the size is below 45 μm. Shi et al. [16] studied glass powders with different sizes and found that the pozzolanic reactivity is inversely proportional to the particle size. Particularly, below 100 μm, glass can be more reactive than fly ash at low cement replacement levels. They also reported that the incorporation of glass powders reduced the mortar expansion, although it was not as effective as coat fly ash.

A large amount of studies on the size-effect of recycled glass took a performance-based approach to assess the effect of different sizes on the mechanical and transport properties, and simultaneously identify the ASR risks or damages. However, in order to gain a fundamental understanding of the size-effect on the ASR damage mechanism, a microstructure-based approach, which utilizes characterization techniques, such as multiscale USANS/SANS [17], SEM [18], X-ray micro-CT [19], etc., is more suitable for this purpose. The USANS/SANS technique can achieve the resolution ranging from 1 to 100 nm. Thus, it was previously applied to investigating the microstructure of the cementitious materials [20,21]. Through combining SANS and other techniques, Gaboriaud et al. [22] found a strong dependence of the structural features of ASR gel and the concentration of the calcium ions. Instead of capturing an image of a particular substance, the neutron scattering methods only provide statistically averaged structural information. Therefore, complementary image techniques would be helpful as a comprehensive examination of the microstructure. The reacted gel microstructure, including the particle size and shape, can be directly examined with SEM techniques [18]. Fernandez-Jimenez et al. [23] used SEM to examine the volume expansion of the ASR gel in alkali-activated slag and ordinary Portland cement mortars, respectively. The results demonstrated the gel expansion in the alkali-activated slag mortar was insignificant compared to the expansion in the ordinary Portland cement mortar. Mitchell et al. [24] investigated the reaction product of lithium hydroxide with opals. The surface morphology of the reaction product was analyzed with SEM. A protective layer was detected on the product surface which was able to reduce the ASR damage. Fernandes et al. [25] analyzed the field gel obtained from a 50-year-old concrete dam with SEM and energy dispersive spectrometer (EDS) test. The ASR product was identified with the elemental and morphology analysis. Particularly, after the field gel was put into the desiccator for three months, the transformation from amorphous structure to needle and tablet structure was observed. Fernandes [26] analyzed the ASR gel from the field samples with the SEM/EDS test. Both texture and composition were found different depending on the location of the gel. Particularly, the calcium content varied significantly among these field gels. Peterson et al. [27] investigated the morphology of the field gel obtained from a concrete structure constructed in 1890s. Both the amorphous and crystalline structures were found in this field gel. Rajabipour et al. [28] used SEM to study the size-effect of glass aggregates and found that the ASR occurred at the pre-existing microcracks within the glass aggregates, rather than the glass-paste interface. The dynamic X-ray CT can monitor the gel morphology change or volume expansion process, which has been used to study the microstructure development of cement pastes [29,30]. Marinoni et al. [19] studied the ASR gel obtained from the concrete samples containing chert aggregates by X-ray CT. The microcracks induced by ASR were detected and analyzed.

To study the size-effect of recycled glass using the above-mentioned analytical models or other numerical models, the characterization information about the microstructure and dynamic change of gel particles is mandatory. This can be obtained through the microstructure-based approach. Thus, the objective of this study is to investigate the size-effect on the microstructure and morphology of synthetic alkali-glass particle and alkali-glass powder reacted gels. The previous studies were all static examinations of the microstructures of ASR gels at a particular reaction stage. In this study, through the applications of the in-situ dynamic X-ray CT on the ASR gel, the development of alkali-glass particle reacted gels and the crack propagation were captured with a fast pace and high resolution. The gathered microstructure information can serve as input for the analytical or numerical models. In fact, the results herein have been used in a numerical simulation of ASR damage, by combining with facture mechanics. This has been reported in a separate paper [31]. In this study, the preparation of gel samples was described in detail, which was achieved by using the controlled alkali-glass particle or alkali-glass powder reactions. The reacted gel particle size and power-law constant were characterized with the USANS/SANS. After that, the USANS/SANS characterization results were validated with the SEM images. The dynamic X-ray micro-CT techniques were applied to investigate the gel development at different reaction stages (up to 64 h) through examining the particle size distribution of reaction products.

2. Alkali-Glass gel Characterization with USANS/SANS

2.1. Alkali-Glass Particle gel Sample Preparation for USANS/SANS Characterization

The recycled glass particle used in this study was obtained from Vitro Minerals. The particle size ranges from 420 to 840 μm. The main contents of glass particle are listed in Table 1 [32]. The remaining contents include Fe_2O_3, MgO, and SO_3 with the mass percentages all below 1%. The alkali-glass particle reacted gel sample (Type 1) was prepared with a Na:Si molar ratio of 1:1. The glass particles were first mixed with 10 mol/L NaOH solution in a Teflon beaker before a heating process for three minutes. Due to the highly basic condition of the solution, the following synthesis procedure was carried out in a fume hood. Therefore, it is expected that the samples might undergo carbonation to some extent. The solution was heated and stirred with a digital hotplate up to 80 °C for one hour to accelerate the reaction and evaporate the excessive water. Finally, the condensed gel was poured into a specially designed mold (Fig. 1(a)) to obtain the thin disk samples

Table 1
Chemical compositions of glass particles and glass powder (%).

Materials	Size	CaO	SiO_2	Al_2O_3	$K_2O + Na_2O$
Glass particle	420–840 μm	11 ± 2	75 ± 5	3 ± 2	13 ± 3
Glass powder	0.5–45 μm	23 ± 3	53 ± 3	17 ± 3	11 ± 3