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Enhancement of thermal neutron shielding of cement mortar by using borosilicate glass powder

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ABSTRACT

Concrete has been used as a traditional biological shielding material. High hydrogen content in concrete also effectively attenuates high-energy fast neutrons. However, concrete does not have strong protection against thermal neutrons because of the lack of boron compound. In this research, boron was added in the form of borosilicate glass powder to increase the neutron shielding property of cement mortar. Borosilicate glass powder was chosen in order to have beneficial pozzolanic activity and to avoid deleterious expansion caused by an alkali–silica reaction. According to the experimental results, borosilicate glass powder with an average particle size of 13 µm showed pozzolanic activity. The replacement of borosilicate glass powder with cement caused a slight increase in the 28-day compressive strength. However, the incorporation of borosilicate glass powder resulted in higher thermal neutron shielding capability. Thus, borosilicate glass powder can be used as a good mineral additive for various radiation shielding purposes.

1. Introduction

Concrete has been considered as one of the promising biological shielding materials. Concrete is effective for shielding gamma radiation because of its dense, complex, and heterogeneous microstructure, and the use of high-density aggregates can effectively increase gamma ray shielding capability (Mehta and Monteiro, 2006). In addition, Davis (1972) reported that high hydrogen content associated with free water (capillary pore solution), physically adsorbed water within calcium silicate hydrate layers, and chemically bound water in the structure of calcium silicate hydrate in concrete effectively attenuate high-energy fast neutrons. Thus, concrete can be a very good neutron moderator if there is a method to protect against thermal neutrons because concrete does not contain any boron compounds that have high neutron capture cross section in its structure (Rinard, 1991). Therefore, it is necessary to include some amount of boron compounds in concrete to increase its efficiency as a biological shielding material.

One of the easiest ways to add boron compound in concrete is to use boric acid and borax. These materials are not known to be compatible with portland cement whose major reaction is based on silicate polymerization that occurs at high pH environment. These materials cause set delay and strength loss of concrete (Davraz, 2015; Olgun et al., 2007; Targan et al., 2002; Volkman and Bussolini, 1992) and thus making it meaningless to add them for thermal neutron protection of concrete. Boron carbide can be used as an alternative material (Kharita et al., 2011), but it is very expensive; moreover, its particle size distribution either for cement replacement or for aggregate replacement is difficult to control because of its high hardness.

The objective of this research is to use borosilicate glass for thermal neutron shielding. Borosilicate glass is a material with very low thermal expansion property. Typical examples are Schott by Duran or Pyrex by Corning. It contains up to 13 wt% of boron trioxide (B_2O_3), and thus can show excellent thermal neutron shielding performance when it is added to the cement-based material. Hence, borosilicate glass has been used as a medium for immobilizing various types of radioactive wastes (Alton et al., 2002; McCloy et al., 2012; Mishra et al., 2008; Tomar et al., 2005). However, borosilicate glass is an amorphous silica and is known to cause an alkali–silica reaction that causes deleterious expansion and cracking in concrete when it is added in the form of fine aggregate. Thus, the size of the borosilicate glass needs to be controlled to avoid such problems.

Shao et al. (2000) reported that incorporation of a soda-lime glass with a particle size of less than 150 µm did not show the alkali–silica reaction. They also showed that smaller-sized waste lime glass (75 and 38 µm) had less expansion than plain cement mortar. Zhu and Byars (2004) reported that there is a critical particle size for colored waste glass to be used as aggregate; pessimum size for amber and green color glass was 0.6 and 1.18 mm for flint color glass. Shayan and Xu (2004)

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found that glass powder smaller than 10 μm can be used to replace reactive pozzolanic materials such as silica fume or fly ash. Shao et al. (2000) also noted that waste glass, finer than 38 μm might have presented pozzolanic activity, but their argument was made without strong evidence. Although the structure of soda-lime glass or colored glass is not the same as that of borosilicate glass, the results from these works can at least provide a guideline to process borosilicate glass for neutron shielding application.

In this work, borosilicate glass powder was used to increase thermal neutron shielding property of portland cement mortar. The size of the borosilicate glass powder was controlled to be much lower than the critical particle size of 38 μm as reported by Shao et al. (2000). Pozzolanic activity of such borosilicate glass powder was investigated. Finally, the thermal neutron shielding property of cement mortar containing borosilicate glass powder was examined. Successful achievements of this research may lead to provide a construction material with desired neutron shielding property that can extend the duration of operation for nuclear power plant or to use it as a shielding material for radioactive waste container.

2. Experimental procedures

2.1. Preparation of borosilicate glass powder

Borosilicate glass, SCHOTT by DURAN®, was chosen for this research. The chemical compositions of SCHOTT borosilicate glass are shown in Table 1. Borosilicate glass was first crushed using a steel hammer. Broken pieces of crushed glass were then ground using a vibrating disk mill (RETZSCH, Germany) for 150 s for facilitating ball mill grinding. Resulting powders were placed in a stainless steel mill with a 10-mm-size stainless steel ball (KOPECO, Korea), and ground for 7 days with constant speed of 240 rpm to achieve particle size of borosilicate glass powder much less than the critical size suggested by Shao et al. (2000). Magnetic separation was applied on the powder by using 13,000 gauss (stainless steel SUS-304) magnet to remove metallic impurity that may be included in the ground borosilicate powder.

Particle size distribution of borosilicate glass powder (Fig. 1) was measured using LS 13320 laser diffraction particle size analyzer (BECKMAN COULTER, USA) and, and the particles were found to have median size of 13 μm and mean size of 9 μm.

2.2. Pozzolanic activity

Cement paste was prepared for the analysis of pozzolanic activity of borosilicate glass powder. Type I portland cement was used, and its chemical composition is shown in Table 2.

Pozzolanic activity is generally understood as a reaction between amorphous silica and calcium hydroxide to form insoluble calcium silicate hydrate or calcium aluminosilicate hydrate. Thus, with pozzolanic reaction, compressive strength increases and pore connectivity decreases. The reactions can be summarized in Eqs. (1)–(3).

$$x\mathrm{Ca(OH)_2} + y[\mathrm{SiO_2},\ \mathrm{Al_2O_3}] \rightarrow l\mathrm{CaO}\cdot m\mathrm{SiO_2}\cdot n\mathrm{H_2O} \quad (1)$$

$$l\mathrm{CaO}\cdot m\mathrm{Al_2O_3}\cdot n\mathrm{H_2O} \quad (2)$$

$$l\mathrm{CaO}\cdot m\mathrm{SiO_2}\cdot n\mathrm{Al_2O_3}\cdot o\mathrm{H_2O} \quad (3)$$

Table 1
Chemical compositions of Pyrex glass.

Compositions	%
SiO_2	81
B_2O_3	13
$Na_2O + K_2O$	4
Al_2O_3	2

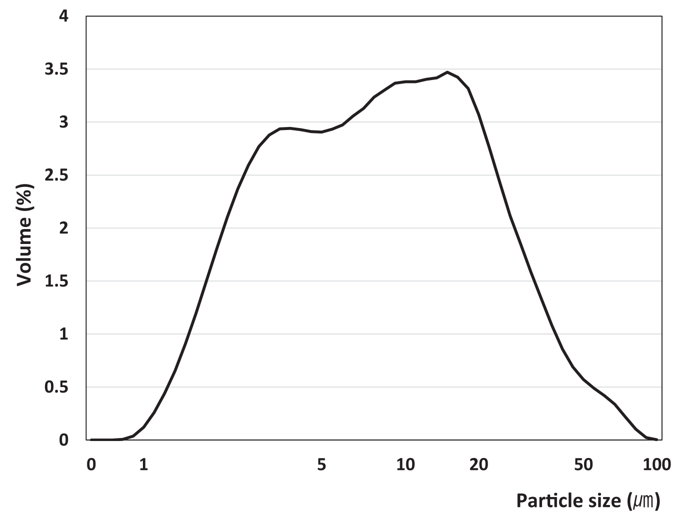

Fig. 1. Particle size distribution of borosilicate glass powder (mean size: 13 μm, median size: 9 μm).

Table 2
Chemical compositions of type I portland cement.

Chemical analysis	Content (wt%)
CaO	53.42
SiO_2	19.48
Al_2O_3	4.69
SO_3	4.08
MgO	3.11
Fe_2O_3	3.04
K_2O	1.32
TiO_2	0.38
P_2O_5	0.20
MnO	0.13
ZnO	0.11

Pozzolanic activity of a material can be evaluated by the amount of calcium hydroxide consumed in the pozzolanic reaction. To use this approach, it is necessary to strictly control mixing and curing environment because some amount of calcium hydroxide is consumed by carbonation. In this work, experiments were conducted in a N_2-filled glove box. Degassed nano-pure deionized (18.30 MΩ·cm) water was also used. Cement paste samples for the analysis of pozzolanic activity were prepared using degassed water in N_2-filled environment. Plain cement paste and cement pastes with 10%, 20%, and 30% replacement of cement with borosilicate glass powder were prepared. The water to binder ratio (w/b) was set at 0.47. Cement paste samples were poured into the 10 mm by 20 mm cylindrical mold and stored in the N_2-filled glove box for a day. The specimens were then demolded and placed in a lime-saturated solution (also in N_2-filled glove box) for additional 27 days to meet the requirement of a total of 28 days of hydration.

XRD analyses were performed using Ultima IV X-ray diffractometer (Rigaku, Japan) equipped with Cu-Kα radiation to identify the presence of calcium hydroxide in the 28-day old cement paste samples. TG/DTA analyses of 28-day old cement paste specimens were also performed using Bruker TG-DTA 2020 model (Bruker, Germany) to quantitatively measure the amount of calcium hydroxide present in cement paste samples. The pozzolanic activity of borosilicate glass powder was evaluated on the basis of the amount of calcium hydroxide present in each specimen.

2.3. Compressive strength

Standard sand, from Jumunjin, Republic of Korea, was used as a source of fine aggregate for the preparation of mortar specimen. Mortar