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SUPPLEMENTARY MATERIAL TO

Influence of fly ash fineness on cesium immobilization in geopolymers

GORDANA G. STANOJEVIĆ[1]*, MIROSLAV M. KOMLJENOVIĆ[1], JANA LJ. PETROVIĆ[2] and RADA D. PETROVIĆ[3]

[1]*Institute for Multidisciplinary Research, National Institute of the Republic of Serbia, University of Belgrade, Kneza Višeslava 1, 11030 Belgrade, Serbia,* [2]*Innovation Center of the Faculty of Technology and Metallurgy, Ltd, 11060 Belgrade, Serbia and* [3]*Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4, 11060, Belgrade, Serbia*

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EXPERIMENTAL PROCEDURE

Synthesis of geopolymers pastes and mortars

The geopolymer pastes were prepared by mixing FA-3, alkaline activator, and water. For Cs containing geopolymers, CsCl was first dissolved in water and then mixed with FA-3 and the alkaline activator. The Cs content was 2.0 % and 5.0 % relative to the mass of FA-3. The water/binder ratio was 0.39. In this ratio, water refers to the total amount of water in the system, including water from the activator and any additional water added to achieve comparable paste consistency, while the binder represents the sum of the FA mass and the solid part of the activator. The flow of the pastes was measured on a flow table and found to be 195 ± 10 mm, according to the standard method described in,[1] to ensure comparable workability of all samples. The concentration of Na_2O for all samples was 10 % relative to the mass of FA-3.[2]

The pastes were mixed for about 3 min, then poured into plastic cylindrical molds (62.5 x 15 mm), after which air bubbles were removed on a vibrating table. The prepared pastes were cured, covered with plastic bags at 95 °C for 24 h, and then at room temperature (20 ± 2 °C) until the testing.[2]

The geopolymer mortar was prepared by mixing FA-3, alkaline activator, water, and standard sand, following the procedure of EN 196-1[3] as closely as possible. The FA-3: sand ratio was 1:3. In the case of geopolymers with added Cs, the mortars were prepared by dissolving CsCl in water, then mixing with FA-3, alkaline activator, and finally sand. The Cs content and the concentration of Na_2O were the same as in the corresponding pastes.

The water:binder ratio was 0.43 for the G-FA-3-0Cs, 0.44 for the G-FA-3-2Cs, and 0.42 for the G-FA-3-5Cs sample. The water/binder ratio was determined as for the pastes. The mortars were homogenized in an automatic mixer for approximately 3 min, then poured into molds containing three mortar prisms of standard dimensions (40 x 40 x 160 mm) on a vibrating table, and cured under the same conditions as the pastes.

* Corresponding author. E-mail: gordana@imsi.bg.ac.rs

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Leaching process

Deionized water was used as the leaching medium, with an electrical conductivity of less than 5 μS/cm on 25 °C. The ratio of the volume of the leaching medium to the total geometric surface area of the samples was kept constant at 10 ± 0.2 cm.

All paste samples were completely immersed in deionized water. The leaching test was conducted for 5 days at 23 ± 2 °C, with the leaching medium being fully replaced after cumulative leaching times of 2, 7, 24, 48, 72, 96, and 120 h.

At the end of each interval, the solution was filtered through a 0.45 μm membrane filter under vacuum. The pH of the filtrate was measured with a pH meter (HANNA Instruments HI 991001). Before measuring the concentrations of leached Cs, Al, and Si, the pH of the filtrate was lowered to below 2 by adding concentrated HNO_3. The concentrations of leached Cs, Al, and Si were measured using an Inductively Coupled Plasma-Optical Emission (ICP-OES) instrument (Spectro-Genesis EOP II 991001).

According to the ANSI/ANS-16.1-2003 standard,[4] the individual leaching rates of Cs were calculated according to equation (S1):

$$LR = \frac{a_n/A_o}{(\Delta t)_n} \tag{S1}$$

where, a_n is the amount of cesium released from the sample during leaching interval n, g; A_0 is the total amount of cesium in the sample at the beginning of the first leaching interval, g, and

$$(\Delta t)_n = t_n - t_{n-1} \tag{S2}$$

is the duration of the nth leaching interval, h.

The diffusion coefficient (D) was calculated using the mass transport equation (Fick's second law):

$$D = \pi \left[\frac{a_n/A_o}{(\Delta t)_n}\right]^2 \left[\frac{V}{S}\right]^2 T \tag{S3}$$

where D is the diffusion coefficient, cm^2/s; a_n is the amount of Cs released from the sample during the leaching interval *n,* g; A_0 is the total amount of Cs in the sample at the beginning of the first leaching interval, g; $(\Delta t)_n$ is the duration of the n-th leaching interval, s; V is the sample volume, cm^3; T is the leaching time, s, represented by the geometric mean time of the nth leaching interval, calculated as follows

$$T = \left[1/2\left({t_n}^{1/2} + {t_{n-1}}^{1/2}\right)\right]^2 \tag{S4}$$

The mean leachability index (L_{mean}) of Cs from G-FA-2%Cs and G-FA-5%Cs was calculated according to the following equation [S5]:

$$L_{mean} = 1/n \sum_{n=1}^{n_{max}\Sigma} [log(\beta/D)]_{n_{max}} \tag{S5}$$

where, n_{max} is the number of D values used for the calculation (7), β is defined constant (1.0 cm^{2}s^{-1}), and D is the diffusion coefficient of Cs calculated from equation (S3), for each step n.

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RESULTS AND DISCUSSION

XRD

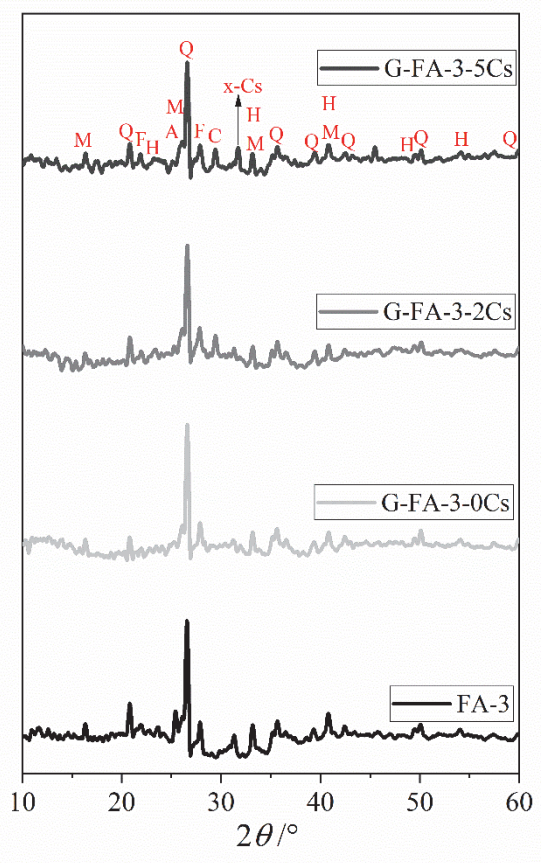

Fig. S-1. XRD patterns of FA-3 and G-FA-3 samples (Legend: M-mullite, Q-qurtz, H-hematite, A-anhydrite, F-feldspar, C-calcite, X-Cs-$Cs_2Al(OH)(SiO_4)_2$)

FESEM

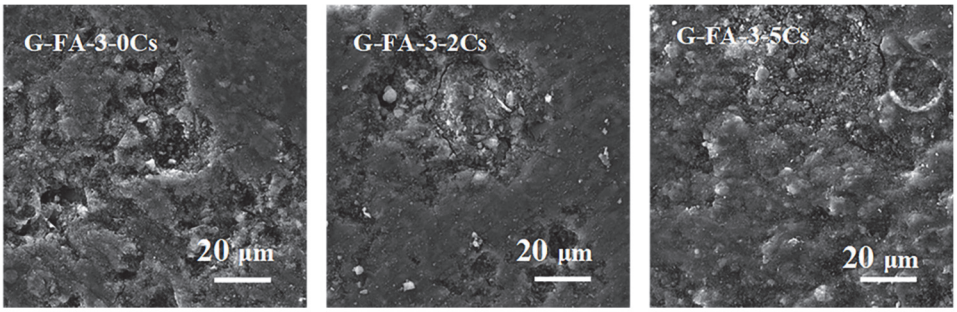

Fig. S-2. SEM images of G-FA-3

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Leaching of Cs from G-FA-4

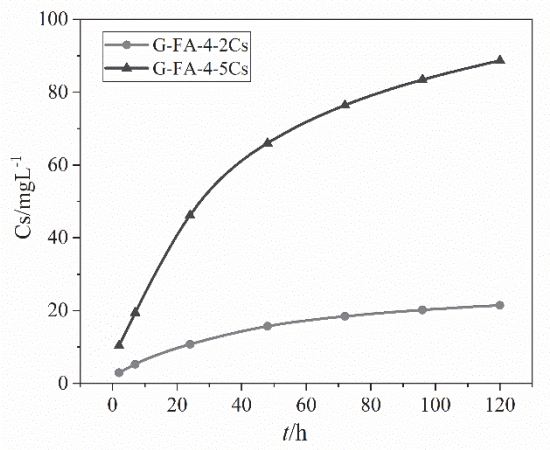

Fig.S-3. Cumulative concentration of leached Cs from G-FA-4

Leaching of Si and Al from G-FA-4

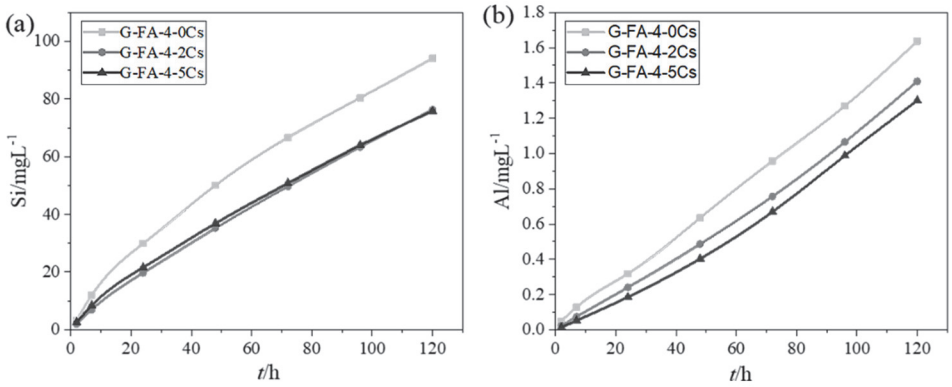

Fig.S-4. Cumulative concentration of leached (a) Si and (b) Al from G-FA-4

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