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Enhancement of sorption capacity to Sr and Cs of a cement composite by addition of brick powder

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Abstract. Concrete is a suitable material for secondary casings in multi-barrier systems used in radioactive waste repositories, because it is durable and radiation- and thermal resistant and it has good mechanical properties. The casings should also limit possible leaching of radionuclides in case of failure of inner barriers. An efficient way to tackle this problem is the addition of a sorption material in the concrete mixture. Here mechanical and thermal properties of concrete containing different amounts of brick powder are studied in relation to the concrete mixture porosity. It is found out that, as expected, the bulk density and compressive and bending strengths decreased with an increasing amount of the brick powder. In addition, the sorption of cesium and strontium chlorides into the mixtures was analyzed under laboratory conditions, and their sorption isotherms and distribution coefficients were calculated. The results indicate that brick has higher sorption capacity for Sr than for Cs. Relatively low distribution coefficients indicate, however, that the studied brick is not a very suitable additive material for sorption of Sr and Cs elements.

1. Introduction

The present solutions of radioactive waste disposal are based on multi-barrier systems of protection to maximize the preservation of the environment surrounding waste repositories, including the safety of living organisms [1–3]. The barrier systems are composed of natural, geological barrier (the surrounding rock) and engineered barriers that include [4]

- durable metallic or concrete canisters (containing sealed hazardous waste);
- clay or cementitious backfills (void filling, limiting water infiltration, sorption and precipitation of radionuclides);
- concrete, reinforced concrete, or clay containment barriers (additional protection of waste and physical stability);
- gravel, sand, ceramic, or concrete drains (control of leachate).

Concrete and cementitious composites are conveniently used in radioactive waste repositories due to its good mechanical properties, durability, low corrosion, and low cost. Concrete casings are usually



reinforced to have sufficient mechanical parameters in order to withstand the manipulation on site and ground pressure after the final emplacement in the repository [5, 6]. In addition to proper mechanical properties, there are requirements on the radiation stability of concrete, its thermal stability and resistance to aggressive water and, perhaps, organisms. Concrete barriers also limit the leaching of radionuclides into the environment in case of an accidental leach from the inner barriers. Natural and synthetic zeolites, presenting pozzolanic activity and having good sorption properties [7, 8], can be used as additives to concrete, taking part in the cement hydration and/or acting as a filler [9, 10].

In this paper we study whether an addition of a brick powder in varying amounts can enhance the capacity of concrete to sorb strontium and cesium. We also investigate changes in mechanical and thermal properties of the concrete in relation to the amount of the brick powder.

2. Materials and methods

2.1. Materials

The principal binder component was Portland cement CEM I 42.5 R (Mokrá Ltd., Czech Republic) with the specific surface area of $408 \text{ m}^2\text{kg}^{-1}$ and bulk density 3110 kg m^{-3} . It was the only binder component for the reference mixture MCR. In four other mixtures, brick powder (Heluz) was added in varying amounts. Namely, 20% of the cement mass (80 kg in 1 m^3) was first substituted by brick powder. Then the addition of brick powder was dosed increasingly by 40 kg m^{-3} , while the cement dosage was kept constant. Brick powder served as an inert mineral additive and took part in the hydration of the binder system. This material has the pozzolanic activity of $840 \text{ mg}(\text{Ca}(\text{OH})_2) \text{ g}^{-1}$, its specific surface area is $402 \text{ m}^2\text{kg}^{-1}$, and its bulk density was 2540 kg m^{-3} . The mixture contained also a fine filler – silicate flour ST6 (Sklopísek Střeleč Ltd., Czech Republic) with the specific surface of $0.4 \text{ m}^2\text{g}^{-1}$. Superplasticizer Stachesil 2000 (Stachema CZ Ltd., Czech Republic) based on polycarboxylate was added to achieve good rheology of the fresh mixture, and the amount of water was dosed in order to meet the prescribed slump of the fresh mixture. Table 1 gives the chemical composition of the input materials. The particle size distributions of fine components are shown in Fig. 1. The composition of the studied materials is presented in Table 2.

Table 1. Chemical composition of the input materials (wt. %).

	Portland cement 42.5 R	Brick powder	Silicate flour
SiO₂	18.7	51.3	99.7
Al₂O₃	4.5	20.0	0.2
Fe₂O₃	3.4	6.0	–
CaO	65.9	11.5	–
MgO	1.3	4.5	–
K₂O	0.8	3.2	–
Na₂O	0.2	1.3	–
TiO₂	0.3	0.8	–
SO₃	4.3	1.0	–
P₂O₅	0.1	–	–

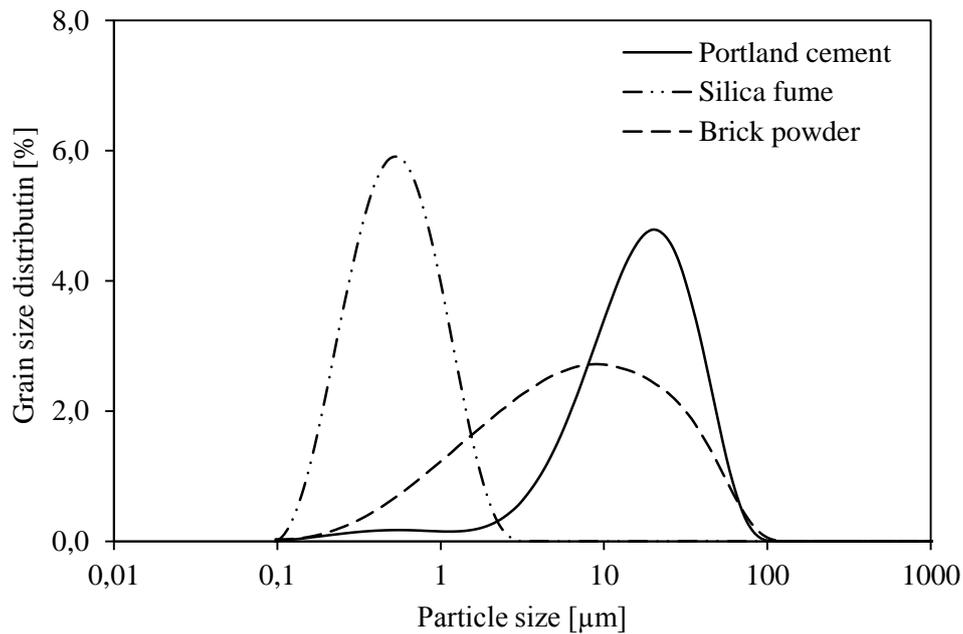


Fig. 1. Grain-size distribution curves of used components.

Table 2. Composition of studied materials (kg m^{-3}).

	RCR	RC1	RC2	RC3	RC4
Portland cement CEM 42.5 R	400	320	320	320	320.00
Brick powder	(0 %)	80 (20%)	120 (30%)	160 (40%)	200 (50%)
Silicate flour ST6	120	120	80	40	-
Superplasticizer – Stachement 2000	6.9	6.9	6.9	6.9	6.9
Water	195	220	220	220	220

2.2. Experimental methods

2.2.1. Basic physical properties

Bulk densities, ρ [kg m^{-3}], were determined from the volume and mass measured for the individual samples.

2.2.2. Mechanical properties

The compressive strength [MPa] and bending strength [MPa] are the main mechanical parameters and were determined according to ČSN EN 12390-3 [11] and ČSN EN 12390-5 [12], respectively. A device EU 40 was used. The strength was determined for 28-days cured samples.

2.2.3. Sorption isotherms

Sorption isotherms were determined by batch experiments. The sorbate solutions of varying concentrations were prepared from CsCl and $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ (p.a.). Namely, 100 mg of pulverized concrete and 100 ml of solution of a proper concentration were agitated for 72 hours and 100 rpm. These conditions proved to be sufficient for equilibration of Cs and Sr between sorbent and solution. The sorbed amount, q (in mg g^{-1} of the sorbent), was determined as

$$q = \frac{V(C_0 - C_e)}{m}, \quad (1)$$

where C_0 is the initial concentration in mg l^{-1} , C_e is the equilibrium concentration, V is the volume of the solution in liters, and m is the mass of the sample (sorbent) in grams. The concentration was determined by Atomic Absorption Spectrometer Agilent AA 280 FS with a flame atomization technique (Flame: Acetylene – air). The distribution coefficient, K_d (in ml g^{-1}), was determined as the ratio q/C_e .

3. Results and discussion

3.1. Basic physical properties

The measured values of bulk densities are given in Table 3. The bulk density decreased with the increasing amount of brick powder in the mixture, which is a result of increasing open porosity. This is caused by a higher porosity of the input brick powder. The decrease of bulk density is by about 10% (RC1) to 35% (RC4).

Table 3. Bulk density of studied materials.

	RCR	RC1	RC2	RC3	RC4
Bulk density [kgm^{-3}]	2161	1984	1998	1997	1612

3.2. Mechanical properties

Results concerning mechanical properties are given in Fig 2. There is a clear decreasing trend for both compressive and bending strengths. The drop in compressive strength for RC1 compared to the reference concrete is 17% and in the case of RC4 the residual strength is at about 35%. In the case of the bending strength, the decrease for RC1 is lower than in the case of the compressive strength – by 4%. However, for RC4 the bending strength showed less than half the reference value.

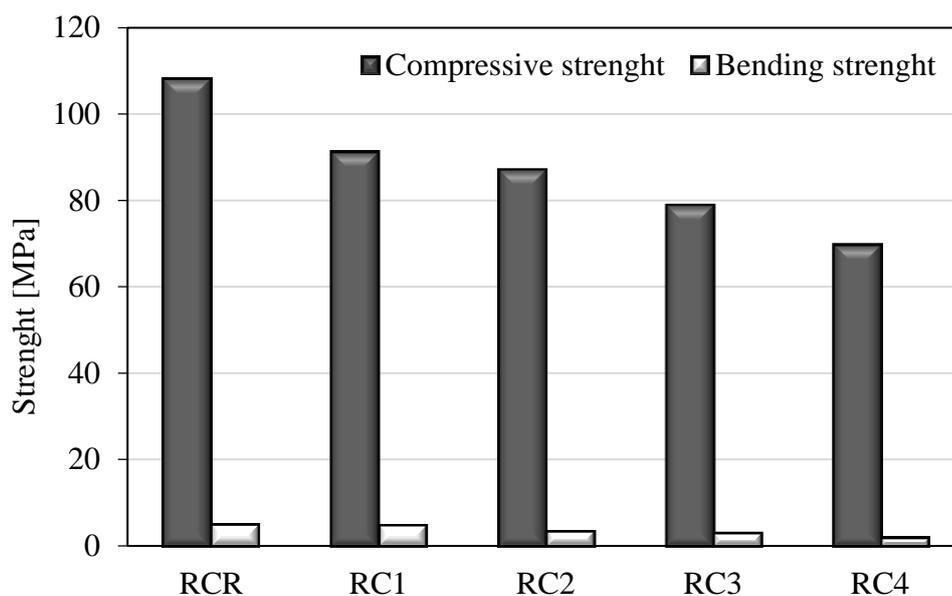


Fig. 2. Mechanical properties of the studied materials.

3.3. Sorption isotherms

The sorption isotherms obtained for the studied samples are shown in Figs. 3 and 4. These results

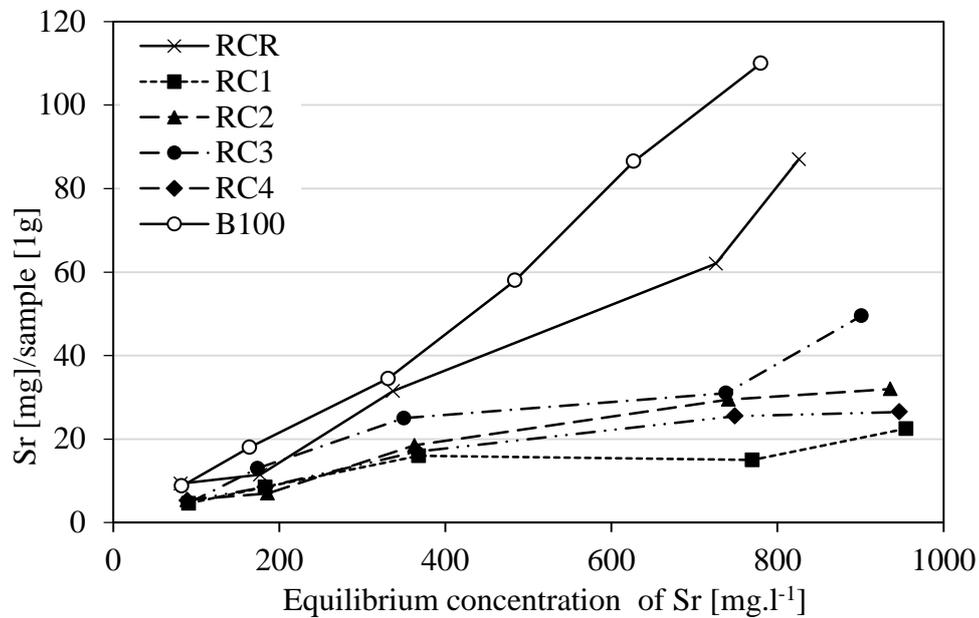


Fig. 3. Relationship between equilibrium concentration of Sr and its amount sorbed into the tested samples.

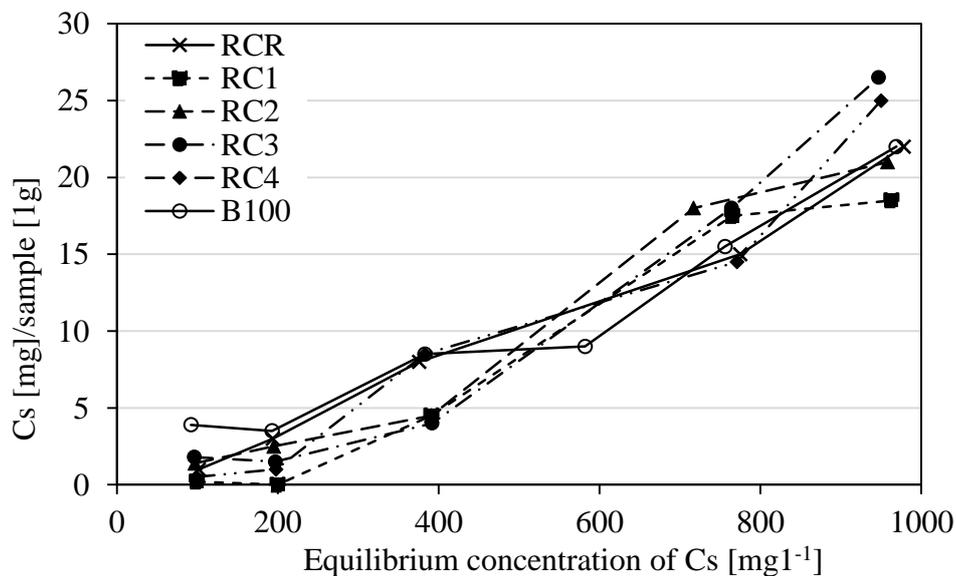


Fig. 4. Relationship between equilibrium concentration of Cs and its amount sorbed into the tested samples.

indicate that the samples containing brick adsorbed strontium to a larger extent than cesium. For samples containing the brick powder, the calculated values of the distribution coefficient K_d (see

Tables 4 and 5) are on an average about twice higher for Sr (from 19.48 mg l⁻¹ to 141.03 mg l⁻¹) than for Cs (from 0 mg l⁻¹ to 57.88 mg l⁻¹). For the reference samples the range of values of K_d is 50.96 – 117.18 mg l⁻¹ for Sr and 0 – 31.91 mg l⁻¹ for Cs, which is a more profound difference than for the samples with the brick powder. Thus, in general, Sr is sorbed in the samples in greater amounts than Cs.

Table 4. Distribution coefficient for all tested concentrations of Sr and all samples.

Sample	RC1	RC2	RC3	RC4	B100	RCR
Initial concentration [mg Sr.l ⁻¹]	K_d [mg Sr.l ⁻¹]					
100	50.66	61.17	51.88	58.66	106.06	117.18
200	46.45	37.63	74.71	46.45	109.76	64.97
400	43.48	50.96	71.43	46.45	104.23	93.47
800	19.48	39.81	42.01	34.05	137.96	50.96
1 000	23.56	34.19	54.94	27.98	141.03	105.33

Table 5. Distribution coefficient for all tested concentrations of Cs and all samples.

Sample	RC1	RC2	RC3	RC4	B100	RCR
Initial concentration [mg Cs.l ⁻¹]	K_d [mg Cs.l ⁻¹]					
100	2.01	14.40	18.67	5.05	42.30	0.00
200	0.00	12.82	7.61	5.05	18.13	15.46
400	11.51	11.51	10.20	22.19	22.19	31.91
800	22.88	57.88	23.56	18.81	29.10	16.13
1 000	19.21	21.92	27.98	26.32	16.00	11.25

4. Conclusions

The application of brick powder as a supplementary sorption material in a Portland cement paste was tested in this paper. Four mixes with 20 %, 30 %, 40 %, and 50% of the cement mass replaced by a brick powder were designed. The sorption capacity of the mixes were quite higher for Sr than for Cs, and so were the distribution coefficients. In comparison with the reference samples (with no brick powder), it turns out, however, that the addition of the brick powder did not significantly increased the sorption of neither Sr not Cs. In fact, a decrease in sorption was observed when for some mixes. Therefore, it should be concluded that the studied brick powder is not very suitable for the sorption of Sr and Cs elements in concrete.

Acknowledgment

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