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Utilization of waste ceramics and roof tiles for radionuclide sorption



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ABSTRACT

The possible utilization of waste ceramic (CT) and roof tiles (RT), as sorbents for liquid radioactive waste (LRW) treatment, was investigated. Following the European directives on waste and politics of saving natural raw materials, it is reasonable and desirable to explore potential applicability of such construction wastes. These materials are low-cost and locally available in high quantities, yet, their sorption characteristics were not evaluated to this point. In the present study, detailed physicochemical characterization of waste CT and RT included determination of mineral composition, surface functional groups, radioactivity, as well as the stability in aqueous media. The batch sorption study of Sr²⁺, Co²⁺ and Ni²⁺ ions from single- and multi-component solutions was performed, as their radioactive isotopes are common constituents in LRW. Sorption equilibrium was best described by Freundlich isotherm model, regardless of the sorbent and the sorbate type. Sorption capacities of CT, defined in single element solutions, increased in the order 0.035 mmol Sr/g < 0.12 mmol Ni/g < 0.17 mmol Co/g, while the affinity of RT was generally lower (0.030 mmol Sr/g < 0.065 mmol Co/g < 0.10 mmol Ni/g). The study of competitive cation sorption was performed following a simplex centroid experimental design matrix. The equations for the prediction of metal sorption capacities from multi-component solution were derived. Utilization of CT and RT might be an efficient way for waste water purification, with simultaneous reduction in construction waste amount on municipal landfills.

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1. Introduction

Civil engineering generates construction and demolition debris, representing nearly 75% of waste worldwide (Mymrin et al., 2015; Zimbili et al., 2014). Although this waste falls into the category of non-hazardous solid waste, it consumes the capacities of municipal landfills. Large amounts of diverse materials such as concrete, ceramic (bricks and roof tiles), asphalt, wood, steel and other metal products, plastic, etc., are being disposed. The problem of accumulation of non-recyclable construction waste has exhibited global proportions, and since it continues to increase rapidly, it is necessary to develop innovative and efficient strategies for its reduction and valorization (Mymrin et al., 2015). Ceramic materials contribute approximately 54% to construc-

tion and demolition wastes, and associated recycling and reuse rates are not available in the EU due to an absence of a systematic European reporting system (European Commission, 2011). One of potentially prosperous fields in ceramic waste utilization is exploitation of its sorption properties. During the last decade, interest in waste materials and by-products utilization as sorbents for purifying contaminated water was markedly increased (Ahmaruzzaman, 2011).

Thanks to their chemical composition and compatibility with solid matrixes (such as cement) used for the disposal of radioactive waste, ceramic materials are prospective sorbents for immobilization of radionuclides present in aqueous solutions. Radioactive waste is generated due to the activity of nuclear power plants, nuclear weapon testing, industrial applications, research institutions, medical uses,

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etc. (IAEA, 1998), and consequently, liquid radioactive waste (LRW) streams of different origin exhibit variable chemical composition. In LRW streams associated with the operation of nuclear reactors, isotopes of ^{60}Co , ^{63}Ni , ^{90}Sr are recognized as abundant and important even many years after the reactor shutdown (IAEA, 1998). ^{60}Co and ^{63}Ni originate from reactor made ferritic materials, while ^{90}Sr is a fission product. The sorption of such radionuclides using different low-cost materials was already investigated (Milenković et al., 2014; Galamboš et al., 2011, 2012; Jiménez-Reyes and Solache-Ríos, 2016; Šljivić-Ivanović et al., 2015; Ofomaja et al., 2015), however, there are no available literature data on the utilization of waste tiles.

The main objective of this study was to evaluate waste ceramic and roof tiles as sorbent materials for the treatment of LRW containing Co^{2+} , Sr^{2+} and Ni^{2+} . For the first time, physicochemical and sorption properties of waste tiles were analyzed to better comprehend the potential hazards and prospects for cation immobilization. The sorption kinetics and the capacities toward Co^{2+} , Sr^{2+} and Ni^{2+} ions were determined using single-component solutions, whereas competitive cation removal was explored using three-component solutions with different metal proportions, according to simplex centroid experimental design matrix.

2. Experimental part

2.1. Collection, preparation and characterization of waste tiles

Samples of roof tiles (RT) and ceramic tiles (CT) were collected from different locations in Belgrade (Serbia). About 2–3 kg of the tile pieces were taken from 5 different building ruins. The tiles were crushed, milled and then sieved. After homogenization, obtained powders were spread over the area of around 1 m^2 , and 50 g were sampled from five randomly found points. Fraction with particle size between 0.3 and 0.6 mm was collected for further experiments, rinsed with deionized water and dried at 100°C .

CT and RT powders were characterized in terms of mineral composition, surface functional groups, content of radioactive elements, pH, and stability in aqueous media. Mineral phases were identified by X-ray diffraction analysis (XRD) using Ultima IV Rigaku diffractometer, equipped with $\text{Cu K}\alpha_{1,2}$ radiation, at generator voltage of 40.0 kV and current of 40.0 mA. The range of $4\text{--}65^\circ 2\theta$ was used for analysis in a continuous scan mode with a scanning step size of 0.02° and at a scan rate of $5^\circ/\text{min}$. For phase identification, the obtained XRD patterns were compared with the ICDD data base (ICDD, 2012).

Fourier transform infrared spectroscopy (FT-IR) analysis was performed to identify the major functional groups at the surface of the materials. The spectra were recorded at ambient conditions in the mid-IR region ($400\text{--}4000\text{ cm}^{-1}$) using Nicolet IS 50 FTIR Spectrometer operating in the ATR mode (resolution of 4 cm^{-1} with 32 scans).

The spectrometry of γ -emitters was conducted for 3000 s in plastic beakers of 250 cm^3 . HPGe p-type detector with relative efficiencies of 18% and one n-type detector with relative efficiency of 20% were used. Calibration of detectors was performed using silicone resin matrix spiked with a series of radionuclides (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{203}Hg , ^{88}Y , ^{113}Sn , ^{85}Sr and ^{137}Cs) with total activity of 41.48 kBq on the day of the thirty first of August 2012 (Czech Metrological Institute, Praha, 9031-OL-420/12, type CBSS 2). The background spectrum was recorded before and after sample counting. The spectra were analyzed using GENIE 2000 program, and the results are given at 95% level of significance.

Stability of investigated materials was explored in the solutions with different initial pH. The pH values of deionized water were preset at 2, 6 and 8, by adding minimum amounts of 0.01 mol/L HNO_3 or NaOH . The suspensions having solid/liquid ratio of 1:200 were shaken in PP bottles on laboratory shaker, at 120 rpm, for 24 h. The suspensions were filtered afterwards, and pH values of clear filtrates were measured, as well as concentrations of released Ca^{2+} ions.

Modified US EPA 9045D method for determination of soil and waste pH (US EPA, 2002) was applied in this study. The pH values of CT and RT were determined for different mass ratios of the powders in deionized water, namely 1:1, 1:10, 1:20, 1:100 and 1:200. Suspensions were shaken, at 120 rpm for 5 min, than centrifuged ($5500\text{--}6000\text{ rpm}$ for 15–20 min) and filtered. The measurements of pH values were conducted in both the suspensions and the filtrates using WTW InoLab pH-meter. Furthermore, electrical conductivities of filtrates were measured by WTW InoLab Cond 7110.

2.2. Sorption experiments

Cation sorption was investigated in the batch system. The amount of 0.1 g of each sorbent was shaken with 20 mL of appropriate Sr^{2+} , Co^{2+} and Ni^{2+} solutions, on a rotary shaker at 10 rpm, at room temperature. The solutions were prepared using nitrate salts of investigated cations (Merck, p.a.). Initial pH values of the solutions were in the range 5–5.6, and they were applied without further pH adjustments. Aforementioned conditions were constant in all sorption experiments.

Process kinetics was investigated to define contact time between the sorbents and the solutions, required for attaining sorption equilibrium. As the equilibrium time generally increases with the increase of initial metal concentration, experiments were conducted taking into account the highest concentration of cations used in further work ($\sim 8 \times 10^{-3}\text{ mol/L}$). In that manner, sufficient contact was ensured for the entire investigated concentration range. Contact time varied between 15 min and 72 h.

The effect of initial cation concentration on the amounts sorbed at equilibrium from single-component solutions was determined in the range from $\sim 10^{-4}$ to $\sim 8 \times 10^{-3}\text{ mol/L}$. Based on kinetics experiments, contact time was set at 48 h.

Competitive sorption of Sr^{2+} , Co^{2+} and Ni^{2+} ions was explored using mixture simplex centroid experimental design methodology. This methodology was selected in order to fit the experimental data with appropriate mathematical models and express response variables as functions of components' proportions. Analysis of mixture experiment enables the modeling of blending surface, so that the prediction of the response for any mixture component, (individually or in combination) can be made empirically. Thus, the proportions of components in the mixtures were set by simplex centroid design with 10 runs (Table 1), while the total metal concentration was fixed at $3 \times 10^{-3}\text{ mol/L}$.

After equilibration, the suspensions were filtered and pH values of filtrates were measured as well as residual metal concentrations. All sorption experiments were run in duplicate.

2.3. Measurements of cation concentrations

The concentrations of Sr^{2+} , Co^{2+} , Ni^{2+} and Ca^{2+} ions in material stability and sorption experiments were determined using Perkin Elmer 3100 Atomic Absorption Spectrometer (AAS). For

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