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Utilization of Spent Adsorbent in a Ceramic Matrix †

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Abstract: Reducing waste, including waste generated through environmentally protective processes, is a primary goal of the circular economy. It is important to properly dispose of waste to promote sustainability and a cleaner environment. This article considered the disposal method for spent adsorbents using ceramic technology. The results indicate that the addition of spent adsorbent does not significantly reduce the performance characteristics of ceramic materials while providing an efficiency of over 99% for Cr (VI) retention in all experiments.

Keywords: adsorption; heavy metals; utilization; clay; iron-containing silicate composites

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1. Introduction

Chromium (Cr) is a toxic element that, with long-term exposure to a living organism, even at low concentrations, can cause damage to the immune system [1]. According to the latest update of the European Pollutant Release register, a total of 512 facilities from EU countries are registered as they release chromium compounds into air and water [2].

Today, adsorption is one of the most promising and effective methods of large treating volumes of water from heavy metals, which are contained in low concentrations compared to other compounds. Most often, carbon adsorbents, zeolites, natural clays, silica, and modified materials based on them are used for this purpose [3,4].

Widely used acid solutions for regenerating spent adsorbents are unsuitable for iron-containing materials due to their high reactivity and dissolution of iron compounds. Consequently, there is a need for their safe disposal to avoid repeated contamination of groundwater and surface water with heavy-metal ions and their subsequent bioaccumulation in living organisms. Usually, materials are disposed of through pyrolysis, burial in a landfill or use in other manufacturing technologies to curtail waste production.

In [5], an overview was given of alternative options for the use of spent biosorbents, particularly in the production of cement and biofuel. The potential for the possible use of a hybrid sorbent based on graphene oxide (GO/Fe-Mn) after the removal of Pb (II) ions as an additive to the cement mix is shown in [6]. The efficiency of Ni (II), Zn (II), and Cd (II) retention of more than 88.5% has been demonstrated by ceramic bricks with the addition of spent sorbents based on agro-industrial waste (sawdust, sunflower, and maize) [7].

Nevertheless, scientific sources give rather limited information on the utilization and toxic effects of spent materials used for the purification of contaminated water systems from anionic forms of toxicants (Cr, As, Mo, etc.) [8,9].

The aim of this study was to investigate the possibility recycling of the spent aluminosilicate adsorbent in the manufacturing process of construction ceramics after its use in the removal of Cr (VI) compounds from aquatic environments.

2. Materials and Methods

The object of this study was a spent adsorbent based on saponite (Ukraine) with an applied layer of ferrihydrite in a ratio of 1:1 (Sap-Fh), synthesized according to [10].

To simulate a contaminated water system, a model solution with a Cr (VI) concentration of 50 mg/dm^3, a pH of 5.2, and an ionic strength of 0.01 based on NaCl was employed. Upon achieving equilibrium, the sample under scrutiny removed 55% of Cr (VI).

The method of utilisation consisted of the addition of Sap-Fh to the ceramic mass during the production of bricks by means of the plastic moulding process. The main component of the ceramic mass consisted of clay from the Veselovske deposit (Ukraine). A series of beams samples, measuring 44.5 × 11.3 × 7.0 mm (l × w × h), were produced with varying amounts of spent adsorbent (5, 10, 15 wt.%). The moulding moisture content of the ceramic masses was 22%. The firing was carried out in a muffle furnace at a temperature of 1050 °C for 2 h.

The flexural strength, water absorption, and porosity of the studied material were determined using conventional techniques [11,12].

Leaching studies were carried out in possible environments of use of this type of building ceramics. For this purpose, model solutions of tap water, water with a high level of mineralisation (3000 mg/L) and water with pH 4 were used. The presence of Cr (VI) ions in the model solutions was analysed by inductively coupled plasma atomic emission spectrometry (Thermo Scientific iCAP 7400 ICP-OES, USA) after exposure of the samples to the solutions continuously for 10 and 60 days.

3. Results and Discussion

Clay of the Veselovske deposit is a kaolin–hydromica raw material by its mineralogical composition, and contains quartz impurities and inclusions of zircon, rutile, ilmenite, and anatase, i.e., it belongs to the group of polymineral clays. This type of clay is used as one of the components of ceramic charge to produce building ceramics, such as tiles and bricks.

The firing results are presented in Figure 1, where the variances in colour intensity of samples are attributed to the incremental concentration of the admixture.

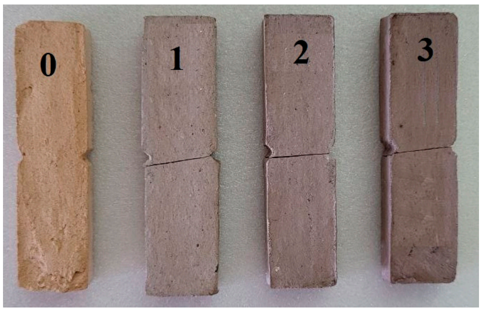

Figure 1. Experimental samples (arranged according to the increasing amount of added Sap-Fh from **left** to **right**).

The composition of the ceramic charge, the main mechanical properties and structural characteristics of obtained samples are given in Table 1.

Table 1. The main mechanical properties and structural characteristics of investigated samples.

Sample	Ceramic Charge, wt.%	Plasticity	Flexural Strength, $\sigma_{f.s.}$, MPa	Water Absorption, W, %	Porosity, %
0	100/0 *	28.91	7.18	9.28	8.33
1	95/5	26.22	5.98	9.05	8.30
2	90/10	23.31	5.85	10.70	9.66
3	85/15	20.50	5.10	11.38	10.22

* mass ratio of components clay/Sap-Fh.

The obtained experimental data demonstrate that the addition of spent adsorbent, from 5 to 15 wt.%, increases the fired samples' porosity by 2%, resulting in a marginal increase in water absorption and a decline in the flexural strength index. The observed changes may be attributed to the physical and chemical processes occurring during firing, as a consequence of the added Sap-Fh.

The immobilisation extent of Cr (VI) oxyanions within the ceramic matrix was assessed following extended exposure to aqueous environments of different chemical compositions (Table 2).

Table 2. Results of Cr (VI) desorption experiments.

Sample	Tap Water		Highly Mineralization Water		Water with pH 4	
	10 Days	60 Days	10 Days	60 Days	10 Days	60 Days
1	<d.l.	0.01	<d.l.	0.01	<d.l.	0.01
2	<d.l.	0.01	<d.l.	0.02	<d.l.	0.02
3	<d.l.	0.01	<d.l.	0.02	<d.l.	0.02

The data obtained indicate that, over 2 months of continuous exposure to potential operating environments, there is minimal leaching of chromium. The solutions' analysis revealed that the chromium content is below the device's detection limit (d.l.) and, therefore, within the maximum allowable concentration (0.05 mg/dm^3). This could be attributed to the fact that, when exposed to temperatures above 700 °C, mobile chromium compounds are assimilated into fusible silicates, generating stable chromium phases with the clay minerals present in the clay of the Veselovske deposit. This ultimately diminishes the bioavailability of Cr (VI).

4. Conclusions

The obtained results suggest that spent iron-containing adsorbents based on clay minerals may find application in ceramic technology, specifically as an additive in the production of building ceramics.

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