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Removal of heavy metals from sewage sludge using combined hydrothermal pretreatment and chelate extraction

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Abstract

The removal of heavy metals from sewage sludge using the combined influence of hydrothermal pretreatment and chelate extraction was tested. The classical method of batch extraction with chelates and the advanced hydrothermal pretreatment (HTP) extraction process with chelates were compared. Both experiments with removal of selected HMs were performed with 0.1 M solutions of 5 different chelating agents: citric acid (CA), S-carboxymethyl-L-cysteine (SCLC), ethylenediamine disuccinic acid (EDDS), methylglycine diacetic acid (MGDA) and ethylenediaminetetraacetic acid (EDTA). The sequential order of potential extraction efficiency at chelates was found as EDDS > EDTA > MGDA >>> SCLC > KC. The mixture of sewage sludge with high sand content soil (A) had in all cases the higher removal efficiency in comparison to the sewage sludge mixture with clay soil (B). The removal of the tested HMs from mixture A and B was better than from the sewage sludge alone. HTP extraction method showed better removal efficiency and significantly shorter time of the process. Also, this extraction method had the higher potential of efficiency in the mixture of soils and sewage sludge. More detailed research on this topic is desirable.

Keywords: sewage sludge; heavy metals; hydrothermal pretreatment; chelate extraction.

1. Introduction

Municipal sewage sludge is a by-product from the waste-water treatment process (WWTP). The common characteristic of sewage sludge is its high environmental and health hazards, primarily due to toxic heavy metals (HMs) content (Usťak and Váňa, 2004; Benitez et al., 2001). On the other hand, it is a valuable substrate for application into the soil due to its high nutrient and organic matter content. Therefore, there is a need to remove high concentrations of heavy metals (HMs) for the use of sludge in fertilization purposes. One of the few prospective suitable sewage sludge treatment processes is the removal of HMs, which is usually accomplished by washing with various chemical agents (Tandy et al., 2004; Peters, 1999). It is the so called biowaste washing, which involves the separation of pollutants from the solid fraction by dissolving them in the washing solution (Babel and Del Mundo, 2006). There are many different ways for such HMs removal, but for our experiments we have chosen both the classical method of batch extraction with chelates, and the advanced hydrothermal pretreatment (HTP) extraction process with chelates (Renu et al., 2016).

Unfortunately, there are not enough experimental results about these processes. Therefore, it was necessary to assess the effect of HTP of sewage sludge in combination with different extractants for removing some HMs.

2. Materials and Methods

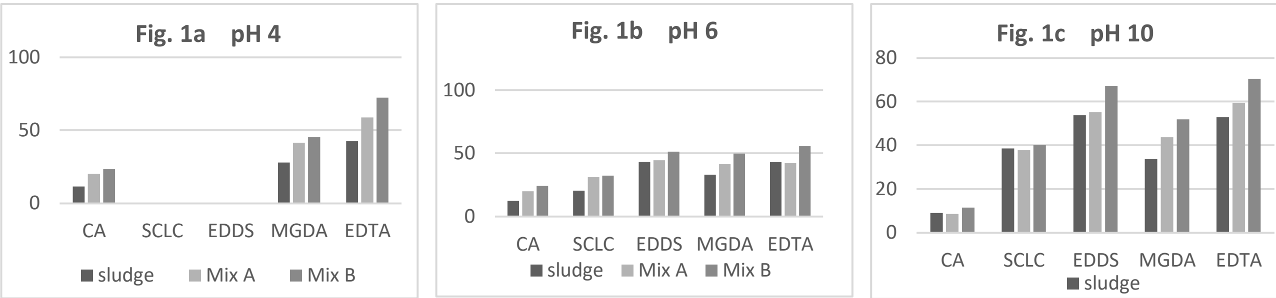
For the experiments was selected a type sewage sludge supplied by the company Wekus Inc. To this sludge was either added clay soil (mixture A) or soil with high sand content (mixture B) in a dry weight ratio of 1: 1. ICP-OES technique (Integra XL, GBC, Australia) was used to determine the concentration of individual HMs in extraction solutions. The total content of 6 selected HMs in the tested sewage sludge, determined by the regia extract, was as follows (mg/kg dry weight): 3.2 Cd, 128 Cr, 118 Cu, 62.6 Ni, 72.4 Pb and 1280 Zn. These values show that the limit values used in the Czech Republic for sludge application on agricultural lands were exceeded for a range of elements (see Decree No. 437/2016 Coll., On the conditions of use for treated sludge on agricultural land).

The experiment methodology was based on the work of Paulauskas et al. (2017). Both experiments with removal of selected HMs were performed with 0.1 M solutions of 5 different chelating agents: citric acid (CA), S-carboxymethyl-L-cysteine (SCLC), ethylenediamine disuccinic acid (EDDS), methylglycine diacetic acid (MGDA) and ethylene-diaminetetraacetic acid (EDTA). In addition to the extraction technique, as variables were also tested pH, temperature and reaction time. EDTA was used as the trisodium salt, i.e. Na_3 -EDTA, while EDDS was used as the S,S isomer because R,R and S,R isomers are not readily biodegradable. The batch extraction was carried out at room temperature on an orbital shaker at 160 rpm for 8 hours, with a solid/liquid ratio of 1:10. The HTP process extraction was performed at 3 different temperatures, 50, 100 and 150 °C and at two-time intervals of 15 and 60 min.

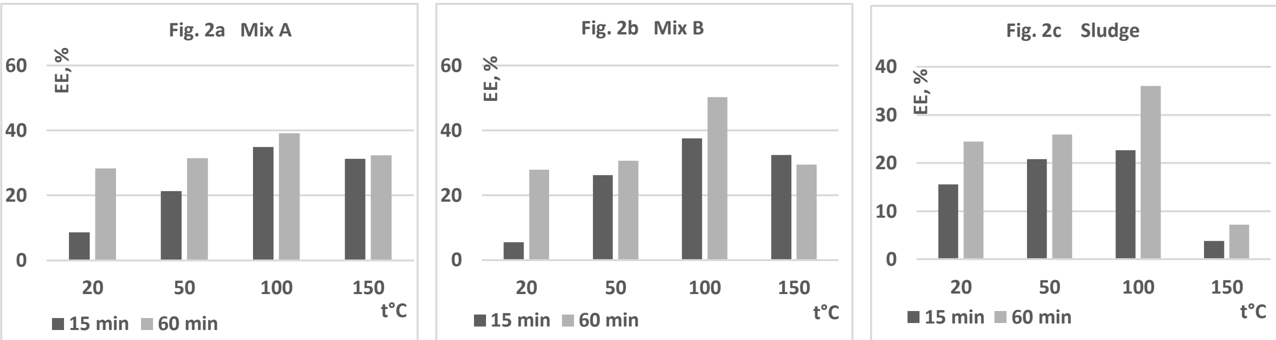
3. Results and discussion

Due to limited content of this article, the results at one of the six observed elements, namely copper (Cu) here are presented for illustration (see Figures 1 a, b, c). The figures for other elements are quite similar. As we can see, EDDS and MGDA were more able to extract high amounts of HMs from the solids than EDTA. The order of the investigated substances according to the potential extraction efficiency was as follows: EDDS > EDTA > MGDA >>> SCLC > KC. Large differences between potential extraction efficiency can be explained by the uneven strength of individual chelates in complexation with HMs. Comparing the removal of HMs in various solids, this showed that the mixture of sewage sludge with high sand content soil (mixture A) had the higher removal efficiency in all cases than the sewage sludge mixture with clay soil (mixture B). Moreover, the removal of the tested HMs from mixture A and B was better than from the sewage sludge alone. This cannot be explained only by the different grain size, but also by the significantly higher proportion of organic matter content in the sewage sludge itself, which obviously reduces the efficiency of chelation. This fact is particularly important for copper, which tends to form stable complexes with organic matter in soil and sludge.

At HTP extraction was shown better removal efficiency and significantly shorter processing time. As an example, there are presented results for one of the six observed elements, cadmium (Cd), see Figures 2 a, b, c. The figures for other elements showed quite similar patterns, but are not listed here due to the limitations of this publication. Under high temperature conditions (150 °C), the amino-polycarboxylic acids examined seemed to exhibit lower chelating ability to HMs ions, or even such metal chelating complexes become unstable and subsequently lose the ability to extract HMs. Similar to batch leaching, the HTP extraction method had a higher potential efficiency in the mixture of soils and sewage sludge than in the sewage sludge itself. On the whole, the achieved results confirmed the conclusions of previous researches based on a similar methodology (Paulauskas et al., 2017).



Figures 1 a, b, c. Extraction efficiency (EE, %) of Cu using 0.1 M chelate solution according to three pH variants and three substrate variants (at pH 4 without SCLC and EDDS due to precipitation)



Figures 2 a, b, c. Examples of Cd Removal by HTP with 0.1 M MGDA with extraction from three different substrates (a- mixture A; b- mixture B; c- sludge)

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