



Stabilization of heavy metals in lightweight aggregate made from sewage sludge and river sediment[☆]



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HIGHLIGHTS

- We use sewage sludge and river sediment to produce lightweight aggregate (LWA).
- We investigate the effects of K on the stabilization of heavy metals in LWA.
- Minimum heavy metals leachability can be obtained at K between 0.175 and 0.2.
- Heavy metal solidification rates above 95% in acidic solutions.
- LWA can be used as an environmentally safe material for civil engineering.

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ABSTRACT

The primary goal of this research is to investigate the stabilization of heavy metals in lightweight aggregate (LWA) made from sewage sludge and river sediment. The effects of the sintering temperature, the $(\text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO})/(\text{SiO}_2 + \text{Al}_2\text{O}_3)$ ratio (K ratio), $\text{SiO}_2/\text{Al}_2\text{O}_3$ and $\text{Fe}_2\text{O}_3/\text{CaO}/\text{MgO}$ (at fixed K ratio), pH, and oxidative conditions on the stabilization of heavy metals were studied. Sintering at temperatures above 1100°C effectively binds Cd, Cr, Cu and Pb in the LWA, because the stable forms of the heavy metals are strongly bound to the aluminosilicate or silicate frameworks. Minimum leachabilities of Cd, Cr, Cu and Pb were obtained at K ratios between 0.175 and 0.2. When the LWA was subjected to rigorous leaching conditions, the heavy metals remained in the solid even when the LWA bulk structure was broken. LWA made with sewage sludge and river sediment can therefore be used as an environmentally safe material for civil engineering and other construction applications.

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1. Introduction

Typical sewage sludge disposal methods, such as landfill, incineration, and agricultural applications, have become subject to increasing public opposition and stricter regulatory pressure because of groundwater pollution, odor emissions, soil contamination and heavy metal pollution [1–8]. Heavy metal pollution affects the use of sewage sludge [9]. Heavy metals such as Cu, Cd, Pb, Hg and Cr are found at relatively high concentrations in sewage sludge [10]. The total heavy metal content of sewage sludge is about 0.5–2.0% (dry weight), and in some cases may be as high as 4% (wet weight), particularly for metals such as Cd, Cr, Cu, Pb, Ni and Zn [11]. Therefore, land application of contaminated sludge releases heavy metals

into the soil when the sludge organic matter decomposes [12]. Furthermore, biomagnification of these heavy metals through the food chain affects human health and the environment [13]. In developing countries, sludge is often disposed of in open fields because of the shortage of appropriate disposal facilities, resulting in serious problems because of heavy metals leaching into groundwater, surface water and soil [14].

In recent years, various protocols for removing or stabilizing heavy metals in sewage sludge have been studied to minimize potential risks to human health and the environment. Heavy metal concentrations in sewage sludge can be reduced by chemical extraction, bioleaching, electrokinetic processes and supercritical fluid extraction [15]. An effective method for decreasing heavy metals leaching from sludge is stabilization, which can be achieved using physicochemical reactions at high temperatures or pressures. Re-use and environmental “neutralization” of waste are among the current energy-efficient and environmental-friendly methods for waste treatment [16].

Using sludge to make lightweight aggregates (LWAs) or ceramics is a promising solution because it not only avoids secondary

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pollution, but also adds value to the sludge by transforming it into a useful material [17–24]. LWA production methods have added sewage sludge or sewage sludge ash to clay. However, these methods require large quantities of clay, which is nonrenewable resource, and excessive exploitation will lead to its depletion, and may also lead to the destruction of arable land. The manufacture of clay bricks is currently prohibited in China. This has prompted research into clay substitutes for the production of LWA to contribute to both the sustainable development of natural resources and the protection of the environment.

River sediment contains SiO_2 , Al_2O_3 , Fe_2O_3 , CaO and MgO , which are the main components of LWA. Previously we verified that LWA produced with sewage sludge and river sediment is an effective and feasible way of reusing this “waste” [25]. To the best of our knowledge, there have only been a few studies investigating the effectiveness of heavy metals in LWAs made from sewage sludge and river sediment. Therefore, the objectives of this study were to:

- evaluate the safety of LWA made using sewage sludge and river sediment as primary materials,
- assess the effect of sintering temperature and K ratio on the stabilization of heavy metals,
- assess the effect of the oxidative conditions on leaching behavior,
- assess the effect of pH and H_2O_2 on the leachability of heavy metals,
- investigate the heavy metal forms present in the LWA, and
- analyze the solidification mechanism and establish effective parameters for evaluating the application of LWA.

2. Materials and methods

2.1. Materials

Sewage sludge was obtained from the Wenchang Wastewater Treatment Plant in Harbin, China. River sediment was obtained from the Hejiagou River, which flows through Harbin. Na_2SiO_3 with a modulus of approximately 3.2 was used. Sewage sludge and river sediment were dried to constant weight at 105°C , then ground and passed through a 0.154 mm sieve. The chemical characteristics of the sewage sludge and river sediment are shown in Table S1 (Supporting Information).

2.2. Methods

2.2.1. Preparations of LWA

Ten grams of sewage sludge, river sediment and sodium silicate mixture, at a mass ratio of 1:1:0.1 (w/w/w), were mixed with 8 mL of water [25]. The mixture was then made into 6–10 mm pellets and dried at room temperature (25°C) for at least 5 days. The samples were dried further at 110°C in a blast roaster for 24 h. The samples were then prepared by heating from an initial temperature of 25°C at a rate of 8°C min^{-1} in a muffle furnace, with 10-min constant temperature periods at 200°C , 600°C and 800°C . Finally, separate samples were baked at the test temperatures (950°C , 1000°C , 1050°C , 1100°C , 1150°C , and 1200°C) for 30 min, before being allowed to cool naturally to room temperature. The samples were stored in a desiccator before analyzing their physical properties and leachability. The LWA production process is shown in Fig. S1 (Supporting Information).

To investigate the effectiveness of the solidification of heavy metals (Cr, Cd, Pb and Cu) in the sewage sludge and river sediment during the production of LWA, a solution of heavy metals (K_2CrO_4 , $\text{Cd}(\text{NO}_3)_2$, $\text{Pb}(\text{NO}_3)_2$ and $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$) was added to the raw materials during the LWA pellet production. The contents of Cd, Cr, Cu and Pb were determined from analysis of activated

sludge at different locations in China as shown in Table S2 (Supporting Information). The simulated heavy metal concentrations in the raw materials (sewage sludge and river sediment mixed, at mass ratio 1:1) were: Cd $50 \mu\text{g g}^{-1}$; Cr $1000 \mu\text{g g}^{-1}$; Pb $1000 \mu\text{g g}^{-1}$ and Cu $500 \mu\text{g g}^{-1}$.

2.2.2. Characterization of LWA

(1) Chemical analysis

The chemical components of sewage sludge and river sediment were determined using a Philips PW 4400 XR spectrometer (X-ray fluorescence, XRF, PANalytical, Amsterdam, the Netherlands). Powder X-ray diffraction (XRD) spectra of LWA were recorded on a D/max- $\gamma\beta$ X-ray diffractometer at 50 mA and 40 kV, Cu $K\alpha$ radiation (Rigaku, Japan).

(2) Heavy metal leaching test

The toxicity of the aggregate samples was determined using a modified method based on the toxicity characteristic leaching procedure [19], a standard method for determining waste leachability, and an update of the hazardous waste extraction procedure provided by the US Environmental Protection Agency. The leaching test was conducted on a solution prepared at a liquid–solid ratio of 1 L 200 g, and stirred at 110 rpm for 24 h. The supernatant was analyzed using a PerkinElmer Optima 5 300 DV inductively coupled plasma atomic emission spectrometer (ICP-AES, Waltham, MA, USA).

3. Results and discussion

3.1. Effect of sintering temperature on the stabilization of heavy metals

To gain a better understanding of the effect of heat treatment on the leaching characteristics of heavy metals in LWAs, the LWAs were broken into pieces of different diameter (D): (1) unbroken LWA, (2) $2 \text{ mm} \leq D \leq 5 \text{ mm}$ and (3) $D \leq 2 \text{ mm}$

The data presented in Fig. 1 show that the leachable Cd, Cr, Cu and Pb contents in the three different diameter LWA samples decreased significantly as the sintering temperature increased from 950°C to 1050°C . The leachable Cd and Cr contents in the LWA samples of each diameter changed only slightly at sintering temperatures above 1050°C . The leachable Cu and Pb contents also only changed slightly at temperatures above 1100°C . Increasing the sintering temperature above 1100°C only had a minor influence on the leachability of the four heavy metals. We also observed that the smaller diameter ($2 \text{ mm} \leq D \leq 5 \text{ mm}$ and $D \leq 2 \text{ mm}$) samples prepared at temperatures between 950°C and 1050°C had more leachable heavy metals, but this difference was less pronounced in samples prepared above 1050°C .

Samples prepared between 950°C and 1050°C had relatively “loose” internal structures and semi-developed crystalline phases, so the heavy metals were not completely incorporated into the LWA and were easily leached. Sintering temperatures above 1050°C promoted the generation of a liquid phase followed by a crystal phase. This resulted in more efficient heavy metal solidification, with the heavy metals being locked inside the crystalline structures, even when the LWA samples were broken. Solidification of heavy metals was consistently observed for LWAs sintered above 1050°C .

XRD analyses identified the form of the heavy metals in the LWA and transformations of heavy metals that occurred in the heating process (Fig. 2A). The major mineral phases in the sintered samples were quartz (SiO_2), albite ($\text{Na}(\text{AlSi}_3\text{O}_8)$) and hematite (Fe_2O_3).

The data shown in Fig. 2B illustrate that heavy metals in the LWA were in stable forms, and that the main compounds were Cd_2SiO_4 , Cr_2O_3 , CuO and PbCrO_4 . The formation of Cd_2SiO_4 reveals that Cd could enter liquid–solid phases and combine with silicate in the

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