



Research article

Feasibility of using demolition waste as an alternative heavy metal immobilising agent

Seelawut Damrongsiri ^{a, b, *}^a Environmental Research Institute, Chulalongkorn University, Bangkok 10330, Thailand^b Research Unit of Site Remediation on Metals Management from Industry and Mining (Site Rem), Chulalongkorn University, Bangkok 10330, Thailand

ARTICLE INFO

Article history:

Received 4 October 2016

Received in revised form

19 January 2017

Accepted 22 January 2017

Available online 3 February 2017

Keywords:

Demolition waste

Heavy metal

Acid neutralising capacity

Immobilising agent

Cement paste

Lightweight concrete

ABSTRACT

Demolition waste consisting of cement paste, lightweight concrete and bricks is a worthless material generated by a growing city. However, research suggests that it may be applied as an alternative heavy metal immobilising agent. The diverse characteristics of demolition waste were examined. Cadmium was selected as there presentative heavy metal to investigate demolition waste adsorption capacity. The solid-liquid distribution coefficients (K_d) were observed. The adsorption isotherms were applied to investigate adsorption characteristics. Carbon content in the demolition waste materials was low and mainly in inorganic form. Cement paste and lightweight concrete had an alkaline pH with very high acid neutralising capacity (ANC). The surface area and cation exchange capacity (CEC) of these materials were low. Cement paste possessed the highest pH, ANC, CEC and surface area, with the highest K_d ; however, its specific surface area and CEC were low compared to activated carbon and organic material. The adsorption isotherms indicated surface heterogeneity with favourable conditions for adsorption and the mean free energy suggested physisorption with multilayer formation. The K_d values of the tested materials were comparable to soil which was not effective in immobilising heavy metal via adsorption mechanisms. However, the high pH and ANC of cement paste and lightweight concrete can improve the heavy metal adsorption capacity of soil and soil ANC that help prevents and controls leaching by heavy metals.

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1. Introduction

Heavy metal contamination in soil is a global problem with increasing environment consequences. Mining activity involving heavy metal extraction from the original subterranean location causes potential toxic contamination of the terrestrial environment. Water and oxygen generate acid from the sulphides in the rock which render the heavy metals unstable and contamination spreads to the surrounding environment through leaching as surface runoff or groundwater (Li et al., 2014; Quinton and Catt, 2007; Wuana and Okieimen, 2011).

The *in situ* immobilisation technique has the potential to reduce this heavy metal distribution and, currently, this is a best demonstrated available technology for remediation of heavy metal contaminated sites (GWRAC, 1997). The advantages of this

approach are low invasivity, simplicity and rapidity, relatively low cost, minimal waste produced and application to a wide range of inorganic pollutants (Wuana and Okieimen, 2011). A stabilising agent is introduced to produce more chemically stable form by various combination mechanisms: adsorption, ion exchange, precipitation, complexation and speciation. Suggested immobilising agents include lime, phosphate salt, fly ash and Portland cement (Guo et al., 2006).

The immobilisation ability of each agent or material is different depending on its various characteristics (Houben et al., 2012; Venegas et al., 2015). The pH is the most significant parameter influencing metal adsorption as it affects metal chemistry both in solution and on the soil surface (Adriano, 1986; Koptsik, 2014; McLean and Bledsoe, 1992). Generally, higher pH values increase metal sorption capacity of soil resulting in decreasing metal mobility (Berbecea et al., 2010). The pH of soil is reduced by various acid input processes such as acid rain, fermentation products and acid wastewater mine drainage. Soil is naturally resilient and acid deposition is neutralised by various reactions between soil solution and soil materials through weathering processes, cation exchange

* Environmental Research Institute, Chulalongkorn University, Bangkok 10330, Thailand.

E-mail address: sdamrongsiri@gmail.com.

and aluminium dissolution (Chesworth, 2008). However, these reactions fail with excessive acid input resulting in decrease of soil pH that increases heavy metal mobility. Acid neutralising capacity (ANC) is a measure of the overall buffering capacity against acid input to the soil system. The cation exchange capacity (CEC) is a measure of the soil's ability to adsorb positively charged ions including heavy metals that decrease their mobility (Adriano, 1986; Bradl, 2004). The CEC of soil varies according to the type and amount of soil materials and pH. Alkaline soil has a naturally higher CEC due to the negative soil surface, with less competition between the cations and protons in the soil solution (Havlin et al., 2011). The adsorption capacity of the immobilising agent is an important characteristic. Materials with high adsorption capacity can accumulate leached heavy metals in their structure. Adsorption capacity is enhanced by large specific surface area which is a remarkable attribute of activated carbon and high CEC which is a notable characteristic of some organic materials. Furthermore, the specific qualities of some materials also influence immobilisation. Organic materials have high CEC which promotes the adsorption of heavy metals; however, they can release dissolved organic compounds into the soil solution which increases metal dissolution via complexation resulting in increased metal mobility (Houben et al., 2012). Some organic material is degraded within a short period and subsequently releases adsorbed heavy metals (Akkajit et al., 2013). Thus, the important characteristics of an immobilising agent to provide long-term stability are the ability to elevate soil pH, high acid neutralising capacity, high CEC, high surface area and lack of dissolved organic compound release.

Demolition waste is a by-product from the construction and destruction of buildings which increases continuously alongside city growth. It is usually used for land reclamation or dumped into a landfill. The major component of demolition waste is concrete (73–88%) with the remainder as bricks, aerated concrete and wood (Uyasatian et al., 2007). Poorahong (2002) investigated the removal of heavy metals from wastewater by concrete grains (0.5–1 mm) (mixture of cement paste, sand and rock). Results determined that the pH of the acidic wastewater was increased to slightly alkaline with high removal efficiency of Cr, Cu, Ni, Pb and Zn through a combination of adsorption and precipitation. The elevated pH was the consequence of dissolution of cement hydration product (Giampaolo et al., 2002). Brick is a widely used material made from clay and rice husk; thus, the composition of brick is similar to soil with SiO₂ as a major constituent (Janbuala, 2012). The Cu adsorption mechanism on brick and successfully Cu removal from wastewater was demonstrated by Djeribi and Hamdaoui (2008). Lightweight concrete or aerated concrete is another popular construction material. This is a calcium silicate hydrate in the form of tobermorite with high porosity of up to 80% and various pore sizes (Laukaitis and Fiks, 2006; Narayanan and Ramamurthy, 2000). However, its application for environmental remediation has not been tested.

Cement paste, brick and lightweight concrete could elevate pH and show good efficiency in adsorbing heavy metals from wastewater. They are not an important source of dissolved organic compounds. Thus, they are feasible for use as immobilising agents. The research objective was to study the feasibility of these three materials as alternative immobilising agents for heavy metals. Cadmium, as a ubiquitous contaminant in Thai soil (Akkajit, 2015) was chosen as the heavy metal representative.

2. Materials and methods

The test materials were 1) cement paste from Portland Cement I, 2) lightweight concrete produced from autoclaved cement based material and 3) brick made with clay and rice husk (X-ray fluorescence analysis of these materials is shown in the supplementary

sections). All these materials are commonly used in Thailand. Cement paste was prepared at a water-to-cement ratio of 0.6 and cured for two weeks. The solid material was ground to obtain a 5:1 wt ratio of grain sizes 0.063–2 mm and less than 0.063 mm. The series of Cd solution was prepared from 1,000 mg L⁻¹ Cd standard solution (1,000 mg L⁻¹ ± 1% in dilute nitric acid, certified grade, Fisher Scientific) and the pH was adjusted to 7 by addition of NaOH solution. Deionised water (DI water) (15-MΩ) was used for all solution preparations.

The acid neutralisation capacity (ANC) of the materials was defined as the quantity of acid (meq kg⁻¹) required to reduce the initial pH to 4 to mirror the weathering process and aluminium dissolution in natural soil (Chesworth, 2008). ANC was determined by a pH titration test using 1:100 sample/DI water ratio. Solutions of 0.1 M and 1 M of HNO₃ or NaOH were applied as titrants. The resulting pH of the suspension was measured 20 min after each acid or base addition. Additions of acid or base were repeated until a pH range of 2–12 was achieved. A total organic carbon analyser (TOC-VCPH, Shimadzu, Japan) was used to quantify the carbon content in the materials and CEC was determined by the ammonium acetate method. The specific surface area, pore volume and pore size were calculated using a surface area analyser (AUTOSORB[®]-1, Quantachrome Instruments, USA).

Solid-liquid distribution coefficients (K_d) and adsorption isotherms resulting from the combination of various physical and chemical parameters of both adsorbent and adsorbate. They were used to indicate the adsorption condition and mechanism. Adsorption isotherm experiments must be conducted under the same environmental conditions. However, cement paste and lightweight concrete elevated the pH of the experimental solutions and obstructed the adsorption isotherm study. Therefore, both the ground cement paste and lightweight concrete were washed with a pH 4 HCl solution. The ground materials were mixed with HCl solution at 1:10 solid to liquid ratio and left overnight. The pH of the washing solution was measured and the washing process was repeated until the pH value of the washing solution was 7 ± 0.2. The materials (washed cement paste, washed lightweight concrete and brick) were then pre-equilibrated with 0.01 M CaCl₂ solution for 24 h using a shaker and dried before use. The prepared materials (1 g) were mixed with 40 mL of Cd solution (0.4–4 mg L⁻¹, pH 7) and shaken for 24 h at room temperature to achieve equilibrium (OECD, 2000). The supernatants were separated by centrifuging and filtered through a 0.45 μm filter. Cd concentration was measured using a flame atomic absorption spectrometer (AAnalyst™ 800, PerkinElmer, USA). Reagent blanks and triplicate analyses controlled the accuracy of the results.

The adsorbed Cd concentration (C_{ad}) was calculated from the mass balance between initial concentration (C_i) and equilibrium concentration (C_{eq}) in the aqueous phase using equation (1):

$$C_{ad} = (C_i - C_{eq})/m \times V \quad (1)$$

where C_{ad} is the adsorbed Cd concentration (mg kg⁻¹), V is the volume of solution (L) and m is the mass of material (kg). The K_d was calculated from each initial concentration using equation (2):

$$K_d = C_{ad}/C_{eq} \quad (2)$$

where K_d is the solid-liquid distribution coefficient measured in L kg⁻¹, C_{ad} is measured in mg kg⁻¹ and C_{eq} in mg L⁻¹.

The C_{eq} and C_{ad} experimental results were plotted using the sorption models of Langmuir, Freundlich and Dubinin-Radushkevich and the fitness was observed by its R square. The Langmuir isotherm assumes that metal ions are chemically adsorbed at certain sites. Each site can only adsorb one ion, the energy of

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