



Heavy metal removal from aqueous solutions by calcium silicate powder from waste coal fly-ash

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ABSTRACT

The removal of Ni (II), Cu (II), Zn (II), and Co (II) ions from simulated aqueous solutions using calcium silicate powder (CSP), a new by-product derived from the production of alumina from coal ash, has been studied. CSP showed high efficiency for the removal of these metal ions. The maximum adsorptions were 420.17, 680.93, 251.89, and 235.29 mg/g for Ni (II), Cu (II), Zn (II), and Co (II), respectively. Total (100%) removal of Ni (II) was obtained when the initial concentration was 100 mg/L, indicating that CSP was highly effective even at an extremely low concentration. Adsorption isotherms and kinetics have been studied using different models. It has been found that the adsorption isotherms can best be described on the basis of the Langmuir model, with the kinetics of adsorption following a pseudo-second-order reaction process. The calcium ion concentration was examined before and after adsorption to investigate the mechanism of removal of the heavy metal ions. It was found that the removal of heavy metal ions is mainly achieved through ion-exchange, combined with some adsorption.

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1. Introduction

Heavy metal pollution has become an increasingly serious environmental problem in recent decades, causing numerous diseases and disorders. An action plan for tackling soil pollution in China was released on 28th May 2017. The Action Plan for Soil Pollution Prevention and Control aims to improve soil quality, ensure safe agricultural products and a healthy living environment for people, according to the State Council, China's cabinet. To control soil pollution by heavy metals, China has vowed to cut the discharge of major heavy metal pollutants in key industries by 10% by 2020. Finding a highly efficient remediation technology remains a bottleneck. Various processes exist for removing dissolved heavy metals from aqueous solutions, including ion-exchange, precipitation, phytoextraction, ultrafiltration, reverse osmosis, electrocoagulation, electro dialysis, and adsorption. Adsorption, particularly using low-cost adsorbents, has attracted a great deal of attention from the research community and industry due to its high

efficiency and simplicity of operation.

Numerous studies have been reported regarding the adsorption of heavy metals by both natural and artificial adsorbents. Natural zeolites, which are abundant, low-cost adsorbents, show high adsorption capacity for heavy metal ions in water (Erdem et al., 2004). The kinetics (Kocaoba et al., 2007; Motsi et al., 2011; Panayotova and Velikov, 2002), equilibrium (Kocaoba et al., 2007), and influence of temperature and pH (Jimenez et al., 2004) have been investigated in relation to these materials. Activated carbon adsorbents are widely used in the removal of heavy metals (Adebisi et al., 2017; Demiral and Güngör, 2016; Jusoh et al., 2007; Kang et al., 2008; Tounsadi et al., 2016). Carbon nanotubes and graphene, as new carbon adsorbents, have also been applied in the removal of heavy metals (Duru et al., 2016; Gu et al., 2015; Gupta et al., 2016; Ihsanullah et al., 2016; Sahraei et al., 2017). Biosorption is an alternative for the removal of heavy metals due to its low cost and eco-friendly nature (Alluri et al., 2007; Basu et al., 2017; Mudhoo et al., 2012; Tang et al., 2017; Wang and Chen, 2009). Reducing the cost of adsorbents is a critical issue for spreading the use of adsorption technologies for the removal of heavy metals.

Synthesized calcium silicates as low cost adsorbents have been used in removing heavy metals. Tobermorites were made from

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waste container glass using hydrothermal method and showed high removal capacities for Cd^{2+} and Zn^{2+} (Coleman et al., 2006, 2014). Functionalized calcium silicate nanofibers were derived from oyster shells for removal of heavy metal ions, showing removal capacity of 203 and 256 mg g^{-1} for Cu(II) and Cr(VI) cations (You et al., 2016). The cement based materials have been widely researched for stabilization or solidification of heavy metals (Chen et al., 2009; Tommaseo and Kersten, 2002). All these studies showed the potential application of calcium silicates in the removal of heavy metals.

Coal remains one of the main energy resources. Indeed, coal accounted for 62.0% of the total energy consumption in China in 2016 (Statistical Communiqué of the People's Republic of China on the 2016 National Economic and Social Development http://www.stats.gov.cn/english/PressRelease/201702/t20170228_1467503.html). Fly-ash is one of the emission pollutants of coal combustion, with about 60 million tonnes being discharged annually in China. Dust and soluble components of fly-ash present great risks to the environment. Comprehensive utilization of fly-ash is considered of great importance by the Chinese government. Fly-ash is mainly composed of alumina and silica, with some other metal oxides. Coal resources with aluminum contents in the coal ash of over 50% are abundant in the northwest of China. Overall, the reserve of this kind of ash in China exceeds 20 billion tonnes. A demo-refinery for coal-ash-based alumina has been constructed in the Inner Mongolia Autonomous Region. The strategy of producing alumina from fly-ash has been a typical case of cyclic economy in China. This alumina production route was released as a Chinese policy in 2013. The fly-ash represents an alternative raw material for alumina due to the shortage of bauxite in China. However, the silica-based by-product constitutes a secondary pollutant and will influence the economic efficiency of the production of alumina from fly-ash if it cannot be effectively reused. Therefore, exploring novel routes for the utilization of this silica-based by-product is a promising strategy. During alumina refining, the silica in the fly-ash is removed as calcium silicate powder (CSP), a new by-product. Employment of CSP as an adsorbent to remove heavy metals in water, thereby controlling pollution with waste, should achieve a win-win situation for the economy and the environment.

Herein, we report the mechanism of Ni^{2+} , Cu^{2+} , Zn^{2+} , and Co^{2+} removal by CSP from alumina production, along with data on its performance, to pave a way for application of CSP. It has been found that the high adsorption capacity of CSP for heavy metal ions results from ion-exchange. The CSP has shown super removal capacities of the four used heavy metals. The environmental benefit for soil pollution control has been analyzed.

2. Experimental method

2.1. Chemicals and materials

CSP was supplied by Inner Mongolia Datang International Renewable Energy Resource Development Co., Ltd. (Hohhot, China) and was used as received. Nickel (II) nitrate and cobalt (II) chloride were purchased from TianJin JINKE Fine Chemical Research Institute (Tianjin, China). Ammonium citrate, dimethylglyoxime, ethylenediamine tetraacetic acid tetrasodium salt, hydroxylamine hydrochloride, copper (II) sulfate, zinc(II) nitrate, and sodium pyrophosphate were purchased from Xilong Scientific (Guangdong, China). Sulfuric acid, sodium citrate, acetic acid, orthoboric acid, sodium hydroxide, and hydrochloric acid were purchased from Beijing Chemicals (Beijing, China). 2,9-Dimethyl-1,10-phenanthroline, zincon monosodium salt, 4-(5-chloro-2-pyridylazo)-1,3-diaminobenzene, ethylenediaminetetraacetic acid

(EDTA), and 2-hydroxy-1-(2-hydroxy-4-sulfo-1-naphthylazo)-3-naphthoic acid were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. (Beijing, China). All reagents were of analytical grade and were used without further purification.

2.2. Adsorption

Solutions containing Ni^{2+} , Cu^{2+} , Zn^{2+} , and Co^{2+} were prepared by dissolving nickel (II) nitrate, copper (II) sulfate, zinc(II) nitrate, and cobalt (II) chloride, respectively, in deionized water. Simulated wastewaters containing different concentrations of these four ions were prepared to test the adsorption capacities of CSP. The initial concentrations of Ni^{2+} and Cu^{2+} are 200, 400, 600, 800, 900 and 1000 mg/L respectively. The initial concentrations of Zn^{2+} are 200, 400, 600, 800 and 1000 mg/L. The initial concentrations of Co^{2+} are 100, 200, 300, 400, 500, 600, and 700 mg/L.

Metal uptake experiments were performed in batch reactors (250 mL flasks) placed in a shaker bath. Each metal ion adsorption tests were conducted using six flasks, each containing 0.1 g of CSP and 100 mL of solution. At predetermined time intervals, the adsorption process in one of the flasks was stopped and the solution was filtered to analyze the metal ion concentration. Each flask was used only once, retrieving data for just one time interval to avoid possible errors deriving from changes in the ratio between adsorbent material and solution caused by the removal of the sample solution for analysis. The Ni^{2+} , Cu^{2+} , Zn^{2+} , and Co^{2+} contents were analyzed by spectrophotometry using dimethylglyoxime (National Standard of the People's Republic of China, GB/T 11910–1989, Water quality-Determination of nickel- Dimethylglyoxime spectrophotometric method), 2,9-dimethyl-1,10-phenanthroline (National Environmental Protection Standard of the People's Republic of China, HJ 486-2009, Water quality-Determination of copper-2,9-dimethyl-1,10-phenanthroline spectrophotometric method), zincon monosodium salt (National Standard of the People's Republic of China, GB/T 10656–2008, Analysis of water used in boiler and cooling-Determination of Zinc-Zincon Spectrophotometry), and 4-(5-chloro-2-pyridylazo)-1,3-diaminobenzene (National Environmental Protection Standard of the People's Republic of China, HJ 550-2015, Water quality-Determination of cobalt-5-Cl-PADAB Spectrophotometry), respectively. The Ca^{2+} concentration was analyzed through the EDTA titrimetric method (National Standard of the People's Republic of China, GB 7476–87, Water quality-Determination of calcium- EDTA titrimetric method).

The adsorption of metal ions by CSP was calculated according to:

$$R(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

where R is the removal of metal ions and C_0 and C_e are the initial and equilibrium concentrations (mg/L), respectively.

$$R(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad (2)$$

where q_e is the equilibrium adsorption capacity, V is the volume of the metal ion solution (L), and W is the weight of the adsorbent (g).

Several models have been proposed in the literature to describe adsorption isotherms. In this study, the Langmuir and Freundlich isotherm models were used to describe the relationship between metal ion adsorption and the equilibrium concentration in solution.

The Langmuir model assumes monolayer adsorption and that the surface of adsorbents consists of identical adsorption sites. The linear form of the Langmuir isotherm is expressed as (Huang et al., 2012):

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