



Research article

Synthetic coal fly ash-derived zeolites doped with silver nanoparticles for mercury (II) removal from water

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ABSTRACT

Coal fly ash-derived zeolites have attracted considerable interest in the last decade due to their use in several environmental applications such as the removal of dyes and heavy metals from aqueous solutions. In this work, coal fly ash-derived zeolites and silver nanoparticles-impregnated zeolites (nanocomposites) were synthesized and characterized by TEM/EDX, SEM/EDX, XRD, XRF, porosimetry (BET), particle size analysis (PSA) and zeta potential measurements. The synthesized materials were used for the removal of Hg^{2+} from aqueous solutions. The results demonstrated that nanocomposites can remove 99% of Hg^{2+} , up to 10% and 90% higher than the removal achieved by the zeolite and the parent fly ash, respectively. Leaching studies further demonstrated the superiority of the nanocomposite over the parent materials. The Hg^{2+} removal mechanism is complex, involving adsorption, surface precipitation and amalgamation.

1. Introduction

Coal is one of the world's most abundant and widely spread fossil fuels, with global proven reserves accounting for nearly 1000 billion Mg. It is still the most used type of fuel in many developing and developed countries such as South Africa (93%), Poland (92%), China (79%), India (69%) and the USA (49%) (Bukhari et al., 2015). According to Yao et al. (2015) coal-fired electricity generation accounted for 29.9% of global electricity production in 2011 and it is projected to reach approximately 46% by 2030. The top-ten countries that produce high amount of coal annually are China, USA, Russia, India, Germany, Poland, Indonesia, Australia, and South Africa; Kazakhstan rounds out the list with 120 million Mg/year (World Coal Institute, 2005). Coal consumption data reveal that nearly 5% of world coal consumption is by Eurasian states and Kazakhstan is amongst the three leading CIS states with a massive 22% consumption (Bukhari et al., 2015).

Coal fly ash (CFA) is a by-product from the combustion of coal in power plants. The amount of annually produced CFA is enormous and thus it requires attention and research on possible recycling routes as it can cause serious environmental and health problems (Blissett and Rowson, 2012; Franus et al., 2015). Several efforts have been undertaken globally to effectively reprocess this waste and convert it into value-added products, such as synthetic zeolites which find several applications in agriculture, medicine, industry and in environmental

engineering such as the removal of heavy metals and other contaminants from water and gas purification (Franus, 2012; Jha and Singh, 2014; Tauanov et al., 2017; Wdowin et al., 2014a). Modified forms of CFA-derived synthetic zeolites and their composites with metals, metal oxides and hydroxides further expand the potential application fields (Goscianska et al., 2018; Zhang et al., 2018a,b).

There are several commonly applied methods for CFA conversion into zeolites, namely conventional hydrothermal synthesis, fusion-assisted hydrothermal synthesis, multi-step treatment, microwave-irradiation and sonication. All synthesis methods involve several steps of crystalline phase formation from amorphous CFA, including dissolution, condensation, nucleation and crystal growth (Bukhari et al., 2015). The advantage of conventional hydrothermal treatment is the simplicity of production, however it may produce a mixture of zeolites depending on reaction conditions, i.e. time, temperature, alkaline concentration, etc. Although the multi-step, microwave- and ultrasound-assisted methods produce single-phase zeolites faster and with high yields, they require high temperature treatment (> 773 K), sonication for a certain duration or several steps, which entail additional costs (Belviso, 2017).

The contamination of water resources with heavy metals, particularly Hg^{2+} and its species, is a serious global environmental threat. Mercury, due to its volatility, chemistry and bioaccumulation properties, is considered as one of the most toxic elements that severely affect

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Table 1
Adsorbents for the removal of Hg²⁺ from aqueous phase.

| Adsorbent type | Initial concentration of Hg ²⁺ [g/m ³] | Volume of the Hg ²⁺ solution [cm ³] | pH | Adsorbent dosage, [g] | Removal of Hg ²⁺ [%] | References |
|---|---|--|--------|-----------------------|---------------------------------|---------------------------|
| Activated carbon from organic sewage sludge | 200 | 100 | 5.0 | 4–10 | 83–100 | (Zhang et al., 2005) |
| Weathered coal | 3 | 100 | 6.0 | 0.8 | > 99 | (Meena et al., 2004) |
| Chitosan-coated Magnetite nanoparticles | 6.2 | 50 | 5.0 | 0.67 | > 99 | (Rahbar et al., 2014) |
| Ag/graphene | 100 | 100 | 5.0 | 0.1 | 98 | (Qu et al., 2017) |
| Ornamental rock solid waste | 10 | 125 | 2–8 | 2 | > 90 | (Dos Santos et al., 2015) |
| Flower-like titanate nanomaterial | 100 | 50 | 5.0 | 0.01 | 98 | (Liu et al., 2015) |
| Linde Type A CFA derived zeolite | 10 | 10 | 2.5 | 0.1–1.0 | 94 | (Attari et al., 2017) |
| ZSM-5-mesoporous/HAPT | 2 | 100 | 2–12.5 | 0.02 | 94 | (Abbas et al., 2018) |
| Ag-X CFA derived zeolite | 13.2–575 | 100 | 5–6 | 10 | 90 | (Czarna et al., 2016) |

human beings and the environment (Yu et al., 2016). According to current regulations of the US Environmental Protection Agency and the European Union the maximum allowable concentration of mercury in drinking water is 2 mg/m³ and 1 mg/m³, respectively (AMAP/UNEP, 2013; The European Parliament and the Council of the European Union, 2013). A variety of methods for the removal of mercury from wastewater have been studied, including the application of electrochemical treatments (electrocoagulation, electroflotation, electrodeposition), physicochemical processes (chemical precipitation, ion exchange) and adsorption (activated carbon, carbon nanotubes, rice husk, etc.) (Azimi et al., 2017). Of all these methods adsorption by utilization of waste materials is of utmost interest, as is a low-cost and effective method for the treatment of polluted water and gases. There are numerous publications on the removal of elemental mercury from flue gas (Liu et al., 2018; Shao et al., 2016; Wdowin et al., 2015, 2014b; Yang et al., 2018), however limited studies have been conducted on Hg²⁺ ions removal from water, particularly with utilization of CFA, synthetic zeolites and composites. Table 1 summarizes the adsorbents used for the removal of mercury from water and their efficiency. It should be noted that most of the experiments presented in Table 1 were carried out at pH 4.0 or higher, which in combination to high mercury concentration may result in precipitates and insoluble complexes rendering the adsorption results questionable.

Among the several metal and metal-oxide containing composites silver and silver oxide nanoparticles (NPs) containing zeolite composites have attracted considerable attention owing to the unique property of silver to form amalgams with mercury. Although some other noble metals also form amalgams they are considerably more expensive than silver. There are some studies on utilization of zeolites derived from CFA for mercury removal, however the research on NP-impregnated CFA-derived zeolites for the removal of mercury from aqueous and gaseous phases is very limited. In this paper the synthesis of CFA-derived zeolites and novel silver NPs-impregnated zeolite nanocomposites is presented, along with full characterization of the derived materials and a study on the removal of Hg²⁺ from aqueous solutions. Moreover, a possible mechanism of the mercury removal by the nanocomposite material is proposed.

2. Materials and methods

2.1. Materials and synthesis

The CFA used in the present study is derived from the electrostatic precipitators of Oskemen city power plant (Karazhyra CFA, East Kazakhstan, 252 MW) and labelled as K-CFA. All chemical reagents used were of analytical grade. Synthetic zeolites (K-ZFA) were produced through a conventional hydrothermal alkaline treatment of K-CFA at 383 K, in a 1-dm³ heavy-walled thermal and chemical resistant reactor using 3 mol/dm³ aqueous NaOH (Sigma-Aldrich) as an activation agent.

The stirring speed was kept constant at 125 rpm, while an incubation period of 48 h was selected in all cases after which the final mixtures were filtered and washed several times for the removal of excess NaOH. The solid residues were dried at 343 K overnight and the products obtained were stored in a tightly closed container for characterization and further experiments.

The K-ZFA was then loaded with silver nanoparticles (Ag NPs) to obtain the nanocomposite materials used for mercury removal from aqueous solutions. The Ag NPs impregnation into the structure of zeolites consisted of two main steps: ion exchange and reduction. Initially, zeolite samples were fully dried at 573 K for 3 h to remove any remaining water molecules in their structure. Then, a slightly modified ion-exchange reaction was conducted by mixing 10 cm³ of 0.01 mol/cm³ aqueous solution of AgNO₃ (Sigma-Aldrich) with 1 g of zeolite and left to react for 12 h (Golubeva et al., 2013). Finally, the obtained Ag-exchanged zeolite slurry was dried at 403 K for 3 h. The obtained Ag⁺-ZFA was then annealed for 3 h at 773 K, followed by 4 h of silver ions reduction using NaBH₄ as the reducing agent, thus producing nanocomposites with various Ag NPs loadings (Ag-ZFA). The reaction container was covered with aluminium foil and stored in dark place to prevent oxidation of silver ions. The same procedure was carried out without adding AgNO₃ solution (R-ZFA) to investigate the effect of reduction on the zeolite structure.

2.2. Characterization of materials

Chemical analysis of both fly ash and produced zeolites, with and without Ag NP impregnation, was done using X-Ray fluorescence (XRF, PANalytical). Their mineralogical composition was identified by X-Ray diffraction (XRD) using a Bruker D8-Focus diffractometer with Ni-filtered CuK α radiation ($k = 1.5406 \text{ \AA}$), at 40 kV and 40 mA. The morphological characteristics of the materials were studied by using Scanning Electron Microscopy (SEM) on JEOL 6380LV Scanning Electron Microscope, operating in LV mode at 20 kV equipped with a backscattered electron detector. Spot and area analyses were carried out using a Si(Li) Energy-Dispersive X-ray spectrometer (INCA X-sight, Oxford Instruments) connected to SEM. The nanoscale investigation was performed with a high-resolution JEOL JEM-2100 LaB₆ transmission electron microscope (HRTEM), operating at 200 kV. The samples (~0.2 g) were suspended in alcohol and treated with ultrasound to disaggregate the agglomerated particles. A drop from the suspension was then placed on a 300-mesh carbon coated copper grid and air-dried overnight. The grain microstructure was also studied using a bright field detector in scanning (STEM) mode. Elemental analysis was carried out using an Oxford X-Max 100 Silicon Drift Energy Dispersive X-ray spectrometer connected to TEM, with a probe size ranging from 2 to 5 nm in STEM mode. The porous structure was determined using low temperature nitrogen adsorption measured on Autosorb-1 porosimeter (Quantochrome, UK). The average pore size and total pore volume were

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