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Nanometer mixed-valence silver oxide enhancing adsorption of ZIF-8 for removal of iodide in solution

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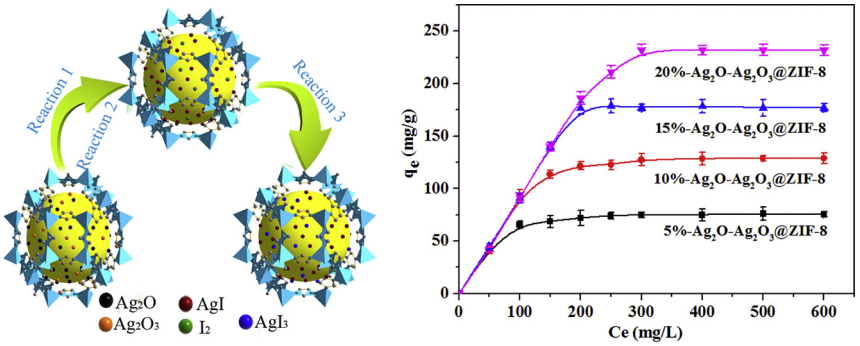
HIGHLIGHTS

- Nano Ag_2O-Ag_2O_3/ZIF-8 composites have been synthesized and characterized for the adsorption of iodide.
- I^- uptake by the Ag_2O-Ag_2O_3/ZIF-8 nanoparticles up to 232.12 mg/g was obtained.
- The high I^- removal originate mainly from the adsorption-induced chemical reaction of Ag_2O and the oxidation of Ag_2O_3.
- Ag_2O-Ag_2O_3/ZIF-8 composites exhibited a wide pH range and interfering ions resistance of I^- adsorption.
- The adsorption mechanism of Ag_2O-Ag_2O_3/ZIF-8 adsorbent was proposed and verified.

GRAPHICAL ABSTRACT



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ABSTRACT

Nano mixed-valence silver oxide (Ag_2O-Ag_2O_3) modified the zeolitic imidazolate framework-8 (ZIF-8) composite (Ag_2O-Ag_2O_3@ZIF-8) was firstly prepared via a simple and efficient method, characterized and applied for iodide ion (I^-) uptake from simulated radioactive wastewater. The results showed that Ag_2O-Ag_2O_3 nanoparticles doped and uniformly dispersed on the surface of ZIF-8 matrix. The adsorption capacity of the as-synthesized adsorbents increased with the increasing Ag doped amount, and the maximum adsorption capacity for 20%-Ag_2O-Ag_2O_3@ZIF-8 was 232.12 mg/g. The calculated thermodynamic parameters indicating that the adsorption was a spontaneous and exothermic. It was worth mentioning that each Ag-based adsorbent exhibited high uptake rate of I^-, and all the adsorption tests were equilibrated for a few minutes. This could be ascribed to its large specific surface area and the absolutely dominant position of chemical adsorption for as-prepared samples. Furthermore, the adsorption was barely affected by pH and competitive anions (e.g. Cl^-, SO_4^{2-}, CO_3^{2-}), even in simulated salt lake water. Additionally, a mechanism explaining the excellent properties for adsorbents could be epitomized into three aspects, namely, the uptake performance of Ag_2O for I^-, the strong oxidization of Ag_2O_3 for I^-, and the adsorption of AgI for I_2.

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1. Introduction

Environmental problems caused by the using of fossil fuels, especially coal, are becoming increasingly severe. It is imperative to seek for alternative clean, safe and efficient non-fossil energy, such as wind energy, water energy. Among them, nuclear energy is playing more and more important role for the production of electrical energy without excessive greenhouse gas emissions (OECD Nuclear Energy Agency, 2003). Studies have shown that the nuclear electricity production was growing steadily with the retirement of existing fossil fuel plants and old nuclear power plants to satisfy the worldwide energy demand (Jang and Lee, 2017; Nandanwar et al., 2016). However, the challenge of radioactive wastes pollution has come into being since the extensive application of nuclear energy (Ma et al., 2014; Montaña et al., 2013). The nuclear power plant accidents, occurred in Chernobyl and Fukushima, have discharged a great number of radionuclides especially ^{129}I and ^{131}I, which were the potential radionuclide contaminants and toxic anion pollutants (Liu et al., 2016; T. Zhang et al., 2017). ^{131}I was isotope of iodine which has a relatively short radioactive half-life of just over 8 days, while ^{129}I was another iodine isotope with the longest radioactive half-life of several million years. Radio iodines or radioisotopes, as the inevitable by-products during uranium and plutonium fission reactions, could easily dissolve in water and thus, become hazardous to jeopardize lives living. Moreover, radioiodine could be accumulated in human body (Fetter et al., 1997). Therefore, effective methods preventing the spread of radioiodine and its safe storage should be developed in order to manage their emissions, so that the use of the important applications can continue to increase significantly in the future with the growth of nuclear energy.

Several studies have focused on the radioactive iodide removal through sorption. As the key of the adsorption, particular attention would be paid to the adsorbents. To develop novel functional materials with strong adsorb-ability is an important direction for radioactive wastewater treatment. In previous studies, various materials, such as cuprous oxide (Mao et al., 2016a; Mao et al., 2017; Zhang et al., 2018), heavy metal (Hg^{2+}, Pb^{2+}) compounds (Ikeda et al., 1994; Mailen and Horner, 1977), silver based materials (Liu et al., 2015; Mao et al., 2016a; Polo et al., 2016), layered double hydroxides (LDHs) (Theiss et al., 2016, 2017), have been proposed to minimize the radioactive I^- concentrations in waste-water. Remarkably, Ag_2O has exhibited outstanding adsorption selectivity due to the strong chemical interaction between silver and iodide (Karanfil et al., 2005; Sánchez-polo et al., 2006). Nevertheless, it was impractical to use Ag_2O directly to entrap I^- in water because of the removal ability and dynamics related to the specific surface area of Ag_2O particles. When silver oxide nanoparticles with a large specific surface area are applied, the adsorption capacity is enhanced. But it will be greatly difficult and costly to separate the thin AgI particles from water. Moreover, Ag^+ in single Ag_2O and I^- anions can only form insoluble AgI with the molar ratio of 1:1, which enormously reduces the utilization efficiency of silver.

A feasible approach to the first conundrum is to immobilize Ag_2O nanoparticles firmly onto the supported-materials with a large specific surface area, and thus can allow the Ag_2O nanoparticles to disperse fully without forming aggregates. To fulfill this goal, Ag_2O anchored $Mg(OH)_2$ (Chen et al., 2017), Ag_2O grafted sodium niobate nanmfibers (Mu et al., 2015) and Ag_2O modified carbon spheres (Yu et al., 2017) have been reported to removal of I^- highly efficient and selectively. However, some challenges and problems still exist and restrict the wide application of those adsorbents, such as complexity of synthesized process, the low stability and sensitivity to solution pH. Thus, the hunt for new support-material that is easily preparation and stable under sophisticated circumstances is quite necessary.

Currently, metal-organic frameworks (MOFs) have received extensive attention as a type of crystalline porous materials due to their large pore sizes, high specific surface areas, tenability and their ability to be chemically tailored (Li et al., 2017; Stock and Biswas, 2012; Wang et al., 2012; Zhang et al., 2017b). The ZIF-8 framework ((Zn $(MeIM)_2$, MeIM) 2 methylimidazole), a typical member of the MOFs group, displayed three-dimensional structural feature, excellent thermal and chemical stability compared to other MOFs. Considering its excellent features, ZIF-8 has been chosen as an outstanding porous carrier in composite adsorbents to remove various pollutants from aqueous solution. For instance, Ag/AgCl/ZIF-8(50%) and Fe_3O_4@ZIF-8 used as the adsorbents for RhB and UO_2^{2+} adsorption respectively. And its exhibited an unprecedented adsorption capacity and selectivity (Liu et al., 2017; Min et al., 2017). Therefore, we believe that ZIF-8 containing nanoparticles have the potential to remove I^- in water.

As for the second challenge, studies have been reported that I^- can be easily oxidized to I_2 owning to the existence of silver nanoparticles under visible light irradiation (Liu et al., 2015). Subsequently, the I_2 could be reacted with the yield AgI precipitates to generate AgI_3, indicating the removal ability of absorbent for I^- was multiplied. However, the adsorbents were greatly influenced by the external environment conditions, and the adsorption capacity was low without UV irradiation (Liu et al., 2015). Therefore, in order to improve the I^- anions adsorption capacity of silver-based adsorbents and full utilization of silver, the high-valence of Ag with strong oxidation is necessary to be introduced. Up to now, high-valence silver oxide has rarely been used as an active component of the adsorbent.

Herein, a novel adsorbent was successfully prepared by anchoring mixed-valence silver oxide on the surface of ZIF-8 with varies contents. The morphology, microstructure and chemical compositions of the Ag_2O-Ag_2O_3@ZIF-8 composites were characterized. Subsequently, the as-prepared Ag_2O-Ag_2O_3@ZIF-8 was used to investigate the adsorption kinetics, isotherms and thermodynamic for I^- from simulated wastewater. Additionally, the effect of pH and interfering ions on I^- removal were also studied. Finally, we proposed the adsorption mechanism for removal of I^- based on the data of adsorption performance and characterizations.

2. Experimental

2.1. Materials

Chemical products, zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$, ≥99.9%), 2-methylimidazole (Hmim, ≥99.0%, Aldrich), trimethylamine (TEA, ≥99.0%, Aldrich), silver nitrate ($AgNO_3$) ethanol (C_2H_6O, ≥99.9%), ammonia ($NH_3 \cdot H_2O$), sodium iodide(NaI, ≥99.0%), potassium chloride (KCl, ≥99.5%), nitric acid (HNO_3, >98.0%), sodium chloride (NaCl, ≥99.5%), calcium chloride anhydrous ($CaCl_2$, ≥97.0%), sodium carbonate anhydrous (Na_2CO_3, ≥99.8%), sodium sulfate anhydrous (Na_2SO_4, ≥99.0%), magnesium chloride hexahydrate ($MgCl_2 \cdot 6H_2O$, ≥97.0%) were all purchased from the Sinopharm Group Chemical Reagent Co., Ltd., China. The I^- anions stock solution (5000 mg/L) was prepared by dissolving NaI in deionized (DI) water, and that solution was diluted to the desired concentration for further use.

2.2. Preparation of ZIF-8 nanocrystals

ZIF-8 composites were synthesized by a typical hydrothermal process according to the Ref. (Sun et al., 2016). Typically, 4.00 g zinc nitrate hexahydrate was dissolved in 24.22 g DI water, which was labeled as solution A. Meanwhile, 4.42 g 2 methylimidazole was dissolved in 96.90 mL DI water. Then 5.80 mL TEA was added in this solution, which was labeled as solution B. Solution A and B were mixed and stirred vigorously at room temperature for 10.0 h. The resulting white precipitates were separated by centrifugation, washed with DI water and ethanol subsequently for 3 times. Finally the white product was dried under vacuum at 80 °C for 12.0 h.