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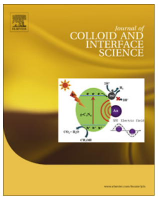

Regular Article

One-step synthesis of $Ag_2O@Mg(OH)_2$ nanocomposite as an efficient scavenger for iodine and uranium

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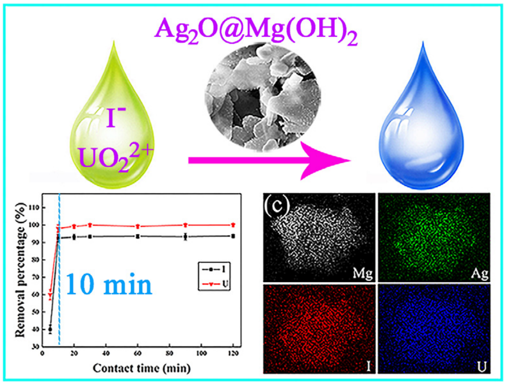
GRAPHICAL ABSTRACT



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ABSTRACT

Ag_2O nanoparticles anchored on the $Mg(OH)_2$ nanoplates ($Ag_2O@Mg(OH)_2$) were successfully prepared by a facile one-step method, which combined the $Mg(OH)_2$ formation with Ag_2O deposition. The synthesized products were characterized by a wide range of techniques including powder X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), transmission electron microscopy (TEM), selected area electron diffraction (SAED), and nitrogen physisorption analysis. It was found that Ag_2O nanoparticles anchored on the $Mg(OH)_2$ nanoplates show good dispersion and less aggregation relative to the single Ag_2O nanoaggregates. In addition, iodide (I^-) removal by the $Ag_2O@Mg(OH)_2$ nanocomposite was studied systematically. Batch experiments reveal that the nanocomposite exhibits extremely high I^- removal rate (<10 min), and I^- removal capacity is barely affected by the concurrent anions, such as Cl^-, SO_4^{2-}, CO_3^{2-} and NO_3^-. Furthermore, I^- and UO_2^{2+} could be simultaneously removed by the nanocomposite with high efficiency. Due to the simple synthetic procedure, the excellent removal performances for iodine and uranium, and the easy separation from water, the $Ag_2O@Mg(OH)_2$ nanocomposite has real potential for application in radioactive wastewater treatment, especially during episodic environmental crisis.

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1. Introduction

Recently, the concern on nuclear waste leakage has drawn increasing attention globally since the nuclear accident at Fukushima in 2011 [1]. The radioactive iodine is one kind of the major byproducts of uranium and plutonium fission, and the half-life of radioiodine isotope differs from about 8 days (^{131}I) to 1.6×10^7 years (^{129}I) [2–4]. In addition, radioactive iodine is also widely applied to the diagnosis and treatment of various diseases [5,6]. For instance, ^{131}I, often used in hyperthyroidism, thyroid cancer diagnosis and metabolic therapies [7], is habitually dumped into domestic sewer system, and thus is one of the radionuclides most often detected in the medical sewage [8–10]. Because of the nonselective uptake of iodine isotope by human body, radioactive iodine can be accumulated in the human thyroid gland and lead to increase of metabolic disorders, mental retardation, and thyroid cancer, especially for children [11]. Studies on iodine speciation in aqueous environment have demonstrated that pH, salinity, natural organic matter (NOM) and microorganisms are involved in regulating iodine biogeochemical processes [11,12]. Dominant iodine species include iodide (I^-), iodate (IO_3^-), and organo iodine in the natural environment [13,14]. It has been proposed that I^- is the major species in deep radioactive waste repositories and is stable almost over the entire pH range [4,15]. Due to the poor removal abilities of many nanomaterials to anions, the implementation in developing nanomaterials for the efficient immobilization of radioactive iodine anions, especially I^-, from aqueous environment is lagged [15].

Various nanomaterials, such as organic anion exchange resins [16], clay minerals [15,17], Ag-based materials [18–27], layered double hydroxide [28–31], and compounds containing Hg^{2+} [32], Bi^{3+} [4,33], and Cu^+ [34,35], have been recently studied for the treatment of water contaminated by radioactive I^-. However, organic anion resin are unstable under high dose of radiation and elevated temperatures [36], and many inorganic materials often display relatively low removal capacities and slow uptake kinetics [4,24]. Among these materials, Ag_2O is a potential capture agent for radioactive I^- because of the efficient I^- immobilization by Ag_2O to form AgI precipitate. Nevertheless, it has been found that the smaller the nanoparticles are, the higher the tendency of aggregation becomes, which stems from high surface free energy. The formation of aggregates could significantly decrease the surface area of Ag_2O, thereby leading to a sharp deterioration of activity and efficiency [24]. Therefore, using Ag_2O directly for the treatment of radioactive I^- is impractical. Anchoring Ag_2O nanoparticles onto the supported-materials could reduce the aggregation of Ag_2O nanoparticles, and thus may improve their performance in I^- uptake. Recently, many Ag_2O-based nanocomposites have been reported for the efficient removal of radioactive I^- [19–24]. For example, Ag_2O anchored titanate nanotubes, nanofibers and nanolamina have been reported to act as scavengers for highly efficient and selective capture of I^- [19,20]. In addition, Ag_2O grafted sodium niobate nanofibers also showed good removal performance for I^- [24]. These findings have provided new insights into the efficient capture of radioactive iodine. However, some shortcomings of the reported supported-materials, such as complexity of preparation, high cost and sensitivity to solution pH, still limit the wide application of Ag_2O. Thus, the search for new substrate that is facilely synthesized and stable under complicated conditions is still crucial and urgent.

As a nontoxic, low cost and environmentally friendly material, nanoscale brucite (nano-$Mg(OH)_2$) is widely applied in the environmental remediation [37–43]. For instance, Li et al. reported that self-supported flowerlike $Mg(OH)_2$ could recycle rare earth elements from low concentration wastewater via ion-exchange [37]. A strategy was proposed by Liu et al. to simultaneously realize both the recycle of $Mg(OH)_2$ nanoadsorbent and enrichment of diluted Cr(VI) from wastewater, which was based on the reversible phase transformation between platelet-like $Mg(OH)_2$ nanoparticles and rod-like $MgCO_3 \cdot 3H_2O$ bulk crystals [39]. In addition, $Mg(OH)_2$ micro/nanorods also showed highly selective adsorption capacity and fast adsorption rate for the removal of dilute anionic dye, and the adsorbed dye was successfully desorbed by carbonation, resulting in a ~4000 fold enrichment of the dye solution [40]. It's well established that nano-$Mg(OH)_2$, having a hexagonal symmetry structure [44], usually exhibits nanoplate-like morphology with a large specific surface area [45], which can potentially provide an ideal substrate for Ag_2O nanoparticle loading. Moreover, nano-$Mg(OH)_2$ could be facilely synthesized by the hydrolysis of periclase (MgO), which is abundant in nature, and thus could be helpful to greatly reduce the preparation cost. In particular, Chen et al. recently reported that a high uranium removal capacity of $Mg(OH)_2$ could be found when the initial uranium concentration crosses over a threshold [46]. Therefore, $Mg(OH)_2$ nanoplates can not only act as a substrate for Ag_2O nanoparticles, but also remove uranium efficiently from solution. That is, Ag_2O coupled with $Mg(OH)_2$ can be potentially applied to the simultaneous removal of iodine and uranium from radioactive wastewater. However, to the best of our knowledge, no research has been reported on the removal of radioactive iodine, especially on the simultaneous removal of iodine and uranium, by $Ag_2O@Mg(OH)_2$ nanocomposite.

Herein, $Ag_2O@Mg(OH)_2$ nanocomposite is successfully prepared by a facile one-step method, which combined the $Mg(OH)_2$ formation with Ag_2O deposition, and the removal ability of the nanocomposite to I^- is systematically investigated. In addition, considering the fact that multiple radionuclides can coexist in radioactive wastewater, the simultaneous removal of I^- and UO_2^{2+} by the nanocomposite is also investigated.

2. Materials and methods

2.1. Materials

All chemicals are commercially available and used as received without further purification. $AgNO_3$, MgO and NaI were purchased from Sinopharm Chemical Reagent Co., Ltd., and are of analytical grade. $UO_2(NO_3)_2 \cdot 6H_2O$ was obtained from Beijing Chemical Industry. Analytical grade nitric acid (HNO_3) and sodium hydroxide (NaOH) were used to adjust the pH whenever necessary. Deionized water was used in all experiments.

2.2. Preparation of $Ag_2O@Mg(OH)_2$ nanocomposite

The $Ag_2O@Mg(OH)_2$ nanocomposite was synthesized using a facile one-step method at room temperature. In a typical procedure, 0.46 g (2.71 mmol) $AgNO_3$ was first dissolved into 50 mL of deionized water. Then 0.40 g of MgO (10 mmol) was slowly added into the $AgNO_3$ solution under continuously stirring. After 12 h of stirring, the precipitate was collected by centrifugation, washed three times with deionized water, and then dried in the vacuum at 60 °C for 12 h. The finally obtained precipitate was designated as sample SOMH-1. For comparison, different amounts of $AgNO_3$ (0.095 and 0.85 g) were used to obtain $Ag_2O@Mg(OH)_2$ nanocomposite with different Ag_2O loadings by the same procedures, which were labeled as samples SOMH-2 and SOMH-3, respectively. The loading of Ag_2O in the nanocomposite is determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES, optima 7300 DV) after sample dissolved by HNO_3 solution. In addition, $Mg(OH)_2$ nanoplates were prepared under the same experimental