



Adsorption of low-concentration arsenic from water by co-modified bentonite with manganese oxides and poly(dimethyldiallylammonium chloride)

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ABSTRACT

Inorgano–organo–bentonite (PMBt) was synthesized through co-modification of bentonite with manganese oxides and poly(dimethyldiallylammonium chloride) (PDMDAAC), and was utilized to remove low-concentration arsenic from water. The hybrid material was characterized by means of X-ray powder diffraction (XRD), scanning electron microscopy (SEM) and N₂ adsorption–desorption. It was revealed that the hybrid material possessed an amorphous structure and displayed uneven manganese oxides particles gathering about the bentonite aggregates with a large BET area of 128.90 m²/g. The effects of adsorption parameters were investigated, such as stirring time, arsenic concentrations, adsorbent dose, temperature, pH, added anions and ionic strength. The adsorption kinetics and equilibrium of arsenic on the co-modified bentonite were further determined. The results revealed that the adsorption of arsenic was dramatically affected with change in solution pH and significantly suppressed by phosphate anion. The adsorption kinetic data was best fitted with the linear pseudo-second-order model and was well with the non-linear Bangham model. The equilibrium data were well simulated with the Brunauer–Emmett–Teller, Freundlich and Redlich–Peterson isotherms by either non-linear or linear regression, respectively. The non-linear Langmuir isotherm showed best fitting and the maximal adsorption capacity was 9.14–9.99 mg/g, dependent on the error function used. It was suggested that the hybrid material had a heterogeneous surface and high affinity towards arsenic, which is highly favorable for arsenic adsorption following a non-ideal monolayer adsorption model. Based on the above comprehensive analysis, the plausible mechanism for arsenic adsorption on PMBt was suggested as a mixed removal mechanism i.e., electrostatic attraction followed by inner-sphere complexation.

1. Introduction

Arsenic enriched groundwater poses a threat of drinking safety of millions for people around the world, especially in developing countries and remote areas. Arsenic pollution of surface waters is still a worldwide environmental problem, mainly resulting from natural mobilization and anthropogenic activities [1,2]. As established by the World Health Organization (WHO), the limit of arsenic contained in drinking water must be further down to 0.01 ppb to prevent the chronic toxic effect of arsenic on human health [3]. Therefore, new and effective remediation techniques to remove low-level arsenic from water have become more and more pressing. Numerous methods have been developed for the removal of arsenic from aqueous solutions, containing modified lime softening, advanced oxidation, chemical precipitation, membrane filtration, coagulation and flocculation, ion exchange, adsorption, solvent extraction, and electro deposition, etc [4,5]. Among these techniques, adsorption is one of the most promising methods

owing to its high-removal efficiency, operational simplicity, cost effectiveness and recycling possibility. The key of adsorption technology lies in utilizing a fast and effective adsorbent with the high adsorption capacity [6,7].

Naturally occurring clay minerals have been widely used as adsorbents for arsenic removal from water and wastewater because they are naturally abundant, easily available and comparatively cost-economical [8,9]. Furthermore, the adsorption capacities of clay minerals can be increased through modifying with acids, bases, cationic surfactants, polyoxocations and/or metal (hydr)oxides [4–6]. There are several investigations involving natural clay minerals in the literatures [10–12], although their adsorption performance is relatively poor. Zhai et al. [10] studied adsorption of trace arsenite (< 200 ug/L) on the two typical clay minerals, i.e. kaolinite and montmorillonite. Their results demonstrated that kaolinite was more advantageous for adsorption of arsenic, as compared to montmorillonite and surface complexation mechanism was proposed to be dominant in adsorption process. Wu

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et al. [11] investigated on adsorption/desorption of arsenate on/from typical minerals contained in soils, such as ferrihydrite, goethite, gibbsite, hematite, kaolinite and montmorillonite. They found that the adsorption capacities of montmorillonite and kaolinite for arsenic were much lower than that of the other clay minerals containing iron and aluminium components. They attributed to the negative character surfaces of the two clay minerals, which is unfavorable for adsorption of arsenic in negative valence state. Mar et al. [12] compared arsenic adsorption on natural geological materials obtained from Indonesia, i.e. lignite, bentonite, shale, and iron sand. Their results showed that the adsorption performance of the four kinds of materials for arsenic had the following order: lignite > bentonite \approx shale > iron sand, and that the amount of arsenic adsorbed was significantly affected by pH with the maximum about near-neutral pH, irrespective of the type of adsorbent and arsenic species. Generally, clay minerals carry a natural negative charge which is not conducive to the adsorption of negatively charged pollutant. But the surface charge of clay minerals can be reversed from negative to positive by modification with cationic surfactant. Moreover, the adsorption capacities of natural clay minerals can be enhanced by means of intercalating with highly charged polymeric or (poly)hydroxy metal species. So far, many studies have been carried out with respect to arsenic adsorption onto raw and single modified clay mineral with metallic polyoxocations (e.g. Al, Fe, Zr, Ti) referred as inorgano-clays [6,13–15], or organocations, such as humic acids (HA), cetyltrimethylammonium bromide (CTMAB), and poly(diallyldimethylammonium chloride) (PDMDAAC) etc. and the resultant is termed as organo-clays [16,17–21]. Doušová et al. [6] reported that the calcined natural kaolin and raw bentonite pretreated with ferrous, aluminous and manganic salts showed remarkable improvements in adsorption affinity towards arsenic oxyanions. Tang et al. [13] synthesized various manganese oxides (i.e. hydrous manganese oxide, birnessite and todorokite) and the oxides coated clay minerals (i.e. kaolinite and montmorillonite). They found that the manganese oxides coated kaolinite illustrated better oxidation property for arsenite, as compared to montmorillonite counterpart. This is due to the improvement in oxidation property of manganese oxide evenly dispersed on the surface of the coated clay minerals. Doušová et al. [14] studied the role of dissolved Fe^{2+} or Mn^{2+} ions played in arsenic adsorption from model groundwater by the calcined product of natural kaolin and bentonite. It was found that Fe/Mn ions were favorably attached to surface sites and co-adsorbed with arsenic onto hydrated Fe/Mn particles formed on adsorbent surface, but only Fe particles contributed to arsenic adsorption considerably. Mishra and Mahato [15] synthesized stable iron and manganese oxide pillared clays as adsorbents towards arsenic. They concluded that the synergistic effect of the nanoxides pillars with active sites and high surface area rather than the clay matrix increased the adsorption capacity substantially. Manganese oxide pillared clays showed better affinity to arsenic owing to in situ oxidation by comparison with the iron oxide pillared. Deng and Zhang [20] compared adsorption of arsenic on natural and modified bentonite with sulphuric acid and/or PDMDAAC and found that the co-modified bentonite showed significantly enhanced uptake of arsenic from real waste effluents. Among the above-mentioned modifiers, PDMDAAC is a kind of eco-friendly organic polymer with being highly positively charged and highly soluble in water and used widely to modify the clay minerals in order to improve both adsorption affinity for anionic contaminants and settling properties of the resultant materials significantly [22]. However, as far as we know, PDMDAAC modified clay minerals are usually used to remove organic contaminants and heavy metals but less applied to strip arsenic from aqueous solution [23–25]. On the other hand, manganese oxides have a higher affinity to arsenic and exhibit rapid oxidation of arsenite to arsenate [26,27]. However, their adsorption capacities are comparatively low thanks to limited specific surface areas, resulting in limiting their applications for water treatment. Nano-structured manganese oxides are renowned for their large surface area and more active and adsorptive sites, allowing excellent oxidation and

good adsorption performance for arsenic [28]. Nevertheless, it is worth noting that nano-adsorbents in the form of a powder have practical drawbacks, such as difficulty in separation of adsorbents from treated water and the high cost of nano materials [29]. In order to overcome these drawbacks, manganese oxide can be embedded with magnetic particles, i.e. oxides of metals such as Fe, Co, Ni and Cu [30–32] and/or immobilized on the surface of low-cost materials with higher specific surface area and/or easy settling property, e.g. activated alumina [29], clay minerals [15,33,34], honeycomb briquette cinders [35], biochar [36], and zeolite [37], etc. Of course, the limitation can also be solved by using manganese oxide-based nanoparticle aggregations of the oxides e.g. iron [32,38], cerium [39], aluminium [40] and zirconium [41].

Recently, arsenic adsorption on co-modified clay mineral with inorganic-inorganic/organic chemicals has been received much more attention, since that the inorgano-organo-clay minerals can possess active sites accessible to adsorption of inorganic and organic pollutants from aqueous solutions at the same time [42]. Moreover, a synergistic effect among two types of modifiers/pillars and clay minerals may help in enhanced uptake of pollutants [43]. Meanwhile, the inorgano-organo modified clay minerals can display significantly additional performances, e.g. the properties of settling, catalytic oxidizing, magnetic and antibacterial, if the modifier/pillar of cationic surfactants [22], manganese oxides [15], iron oxides [44] and silver species [45] are introduced, respectively. To date, little study on clay mineral co-modified with metal oxide and cationic polymer as adsorbents for arsenic removal from water has been presented [46,47]. Earlier we have synthesized a series of single and complex modified bentonites with metal (hydr)oxides (i.e. $\text{Fe}_x(\text{OH})_y$, Mn_xO_y) and/or cationic polymers (i.e. CTMAB, PDMDAAC) and found that the manganese oxides and PDMDAAC modified showed the best adsorption properties for arsenic [48]. In this work, larger quantity of manganese oxides and PDMDAAC co-modified bentonites with proper addition was prepared for the adsorption of low-concentration arsenic from water. X-ray powder diffraction (XRD), scanning electron microscopy (SEM) and N_2 adsorption-desorption techniques were utilized to elucidate the structure and surface morphology of co-modified bentonite. The effects of the adsorption parameters were investigated in detail, such as stirring time, arsenic concentrations, adsorbent dose, temperature, pH, added anions and ionic strength. Kinetics and isotherms of adsorption of arsenic on the manganese oxides and PDMDAAC modified bentonite, and its plausible mechanisms of the adsorbent were also explored.

2. Experimental

2.1. Materials and chemicals

The bentonite sample was obtained from Shanghai Four Hervey Chemical Co., Ltd., China. All the reagents used were analytical grade without further purification. All standard solutions were prepared by dilution with single-distilled water. Anhydrous sodium carbonate (Na_2CO_3 , $\geq 99.8\%$), hydrochloric acid (HCl, 36%–38%), concentrated nitric acid (HNO_3 , 65%–68%), triethanolamine ($\geq 85\%$), copper sulfate pentahydrate ($\geq 99.0\%$) and potassium iodide ($\geq 99.0\%$) were provided from Guangdong Guanghua Science Stock Co., Ltd., China. Zinc particles free from arsenic, stannous chloride dehydrate and silver diethyldithiocarbamate were purchased from Sinopharm Chemical Reagent Co., Ltd., Beijing, China. Fumaric acid ($\geq 99.5\%$) and polydiallyldimethylammonium chloride solution (Mw 200,000–350,000, 20 wt.%) were supplied from Aladdin industrial corporation, Shanghai, China. Concentrated sulfuric acid (H_2SO_4 , 95%–98%) and sodium hydroxide (NaOH) were afforded from Quanzhou Donghai Reagent Co. Ltd., China. Standard solution of As single element (1000 $\mu\text{g}/\text{mL}$, 1.0 mol/L HNO_3 as medium) and potassium permanganate ($\geq 99.5\%$) were buied from National Center of Analysis and Testing for Nonferrous Metal, Xilong Chemical Stock Co., Ltd. Chloroform ($\geq 99.0\%$), sodium

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