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Review article

An overview of As(V) removal from water by adsorption technology

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Abstract

Adsorption has been widely used for arsenic-contaminated drinking water treatment. The mechanism of As(V) removal from water by adsorption technology was briefly introduced, and the influence factors of different adsorption pathways were also illustrated. The materials used as adsorbents were classified according to physical, chemical and biological processes, and the effect on As(V) removal from water using a variety of adsorbents were overviewed in this study. The current problems of this technology were also discussed, then, the solutions and development directions were put forward. Adsorption capability provides significant reference when selecting an adsorbent. Therefore, to develop environmental friendly materials achieving efficient adsorption capabilities, optimize the synthesis and modification methods of the current adsorbents, and improve regeneration efficiency are the trends of developing adsorption technique on As(V) removal from water. Biological arsenic removal technology has the advantages due to good effect, low cost and less secondary pollution, which have bright development and application prospects. Local conditions of arsenic-contaminated water, practical feasibility of the adsorption technology and applicable conditions of each material should be comprehensively compared and evaluated, when selecting specific adsorbent for As(V) removal from water.

Introduction

Arsenic is a chemical element which is well known to human for its toxicity [1]. Chronic toxicity of arsenic via drinking arsenic-contaminated water will threaten human's health [2]. Previous studies reported that long-term exposure to arsenic-contaminated water will cause harmful effects on people's skins, lungs, digestive systems and nervous systems [1,3]. Arsenic is widely distributed in natural water, due to natural geological processes and human activities, such as mining, metallurgy and chemical industry [4,5].

The problem of water pollution caused by arsenic contamination is worldwide for a long time, and it is reported that drinking arsenic-contaminated water had seriously influenced more than 100 million people all over the world [6]. Arsenic removal from water has been an urgent problem to be solved, and reducing the limit of arsenic in drinking water is of great benefit to human's health. Therefore, the contaminant acceptance threshold of arsenic in drinking water in the related standards were more stringent in the

past decades [7]. According to the regulation of World Health Organization (WHO), the maximum permissible limit for arsenic concentration in drinking water was revised from 50 µg/L to 10 µg/L [8].

Arsenic usually exists in two valence states in natural water: As(III) and As(V). The toxicity of As(III) is much higher than that of As(V). As(III) mainly exists in the form of molecular, and As(V) mainly exists in the form of H_2AsO_4^- or HAsO_4^{2-} under neutral conditions, in general, ionic arsenic is more easily removed by physical or chemical processes [9]. Therefore, As(III) can be converted into As(V) by chemical or biological methods to reduce its toxicity and the difficulty of subsequent treatment [10-12].

The main methods for As(V) removal from drinking water include adsorption technology [13-15], coagulation technology [16,17] and membrane technology [18,19]. Arsenic-contaminated water treatment by adsorption technology has been widely adopted due to its advantages such as simple operation, economic benefits, and a variety of materials which



are proven to be used as adsorbents. This study provided an overview of As(V) removal from water by adsorption technique, in order to give a reference for the application of adsorption technology to remove As(V) from water in practice.

The mechanism of arsenic removal by adsorption

Insoluble solid materials with high specific surface area or adsorption groups are chosen as adsorbents, and arsenic in drinking water can be fixed to the adsorbent by physical or chemical process including electrostatic attraction, surface complexation, and ion exchange [20]. The characteristics and main influencing factors of adsorption pathways are summarized in Table 1. The adsorption capability of an adsorbent is an important indicator to evaluate its performance. The effects of As(V) adsorption are influenced by pH, temperature, initial concentration of As(V), contact time and the presence of other competing ions in water [14,21].

Table 1: The characteristics and main influencing factors of different adsorption pathways [13,22-26].

| Adsorption pathway | Force | Main influencing factors |
|--------------------------|----------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------|
| Electrostatic attraction | Van der Waals, chemical bond forces | properties of adsorbent and solution, such as the specific surface area, the number of surface functional groups, pH |
| Surface complexation | the complexation of surface hydroxyl groups or carboxyl groups with metal or metalloid oxide | concentration, pH, ionic strength |
| Ion exchange | concentration gradient, affinity capacity | pH, temperature, the presence of competing ions in water |

Furthermore, with the development of science and technology, a number of advanced techniques and precision instruments can be used for studying the mechanisms of arsenic removal from water by adsorption methods. It is reported that the characteristics of adsorbents can be investigated by means of X-ray diffraction (XRD), scanning electron microscope (SEM), Fourier transform infrared (FTIR), X-ray absorption spectroscopy (XAS), X-ray photoelectron spectroscopy (XPS), the extended X-ray absorption fine structure (EXAFS), etc [23,26-28].

Physical and chemical adsorption

Traditional adsorption materials

Activated carbons: Activated carbon is well-known to be used as adsorbents, which is a kind of carbide made from coconut shell, charcoal, lignin, sawdust, rice husk and other carbonaceous materials by carbonization and activation processes. The strong adsorption capacity is attributed to rich pore structure, huge specific surface area and surface functional groups [13].

Both activated carbon and modified forms can be used as adsorbents to remove As(V) from water. Natale, et al. investigated the effect of As(V) removal using granular activated carbon (GAC) [29], and the results indicated that the

saturated adsorption capacity can reach 2.5 mg/g at pH 2–11.5 and 20°C after 72 h, using 2 g/L GAC, which was of 950 m²/g specific surface area (BET), treating raw water with initial As(V) concentration of 0.5–10 mg/L. This study also indicated that H₂AsO₄⁻ and HAsO₄²⁻ in natural water were more conducive to be removed efficiently under acidic condition (pH=3). Lodeiro, et al. [30] compared adsorption capabilities of four adsorbents: activated carbons prepared from sugar beet pulp (BP) using steam activation and with or without Fe or Mn oxidation (BP-H₂O, BP-H₂O-Fe, BP-H₂O-H₂O₂-Fe and BP-H₂O-MnO₂-Fe). Their BETs reached 821 m²/g, 762 m²/g, 858 m²/g and 741 m²/g, respectively, for treating raw water with initial As(V) concentration of 100–1000 µg/L, using the dosage of 0.05 g/100 mL. The adsorption capacity of these four adsorbents on As(V) were 0.69 mg/g, 2.94 mg/g, 3.25 mg/g and 16.8 mg/g, respectively, calculated according to Langmuir equation. The results showed that it is necessary for the activated carbon to be modified in order to achieve a better adsorption ability.

Minerals: Clay belonging to aluminum silicate minerals contains small amount of iron, magnesium and other elements, resulting in small particles, large specific surface area, good adsorption capability and ion exchange performance [13]. As typical of clay minerals, kaolinite, montmorillonite and illite can effectively adsorb As(V) from water [31]. In addition, natural iron ores (goethite, hematite, etc) and manganese ores (sodium manganese, manganese potassium, etc) also have good adsorption capacities for As(V) removal from water [32].

Chen, et al. [33] took high-quality clay as the main material, and processed into flake, spherical or columnar shape by a series of processes. Its surface was porous, with a BET of 38.19 m²/g and a pore volume of 0.0869 mL/g at 25°C under neutral condition (pH=6.9). According to Langmuir and Freundlich adsorption isotherm equations, the adsorbent can effectively treat raw water with initial concentration of As(V) ranging from 5 to 100 mg/L, and the adsorption capacity was about 4.19 mg/g. Zeolite is a kind of aluminosilicate crystals with regular cage structure [34]. Jimenez-Cedillo, et al. [35] loaded Fe or Fe-Mn onto two natural clinoptilolites (ZC and ZT) with BETs of 5.97 m²/g and 14.68 m²/g, respectively. Four kinds of modified adsorbents (ZCFe, ZTFe, ZCFeMn and ZTFeMn) were prepared, and their BET values were 40.44 m²/g, 38.20 m²/g, 36.40 m²/g and 35.54 m²/g, respectively. According to Langmuir equation, the adsorption capacities of these four modified materials for As(V) removal were about 0.08–0.1 mg/g, specifically, ZTFeMn=ZCFeMn>ZCFe>ZTFe.

Metal based materials: Metal based materials include metals, metal oxides and metal hydroxides [36]. Activated alumina is often used in drinking water treatment due to its large specific surface area and rich pore structure, which is often used for arsenic removal from water [37]. Previous studies reported that the adsorption capacity of As(V) using activated alumina as an adsorbent can be improved under acid conditions or by modification methods [38-40].

Zero valent iron, iron oxides and their hydroxides also have good adsorption performances on As(V). Bang, et al. [41] showed that high dissolved oxygen (4.3–5.5 mg/L) and low pH

value (pH=6) could promote zero valent iron corrosion, and iron hydroxide formed by iron oxidation could adsorb As(V) in water. The removal rate of As(V) reached 99.8% after treating for 9 h, using zero valent iron 1 g/L to treat raw water with initial As(V) concentration of 100 mg/L. Guan, et al. [23] used granular iron hydroxide (GFH) as an adsorbent (BET 206 m²/g, pore volume 0.76 cm³/g, average pore diameter 14.8 nm) to treat raw water with initial As(V) concentration of 5–2000 mg/L under different pH levels of 1–13. The results indicated that the suitable pH changed from weak alkaline (pH 8) to strong acid (pH 2) with the increasing As(V) concentrations. The structure of adsorbent was also analyzed by Fourier transform infrared spectroscopy (FTIR) and extended X-ray absorption fine structure (EXAFS) to explore the adsorption mechanism of As(V).

In addition, the composite iron aluminum hydroxide, manganese oxide, rare earth oxide (such as lanthanum and cerium), zirconium oxide, titanium oxide and tin oxide can also be used for As(V) adsorption from water [13,42].

Industrial and agricultural wastes: A large amount of wastes are often produced in the process of industrial and agricultural production, and these resources can be efficiently reused. With the acceleration of industrialization and the rapid development of coal-fired power industry, the emission of fly ash rises sharply, and the pollution problems caused by fly ash need to be solved effectively. Medina, et al. [43] used fly ash to synthesize zeolite, which can be utilized as an adsorbent to treat As(V)-contaminated water. As(V) concentration decreased from 740 µg/L to less than 10 µg/L, and the removal rate reached 99% under neutral condition after contacting for 5 min. Red mud is a kind of waste produced in the process of alumina production, which needs to be treated properly. Altundoğan, et al. [44] reported that the adsorption capacity of As(V) can be effectively improved after the acid activation treatment. The initial concentration of As(V) in water was 10 mg/L, and the dosage of activated red mud was 20 g/L, resulting in the removal rate of 96.52% and the effluent pH of 7.25 at 25 °C. Pehlivan, et al. [24], made use of lignocellulosic materials extracted from rice husk (RH) to form RH-FeOOH by hydrolysis and impregnation of Fe(III) under alkaline condition. As(V) ions with negative charge in water can be effectively adsorbed on RH-FeOOH with positive charge on the surface via Coulomb interaction under slightly acidic condition, thus, As(V) can be removed from water effectively. The result reported that the removal rate reached 99.6%, by using the dosage of 0.2 g/50 mL to treat initial As(V) concentration of 3 mg/L, at pH 4 and 22°C for 6 h. The adsorbent was regenerated by 0.1 M NaOH solution for 20 h at pH 12–14, and the regeneration rate can reach 90%.

Ion exchange resins: Ion exchange resins are made of insoluble polymer networks with functional groups, and As(V) ions in water are exchanged with functional groups and absorbed on the resins. The synthesis of ion exchange resins can be divided into two steps: preparing polymer skeleton and introducing functional groups into the skeleton [45].

Ligand exchange cellulose adsorbent was prepared by Awual, et al. [15], who loaded Zr (IV) onto the cation exchange

resin (Zr-FCPS) with bifunctional groups (phosphate and sulfonate). The results indicated that the influence of initial As(V) concentrations of 1.115 mg/L can be purified to less than 10 mg/L by adding 0.5 g Zr-FCPS into 1.93 L of As(V)-contaminated water, and 0.1 M NaOH solution can be used as the eluent reagent. An ion exchange/electrodialysis (IXED) flow cell was introduced by Rivero, et al. [46], to remove As(V) from water. A central compartment packed with anion exchange resin, and two anion membranes are placed as its boundary of compartments on the anode side and cathode side, respectively. As(V) anions can be exchanged by OH⁻ anions produced by water dissociation in cathode side, and the transport rate of anions was influenced by several factors, such as the potential profiles, Donnan potential and concentration polarization.

New adsorption materials

Nanomaterials: Nanomaterials have smaller particle size, higher specific surface area and stronger adsorption capacity, compared with the traditional adsorption materials [14,47]. Previous studies have shown that nano-Cu(II), zero-valent iron, crystal TiO₂, granular ZrO₂, ZrO(OH)₂, Al₂O₃, magnetic Fe₂O₃, magnetic bimetallic oxide (MnFe₂O₄ and CoFe₂O₄) have good adsorption performances on As(V) removal from water [14,21,27,48–51].

On this basis, previous studies reported that nanoparticles loaded onto adsorbents can deal with the problems of high price and difficult operation. Ntim and Mitra [52] loaded carbon nanotubes with nano zirconia particles (MWCNT-ZrO₂), with a BET of 152 m²/g, to treat raw water with initial As(V) concentration of 100 µg/L. The contact time was extended from 10 min to 60 min, correspondingly, the removal ratio was increased from 50% to 99%. The results indicated that As(V) can be removed effectively from water, and its concentration was as low as 1 µg/L in effluent. The adsorption capacity was 5000 µg/L calculated by Langmuir and Freundlich adsorption isotherms. Kanematsu, et al. [53] filled the adsorption fixed bed with nano goethite particles (BET 158.1 m²/g), for treating As(V)-contaminated water with initial As(V) concentration of 120 µg/L at pH=7. The empty bed contact time (EBCT) was controlled at 0.328 min, to ensure the discharge to meet the standards of As(V) in drinking water. In addition, silicate was not conducive to be used for As(V) removal because of strong adsorption competition with the existing As(V) ions.

Mesoporous materials: Mesoporous materials is a kind of adsorption material with pore sizes ranging from 2 nm to 50 nm, with large specific surface area, highly ordered pore structure and high adsorption capacity [28]. Patra, et al. [54] showed that mesoporous γ-Al₂O₃ spherical nanoparticles with BET of 497 m²/g had a high affinity for As(V) in water. The dosage of adsorbent was 0.1 g/100 mL, and the contact time was prolonged from 1 h to 6 h, then, the removal rate can be increased from 60% to 80% when treating the water with initial As(V) concentration of 100 µg/L. The structure, surface and optical property of the particles were analyzed by X-ray diffraction, transmission electron microscopy and ultraviolet visible spectroscopy. Pillewan, et al. [55] prepared copper oxide incorporated mesoporous alumina (COIMA) (BET 189.25 m²/g)



by impregnating mesoporous alumina with copper sulfate solution to treat raw water with initial As(V) concentration of 1.0 mg/L for 24 h at 30°C, using the dosage of 0.4 g/L, the calculated adsorption capacity of COIMA for As(V) was 2.017 mg/g according to Langmuir equation.

Biological adsorption

Biological adsorbents include plants, microorganisms and biomaterials, which can adsorb, transform and degrade As(V) in water through a series of physical, chemical and biological processes. Biological adsorbents have promising application prospects due to the advantages of high efficiency, cost effective and less secondary pollution.

Many species of algae in huge quantities are widely distributed in natural water, which have been reported to be associated with adsorption and degradation of pollutants in water. As(V) can be adsorbed on the surface of algae and accumulated in their cells, and then reduced to As(III), which can be further degraded and detoxified by methylation process, to achieve arsenic removal from water [56].

Microbial cell walls are composed of polysaccharides, lipids and proteins, which are rich in binding sites, therefore, they can be used as adsorbents for water treatment [13]. As(V) in water is adsorbed on the surface of microorganism and degraded by a series of metabolic activities. It has been proven that several fungal strains of *Aspergillus* and *Trichoderma*, and other fungi were isolated from soil [57]. *Aspergillus candidus* belonging to a facultative marine fungus also showed good adsorption performance on As(V) removal from water [58].

In addition, previous studies have shown that biomaterials can also adsorb As(V) in water [59]. For example, chitosan converted from crustacean shell by deacetylation process is used as an adsorbent to treat As(V)-contaminated water, which has good adsorption capacity for As(V), due to its high affinity for ions caused by its high molecular chain structure, rich hydroxyl and amino groups [13].

Furthermore, the adsorbents can also be modified by chemical methods to improve the adsorption capacities of As(V). A novel Fe(III)-loaded ligand exchange cotton cellulose adsorbent was prepared by loading Fe(III) onto cotton cellulose (BET 2.23 m²/g, water content 87%) by Zhao, et. al. [60], and this adsorbent was filled into a glass filter column (φ 9.5 × 300 mm). The raw water with initial As(V) concentration of 1 mg/L (coexisting ions SO₄²⁻ 250 mg/L, Cl⁻ 250 mg/L) could be purified until the effluent was less than 10 µg/L, under the conditions of pH=7.1, 25 ° C and the filtration rate of 26 BV/h. The desorption rate of As(V) was higher than 96% when the filter column was regenerated with 1 M NaOH. Boddu, et al. [61] coated chitosan on alumina ceramics to prepare a new composite chitosan biosorbent (BET 125.24 m²/g), and then the biological adsorbent was filled in a filter column (inner diameter 1.1 cm, height 30 cm). The results showed that the raw water with initial As(V) concentration of 101 mg/L can be treated until

there was no As(V) in effluent (downward filtration, filtration rate 2.5 mL/min, pH=4), thus, the adsorption capacity was as high as 96.46 mg/g. It has also indicated that acidic condition was more conducive to the removal of As(V), and 0.1 M NaOH solution can be used as regeneration agent.

Problems and suggestions

As(V) removal from water by adsorption will not produce chemical sludge and concentrated water needed subsequent treatment, compared with treating As(V) using coagulation or membrane technologies. However, the adsorption capacity of an adsorbent is limited, and the effect of arsenic removal is easily affected by the competition of coexisting ions in water, therefore, it is not beneficial to select adsorption technology to treat As(V)-contaminated water with complex ionic composition, and the costs will be highly increased due to the needed extra pretreatment process. Furthermore, the separation of adsorbents from water has been considered as a big challenge for drinking water treatment processes.

In general, the adsorption technology is suitable for treating the water containing As(V) with low concentration and simple composition. It is wise to use different adsorption materials in different practical application directions, and it needs to make an overall comparison comprehensively between the adsorption capability and economic feasibility. The adsorbents reused after regeneration by chemical methods complies with the trend of sustainable development. In the future, to develop environmental friendly materials achieving more efficient performance, explore methods enhancing regeneration capacity, optimize the synthesis and modification methods and try to reduce the overall cost are the development directions, in order to promote this technology to be commercialized. The adsorption performance of the adsorbents can be further improved by modifying the powder adsorbents into granular forms, loading metal ions or bacteria on adsorbents, fixing nano or mesoporous particles on the adsorbents with relatively low price or large volume, adding magnetic components, using symbiont of bacteria and algae as adsorbents, and modifying biological adsorption materials by chemical methods.

Conclusions

Arsenic discharge into water environment should be effectively minimized in productive activities to prevent arsenic pollution. It is necessary to select the technology on the basis of detailed investigation of the water quality of water sources combined with the applicable conditions of various technologies. The adsorption technology is suitable for purifying the water containing As(V) with low concentration and simple composition. It is important to weigh the advantages and disadvantages of each adsorption material for arsenic removal when selecting a feasible material for As(V)-contaminated water treatment. This technology needs to be further explored in search of efficient and environmental friendly adsorption materials, comprehensive utilization of chemical and biological methods, and improvement of regeneration methods, to optimize the synthesis and modification of adsorbents.

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