# REMARK ON MECHANISM OF ARSENIC REMOVAL FROM WATER USING METAL (HYDR) OXIDE NANOPARTICLES

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## ABSTRACT

*Effective removal of arsenic from water and groundwater is crucial to obtain safe drinking water. Many researchers have studied metal (hydr)oxide nanoparticles (NPs) as promising adsorbents for adsorption of arsenic from water due to their high selectivity and efficiency. This study critically reviewed the adsorption mechanisms of As(III) and As(V) species on metal (hydr)oxide NPs during the adsorption process and the perspectives involved in their utility. The study discussed the inner-sphere complexation between arsenic species and active surface metal (hydr)oxide NPs, which is attributed to the high selectivity toward arsenic species over other common ions (such as sulfate and chloride). In addition, the As(III) removal involving an oxidation process using oxidant oxides such as manganese oxide, zero valent iron and titanium oxide for enhancing adsorption performance was provided. This paper provides the readers to deeply understand the role of metal (hydr)oxide NPs on the arsenic removal and to take up further research required for arsenic removal using metal (hydr)oxide adsorbents.*

***Keywords:*** *Arsenic; metal oxide; adsorption; adsorption mechanism; adsorbent.*

## 1. INTRODUCTION

Water pollution, especially arsenic (As) contamination in water and groundwater is a worldwide concern. It has been reported that many countries have been seriously concerned with arsenic pollution in water resources such as Bangladesh, India, Chile, Argentina, South East Asia, China, Mongolia and the United State. Long-term exposure to arsenic can cause many dangerous diseases to humans including cancers (skin, lung, liver and bladder and kidney) and other non-cancer effects (muscular weakness, loss of appetite and nausea) [1, 2]. Due to its harmful properties, the US Environmental Protection Agency (US EPA) has established the guideline for arsenic concentration permitted in drinking water to be lower than 0.01 mg/L [3]. The Eh-pH diagram provides a view of arsenic forms and its possibility of mobilization in the water environment depending on the solution pH and oxidation potential Eh values (Fig. 1) [3, 4]. Positive Eh indicates oxidative condition, whereas negative Eh represents reducing condition. In natural water with pH often from 6 to 9, arsenic is almost predominated in two inorganic forms: arsenate (As(V)) and arsenite (As(III)) under several forms (such as $H_2AsO_4^-$, $HAsO_4^{2-}$ and $H_3AsO_3$). Generally, As(III) mainly occur under anaerobic conditions (e.g. groundwater), whereas As(V) is the major species under aerobic conditions (e.g. surface water) [5]. In order to obtain safe water, which meets the arsenic standard in local communities, it is required to develop relatively simple, inexpensive and effective arsenic removal technologies [6].

Most arsenic removal technologies are classified into four processes: membrane filtration (including RO), coagulation–filtration precipitation (including lime softening), ion exchange, and adsorption. Among them, the adsorption technology has been commonly used due to its simplicity, effective removal and cost effectiveness [3,

8]. Be usually occurring in trace level compared with other coexisting ions in water and groundwater, the removal of arsenic requires selective adsorbents which should have a much greater affinity toward arsenic species than other ions. A variety of polyvalent metal (hydr)oxide NPs have been extensively employed for removal of arsenic species due to their extremely large surface area and high specific functionalities [9]. These adsorbents could be single oxides (such as, Al oxide/hydroxide [10], Fe oxide [11], Zr oxide [12], Cu oxide[13], Mn oxide [14], Ti oxide [15], Sn oxide [16]) and binary oxides (such as, Fe(III)-Mn(IV) [17], Fe(III)-Zr(IV) [18], Fe(III)-Cu(II) [19], Fe(II,III)-Ti(IV) [20], Fe(III)-Sn(IV) [21], and Al(III)-Mn(IV) [22].

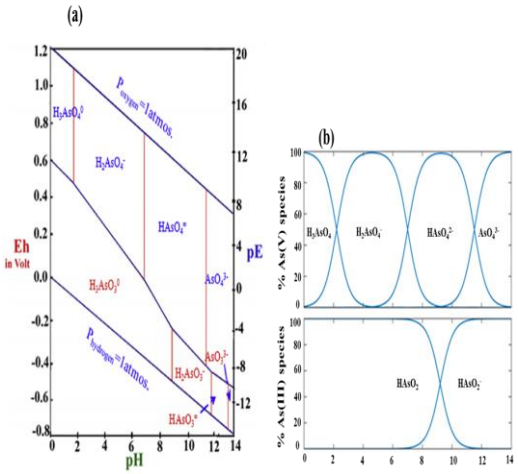


***Figure 1.*** *Species distribution of dissolved arsenic in water with respect to water pH and Eh at °C temperature and 1 atmospheric pressure [7].*

There are some review papers available on arsenic removal using metal oxide, but this paper critically reviewed the mechanism of arsenic adsorption on metal (hydr)oxide particles. Then the challenges of selecting metal (hydr)oxides as adsorbents for removal of arsenic from drinking water were also discussed.

## 2. APPLICABILITY OF VARIOUS METAL (HYDR)OXIDES

Iron oxides have been mostly used because iron is an abundant, inexpensive and innocuous source. Many forms of iron oxide have been considered as promising adsorbents for arsenic and phosphate removal, including hydrous ferric oxide (FeOOH) [11, 23], goethite (α-FeOOH) [24] and hematite (α-$Fe_2O_3$) [25], magnetite ($Fe_3O_4$) [26, 27]. In particular, the use of magnetite iron oxide could allow magnetic separation of adsorbents, which bring advantages in technical design of filtration process [28]. Moreover, the addition of manganese oxide to the composition of adsorbent has shown unique characterization favoring arsenic compared with individual elements. Zhang et al. synthesized the Fe-Mn binary oxide by the co-precipitation method using solutions of iron (II) sulfate and potassium permanganate. This material proposed a high arsenic affinity of iron oxide and an effective oxidation properties of manganese oxide to oxidize As(III) to As(V) [29]. In addition, the incorporated oxides could provide additional active surface sites for adsorbing arsenic species. In a similar way, the material from San et al. showed very effective for both As(III) and As(V) removal and easy regeneration with a simple process [30]. Nevertheless, iron oxides can be dissolved in acidic condition and in contact with organic substances. In addition, the reduction of Fe(III) to Fe(II) by reducing agents can increase their dissolution, which could increase risk of arsenic release from the arsenic-laden to landfill conditions[31]. To compare with iron oxides, zirconium oxide has been recently reported with high stability against pH ranges, organic substances and oxidants [32]. An increasing study of zirconium oxide and its binary oxide has been extensively introduced for high efficient removal of arsenic from water [12, 18].

## 3. MECHANISM OF ARSENIC ADSORPTION

### 3.1 Inner-sphere complexation

Metal oxides are able to selectively adsorb arsenic species on their surface groups in the presence of high concentrations of competing anions such as sulfate, nitrate,

chloride, and bicarbonate. Fig. 2 shows a modelling illustration of the interaction between hydrated metal oxide and arsenic in electrolyte solution, which is mainly taken place on their surface functional groups, −OH groups [33]. The hydrated metal oxides can be viewed as diprotic weak acids that can deprotonate as Eqs. (1) and (2)[34]:

$$\overline{MOH_2^+} \leftrightarrows \overline{MOH} + H^+ \overline{MOH_2^+} \leftrightarrows \overline{MOH} + H^+ \quad (1)$$

$$\overline{MOH} \leftrightarrows \overline{MO^-} + H^+ \overline{MOH} \leftrightarrows \overline{MO^-} + H^+ \quad (2)$$

Surface charge of metal oxides could be positive ($\overline{MOH_2^+}\overline{MOH_2^+}$), neutral ($\overline{MOH}\overline{MOH}$) or negative ($\overline{MO^-}\overline{MO^-}$) which depends on the solution pH. When the pH < $pH_{pzc}$ of the oxides, the protonated form ($\overline{MOH_2^+}\overline{MOH_2^+}$) will be favorable for phosphate or arsenate ion. Arsenic was effectively adsorbed on the hydrated surface of metal oxides through their inner-sphere complexation between arsenic and surface hydroxyl groups [35, 36] (Fig. 2). However, when pH > $pH_{pzc}$, the deprotonated form ($\overline{MO^-}\overline{MO^-}$) of metal oxide surface was created, resulting in decreasing the removal efficiency due to the electrostatic repulsion [37].

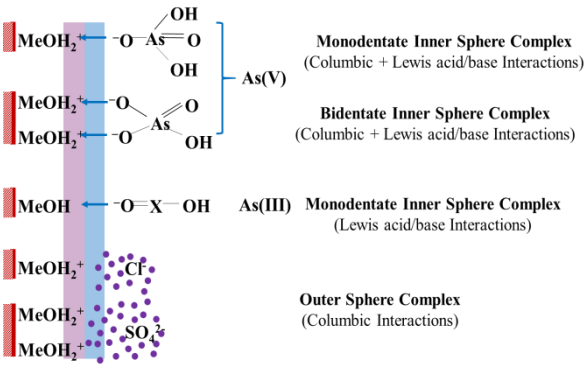


***Figure 2**. Selective interaction between metal oxide surface and arsenic species compared with other common ions in water.*

X-ray photoelectron spectroscopy (XPS) spectra was used to examine the insight adsorption mechanism [38, 39]. The binding energies of O 1s core level was characterized before and after arsenic adsorption on adsorbents. In general, the O 1s spectrum of metal (hydr)oxide consists of three component peaks for metal oxide (M–O), hydroxyl group bonded to metal (M–OH) and adsorbed water ($H_2O$), respectively, with binding energies varying at about 530 eV (Fig. 3). After arsenic adsorption, the relative ratios of M–OH decreased, corresponding to the increase of the relative ratios of M–O peak. This phenomenon indicates that some surface hydroxyl groups were consumed in surface reactions and form As–O after surface reactions. Therefore, the XPS spectra verified that the hydroxyl groups on metal (hydr)oxide reacted with those on arsenic species during adsorption, to form inner-sphere complexes.

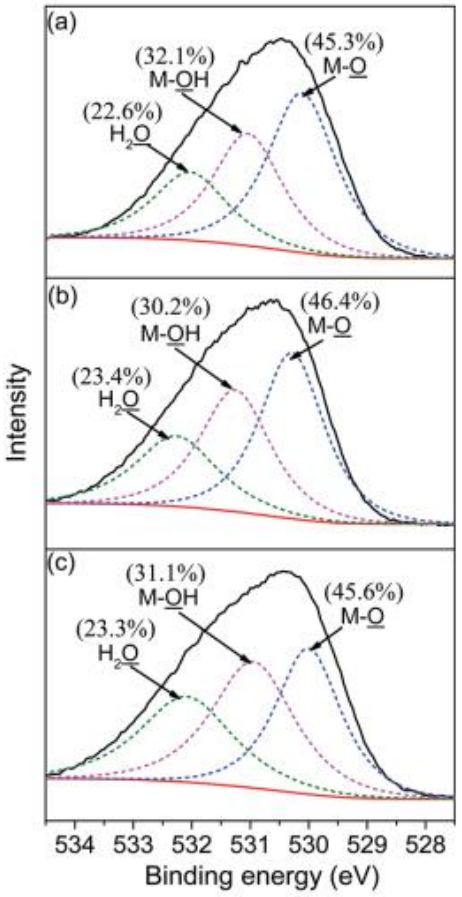


***Figure 3**. As 3d core levels of (a) fresh, (b) As(III)-adsorbed, and (c) As(V)- adsorbed iron oxide NPs [38].*

### 3.2 Oxidation of As(III) to As(V)

To compare with As(V), As(III) removal is more challenging under a wide range of conditions because As(III) generally has a

weaker physicochemical affinity towards most absorbents. In order to remove together with As(V), a pretreatment process is usually introduced for As(III) oxidation, then the resulting As(V) was selectively absorbed onto the surface of solid materials [29, 40-42]. Recently, several manganese-containing oxide have been developed (such as Fe-Mn or Al-Mn binary oxide [22, 43]) for effective removal of both As(III) and As(V) adsorption. During the adsorption process, the oxidation of As(III) to As(V) by Mn(IV) component mainly occurs at the adsorbent's surface. The plausible mechanism for the sorption reaction of As(III) with MNHFO-1 can be described in Eqs. (3)–(6).

$$S_{Fe-Mn}(MnO_2) + As(III) \rightarrow S_{Fe-Mn}(MnO_2)\text{–}As(III) \quad (3)$$

$$S_{Fe-Mn}(MnO_2)\text{–}As(III) + H_2O \rightarrow S_{Fe-Mn}(MnOOH) + As(V) + OH^- \quad (4)$$

$$S_{Fe-Mn}(MnOOH) + As(III) \rightarrow S_{Fe-Mn}(MnOOH)\text{–}As(III) \quad (5)$$

$$S_{Fe-Mn}(MnOOH)\text{–}As(III) + H_2O \rightarrow S_{Fe-Mn} + As(V) + 3OH^- \quad (6)$$

Nano zero-valent iron (ZVI) was reported as one of the effective adsorbent for removal of arsenic including As(III) and As(V) in aqueous solution [44, 45]. The arsenic adsorption on nZVI was well explained by a series of reactions on its surface (Eq. (7)–(13)) [33]. The $Fe_s^0$(nZVI) surface is easily oxidized by dissolved oxygen to form different type(s) of oxide layers on the surface (including $Fe(OH)_2$, $Fe(OH)_3$ and $Fe_3O_4$ as shown in Eqs. (7)–(12)). Iron (hydr)oxide products play an important role as creating active adsorption sites for As(III) and As(V) adsorption with the formation of inner-sphere bi-dentate or mono-dentate complex between arsenic and iron (hydr)oxide [46]. As(III) can be oxidized to form As(V) binding on the adsorbent's surface Eq. (13). The oxidation process of nZVI is more effective in acidic condition but the loss of nZVI mass is increased consequently due to the corrosion.

$$Fe_s^0(nZVI) + O_2 + 2H^+ \rightarrow Fe^{2+} + H_2O_2 \quad (7)$$

$$Fe_s^0(nZVI) + H_2O_2 + 2H^+ \rightarrow Fe^{2+} + 2H_2O \quad (8)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\cdot}OH + OH^- \quad (9)$$

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \quad (10)$$

$$Fe^{2+} + 3OH^- \rightarrow Fe(OH)_3 \quad (11)$$

$$6Fe(OH)_2 + O_2 \rightarrow 2Fe_3O_4 + 6H_2O \quad (12)$$

$$As(III) + {}^{\cdot}OH + H^+ \rightarrow As(V) + H_2O \quad (13)$$

A number of groups have been studied on $TiO_2$ NPs as a promising technology for oxidation of As(III) to As(V) [47]. Under UV light or sun light, $TiO_2$ can work as both functions of adsorbent and photocatalyst. Meanwhile, only adsorption property was observed for $TiO_2$ without light irradiation. It was noticed that the photocatalytic reactivity of $TiO_2$ toward As(III) was affected by the band gap change and the specific surface area (or particle size) of the $TiO_2$ particles [48]. The arsenic species adsorption on $TiO_2$ can be presented in Fig. 4.

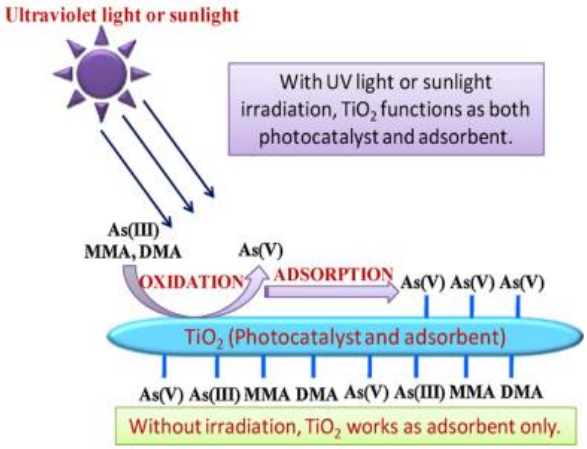


***Figure 4**. Schematic illustration of arsenic removal using $TiO_2$ adsorbent [47].*

As 3d XPS deconvoluted spectra were used to characterize the arsenic species adsorbed on adsorbents. The 3d line could be fitted with separated peak of As(III) or As(V) of specific binding energies of the different chemical states of the As 3d core level for As(III) and As(V)are 44.3–44.5 and 45.2–45.6 eV, respectively (Fig. 5) [33, 49]. As shown in Fig. 5 (a), the oxidation of adsorbed As(III) on the surface of Fe-Mn oxide (OMIM) due to the role of Mn(IV) was obviously provided. This resulted in two

peaks at 44.6 and 45.6 eV assigned for As(III) and As(V), respectively. The much higher ratio of As(V) peak indicates that most of the As(III) was oxidized to As(V), which was attributed to the $MnO_2$ in the adsorbent. Meanwhile, the As(V) adsorption on OMIM showed only one As 3d peak appeared at 45.4 eV for As(V) in Fig. 5(b), attributable to the As–O bonding.

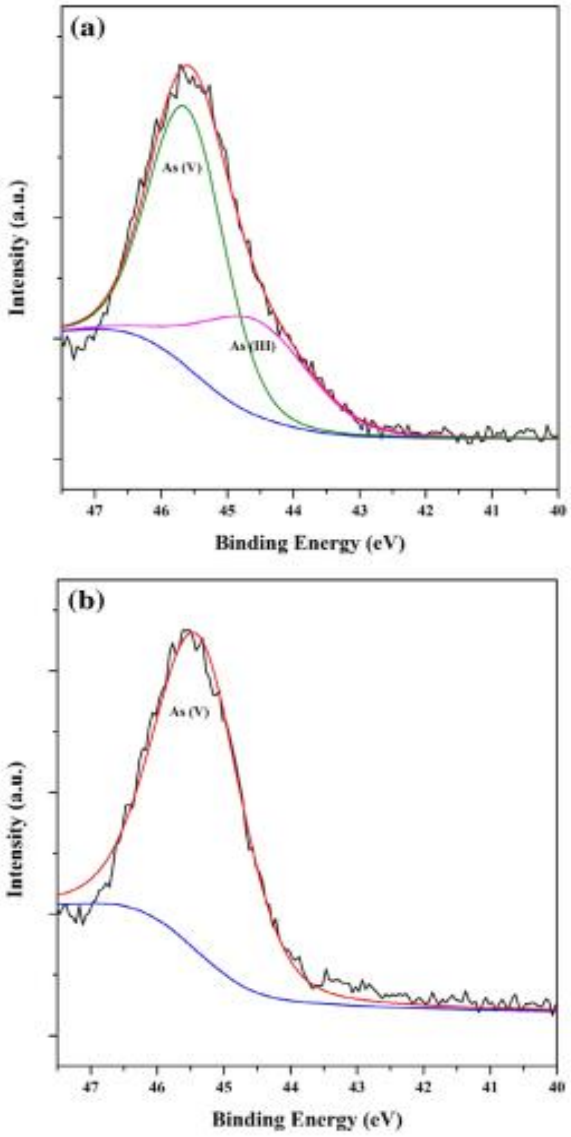


***Figure 5**. As 3d core levels of OMIM after the adsorption of As(III) (a) and As(V) (b) [49].*

## 4. PERSPECTIVES

As fine particles, the use of metal (hydr)oxide NPs can deal with hard problems including aggregation effect, difficult manipulation and separation, and high pressure drop during operation, which could lead to limitation of their field application. In addition, risk of leaking of NPs to the environment causes another environmental concern [31, 50]. Recently, composite materials of incorporating metal oxide NPs within a large-sized or porous support has been recently studied for overcoming technical problems of NPs because it could combine desired properties from a high mechanic host and the outstanding properties of embedded NPs. Thus, it can provide better performance toward arsenic adsorption. However, a significant challenge of controlling the dispersion of NPs in/on the porous host is still considered by researchers. In addition, this technology could limit the content of loaded metal oxide, leading to lower the desired adsorption capacity of the resulting adsorbents. One of the promising composites is a hybrid adsorbent of metal oxide NPs impregnated a strong anion exchange resin. This type of material can maintain the dispersion of metal oxide NPs within the porous host and utilize the advantages of Donnan membrane effect from the polymeric phase, leading to significantly enhance the arsenic adsorption.

## 5. CONCLUSION

Different types of metal (hydr)oxide NPs have been employed for the arsenic removal from aqueous solution. Among them, Fe-based nanomaterials, particularly iron oxide NPs, displayed advantages in arsenic removal due to their environment-friendly, inexpensive and abundant source, especially their high affinity toward arsenic species. The mechanism of metal (hydr)oxide NPs toward both As(III) and As(V) adsorption was attributed to active surface hydroxyl groups. One or more lone pairs of electrons in arsenic species form surface complexation with metal hydroxide sites through a Lewis acid-base interaction (specific adsorption) as inner-sphere complexes. This could provide the selective adsorption toward arsenic species compared to most common ions in natural water. Moreover, some nanomaterials like manganese(IV) oxide, nZVI and $TiO_2$ and their based-nano materials can work as

oxidizing agents for the oxidation of adsorbed As(III) to As(V), and thus enhance the arsenic removal. Recently, the challenges of tiny particle size could be overcome by using composite adsorbents where metal (hydr)oxide NPs were incorporated with a large-size and porous support. Further study is needed to maintain the dispersion state of NPs within the porous host as well as to increase the content of loaded NPs to provide better performances.

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