

Article

Elementary Iodine-Doped Activated Carbon as an Oxidizing Agent for the Treatment of Arsenic-Enriched Drinking Water

Fabio Spaziani ^{1,2,*}, Yuli Natori ¹, Yoshiaki Kinase ¹, Tomohiko Kawakami ¹ and Katsuyoshi Tatenuma ¹

¹ KAKEN Inc., Mito Institute 1044 Hori, Mito, Ibaraki 310-0903, Japan

² ENEA Casaccia, Via Anguillarese 301, 00123 Roma, Italy

* Correspondence: fabio.spaziani@enea.it

Received: 18 July 2019; Accepted: 23 August 2019; Published: 27 August 2019



Abstract: An activated carbon impregnated with elementary iodine (I₂), named IodAC, characterized by oxidation capability, was developed and applied to oxidize arsenite, As(III), to arsenate, As(V), in arsenic-rich waters. Batch and column experiments were conducted to test the oxidation ability of the material. Comparisons with the oxidizing agents usually used in arsenic treatment systems were also conducted. In addition, the material has been tested coupled with an iron-based arsenic sorbent, in order to verify its suitability for the dearsenication of drinking waters. IodAC exhibited a high and lasting oxidation potential, since the column tests executed on water spiked with 50 mg/L of arsenic (100% arsenite) showed that 1 cc of IodAC (30 wt% I₂) can oxidize about 25 mg of As(III) (0.33 mmol) before showing a dwindling in the oxidation ability. Moreover, an improvement of the arsenic sorption capability of the tested sorbent was also proved. The results confirmed that IodAC is suitable for implementation in water dearsenication plants, in place of the commonly used oxidizing agents, such as sodium hypochlorite or potassium permanganate, and in association with arsenic sorbents. In addition, the well-known antibacterial ability of iodine makes IodAC particularly suitable in areas (such developing countries) where the sanitation of water is a critical topic.

Keywords: elementary iodine; activated carbon; arsenic; water purification; drinking water

1. Introduction

Water is essential to life, and ensuring safe drinking water is a priority in any country. The health risks associated with the consumption of unsafe drinking water are related to infectious diseases but also to environmental components such as fluoride, arsenic, lead, cadmium, nitrates and mercury [1]. There is an urgent need to develop suitable water treatment strategies especially for small rural and remote communities in low-income developing countries [2]. However, providing safe drinking water is also a crucial task in the managing of emergency situations (such as earthquake, flooding, etc.). Therefore, even in the so-called “more developed countries”, bacteria and chemical contamination must be constantly monitored and, if detected, removed.

Arsenic is one of the most widespread drinking water-related concerns, due to its toxicity, and because this element occurs naturally in many groundwater sources around the world [3–6].

Soluble arsenic can be found in water in both inorganic and organic forms. Inorganic arsenic compounds (such as those predominantly found in groundwater) are highly toxic, while organic arsenic compounds (mainly found instead in seafood) are less harmful to health. Moreover, inorganic arsenic can occur in several oxidation states (+5, +3, 0 and –3), depending on the physic-chemical characteristics (pH and Eh) of the water, but in natural water systems it is mostly found as trivalent

arsenic (As(III), arsenite) or pentavalent arsenic (As(V), arsenate). Under oxidizing conditions, such as the ones typically found in surface waters, the major species is As(V), mainly present as its oxyanionic forms (H_2AsO_4^- , HAsO_4^{2-}) at the common pH range of natural waters. In contrast, under mildly reducing conditions, such as in groundwater, As(III) is the thermodynamically stable form, which, at the common pH values found in natural waters, occurs as non-ionic form of arsenious acid (H_3AsO_3) [7–9].

The health risks associated with the long-time consumption of arsenic contaminated water are well documented [10,11], and the World Health Organization (WHO), European Union (EU) and the US Environmental Protection Agency (US-EPA) have consequently defined the arsenic standard for drinking water as 10 $\mu\text{g/L}$. The toxicity of inorganic arsenic is related to its aptitude to combine with sulfhydryl groups in proteins and, therefore, to accumulate in the body. In this regard, it is important to notice that arsenite is potentially more toxic since it has a higher affinity for protein than arsenate [12].

The natural occurrence of arsenic in groundwater is mainly linked to the influence of geothermal systems, such as mixing with arsenic-rich deep-rising fluids [13,14] or to the release/mobilization of the element from the minerals in the aquifer [15]. These mechanisms are the result of a complex conjunction of stratigraphy, hydrogeology, and geochemistry, therefore the administration of groundwater in the affected area is a very challenging and difficult task.

Fortunately, several methods for the removal of arsenic from contaminated water have been proposed, including adsorption, coagulation/flocculation, chemical coprecipitation, membrane filtering, and ion exchange. Among these technologies adsorption is distinguished as the most promising one, as it is cost-effective and efficient even at low concentrations. Due to its ease of operation, lower costs and lower amounts of resultant sludge, adsorption is widely used as the preferential method for the removal of arsenic and other heavy metals. Furthermore, there are several types of adsorbents available, such as metal oxides and hydroxide, mineral clay, activated carbon and biochar [16–20].

Owing to the slow redox transformations, the prevailing forms of inorganic arsenic, As(III) and As(V), are present in both reduced and oxidized environments [21]. However, it is important to notice that the most commonly used sorbent materials are more effective on As(V) [22]; indeed, as already noticed, As(III) basically occurs in non-ionic form at the common pH range and therefore is less adsorbed than the As(V) anions on mineral surfaces [12]. Consequently, to maximize the removal performance, a pre-oxidation step is frequently applied. The predominantly used oxidizing agents are liquid chemicals (sodium hypochlorite, potassium permanganate) or gases (ozone, chlorine), and their use implies a rigorous dosage and sometimes a handling-related danger (e.g. chlorine).

In this paper we introduce an innovative “elementary iodine impregnated activated carbon” (IodAC from now on), developed in our laboratories, which shows potential applicability in the field of water treatment, combining in a solid media the disinfectant capability and also the abovementioned oxidation capability useful in the arsenic removal systems.

2. Materials and Methods

2.1. Materials Used for IodAC Preparation

IodAC was manufactured by means of the impregnation of elementary iodine (I_2) on granular activated carbon, through polyiodide wet treatment. The polyiodide chemical precursor was NaI_3 , while the activated carbon was Shirasagi LH2 C-32/60ss supplied by Osaka Gas Chemicals Co., Ltd. (Osaka, Japan).

NaI_3 was prepared from the reaction between NaI and I_2 in aqueous solution. The activated carbon was then added to the NaI_3 solution in order to achieve the impregnation of I_2 . Lastly, the mixture was filtered, washed with distilled water (to remove the unreacted iodine) and dried to yield the final material IodAC.

Various types of IodAC, with different loads of polyiodide, were produced in our laboratory and tested for a wide range of purposes and applications. In this report, for preliminary investigation of water treatment usages, we are discussing IodAC(30) and IodAC(10), which means that 100 g of

IodAC respectively hold 30 g and 10 g of I₂ (the type of IodAC used is specified in each paragraph of the results).

2.2. Materials Used for the Experimental Procedures

The arsenic oxidation capabilities of IodAC were tested on As(III) enriched distilled water and As(III) enriched groundwater. This differentiation was important to compare the IodAC oxidation performance in a laboratory matrix and in a natural matrix. The groundwater was selected and analyzed prior to the enrichment, in order to assure the absence of preexisting arsenic.

Sodium meta-arsenite (NaAsO₂, assay ≥ 90%) from Sigma-Aldrich (St. Louis, MO, USA) was used as the source of As(III). It was dissolved in distilled water, to obtain a 1000 mg/L As(III) stock solution, and the final pH was adjusted to 7 using hydrochloric acid (HCl, reagent grade, 37%) from Sigma-Aldrich (St. Louis, MO, USA). Small amounts of the stock solution were then diluted, using distilled water or the abovementioned arsenic-free groundwater, to prepare the As(III) enriched solutions.

In addition, to compare IodAC with the oxidizing agents commonly employed for water treatment, sodium hypochlorite (NaClO, 5 g/L) and potassium permanganate (KMnO₄, 0.02 mol/L) were also used. The chemicals were reagent grade, purchased from Kanto Chemical Co., Inc. (Tokyo, Japan). Appropriate dosages, calculated to fulfill the stoichiometric oxidant demand required by the tests, were added to the experiment solutions (more details in the results section).

All the tests were conducted at room temperature and the experimental solutions had initial conditions of T = 20 °C and pH = 6.5.

2.3. Characterizations Techniques

Direct analysis of arsenate content, As(V), was performed by ion chromatography, using a Dionex-DX-120 (Sunnyvale, CA, USA) equipped with an AS12A analytical column. An ICP-MS, ELAN-DRC-II from Perkin-Elmer (Waltham, MA, USA), was used to analyze the total arsenic (such as to check the initial amount of As(III) in the stock solution and in the experimental solutions before the experiments).

SEM-EDS, JSM-6510 from JEOL Ltd. (Tokyo, Japan), was used to investigate the morphological features of IodAC and to confirm the iodine assimilation on the activated carbons.

2.4. Arsenic Oxidation: Batch Experiments

Amounts of 0.1 cc and 0.2 cc of IodAC were added to 100 ml of 50 mg/L As(III) solutions (prepared with distilled water or groundwater) and shaken for 4 hours in polypropylene containers. 1 ml aliquots were sampled after 10 minutes, 1 hour, 3 hours and 4 hours. Each sample was filtered through a 0.45 µm syringe filter and analyzed for total arsenic (ICP-MS) and arsenate (ion chromatography).

A similar batch protocol was applied also for tests aimed to compare the IodAC oxidation ability with some commonly used oxidizing agents.

2.5. Arsenic Oxidation: Rapid Small-Scale Column Experiments

A 5 ml syringe, with a diameter of 1.2 cm, was packed with 3 cc of IodAC, to create a 2.5 cm long packed column. A cotton filter was placed at the end of the column, to avoid loss of IodAC. The 50 mg/L As(III) solution was continuously flowed in the column, from the top to bottom, using a peristaltic pump set at 10 ml/min. The solution passed in the column, exiting at the bottom nozzle, and was collected in portions of 150 ml for periodic pH monitoring. Moreover, after completing each 150 ml recovery, the following 1 ml of solution exiting from the column was sampled from the nozzle in a plastic vial for arsenic chemical analysis. The test was discontinued when the oxidation efficiency on the 50 mg/L As(III) solution felt lower than 90% (meaning an arsenate concentration amount lower than 45 mg/L).

2.6. Combined Use with Arsenic Sorbent

Based on the results of the oxidation experiments, in order to test the applicability in a full “arsenic treatment system”, that is a system able both to oxidize arsenite and to remove arsenic, we also executed batch and column tests using IodAC in combination with iron-impregnated activated carbon (FeAC). The preparation of FeAC was conducted using $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, following a method based on impregnation by iron-salt evaporation similar to the one illustrated by Chen et al. (2007) [23]. The batch experiment compared the kinetics of arsenic adsorption from an arsenite contaminated groundwater, with or without preliminary oxidation by IodAC. The column experiments, instead, tested the real-time synergy of the two mechanisms through a “sequential column experiment”, where arsenite contaminated groundwater was first flowed in a column filled with IodAC and then in a column filled with FeAC.

2.7. Influence of Coexisting Anions on Oxidation Ability

The effects of coexisting anions were studied using 50 mg/L As(III) solutions, each of them spiked with 100 mg/L of one of the following anions: Cl^- , NO_3^- , SO_4^{2-} , H_2PO_4^- . An additional sample, used as reference, was characterized by the same amount of As(III) but without any coexisting anion inside. After the addition of IodAC, the solutions were agitated for 4 hours, filtered through a 0.45 μm syringe filter and used for arsenate estimation.

2.8. Statistical Analyses of Data

All tests were conducted in triplicate ($n = 3$). All data given in text and figures are expressed as the mean of the results; the error bars in figures are expressed as standard deviation. All statistical significance was noted at $\alpha = 0.05$.

3. Results

3.1. IodAC Characterization

Figure 1 shows the SEM images of IodAC. The production process did not affect the surface of the activated carbon, and no superficial differences were observed comparing the IodAC and the activated carbon before the impregnation process. The EDS results, showed the presence of iodine on the granules, confirming the success of the impregnation.

Other physical and chemical characteristics of IodAC are summarized in Table 1.

Table 1. Main characteristics of IodAC.

| Properties | IodAC |
|-----------------------|--|
| Color, form | Black, granular |
| Granular size | 0.25–50.5 mm |
| Bulk density | 0.50–0.56 g/cc |
| Element composition | Carbon (C), Iodine (I) |
| Heat-resistance | 184 °C (I_2 gradually volatile at higher temperature) |
| Weight loss by drying | $\leq 5.0\%$ |
| pH | 4.0–6.0 (Measured by pressing the electrode against the IodAC soaked in water) |
| Chemical durability | Strong in acid |
| Adsorption capacity | Weak in high concentrated alkaline, alcohol, org. solvents, reductants |
| Corrosive | I_2 30 g/AC 100 g (for IodAC(30)) |
| Solubility in water | Low < 0.02 mmpy (SUS316 in seawater) |
| BET surface area | Iodide (I^-) < 0.05%, elementary iodine (I_2) < 0.05% |
| Pore volume and size | 1620 m^2/g |
| Ash | Pore volume of 0.6 mL/g, <2 μm pore; pore volume of 0.75 mL/g, <30 μm pore ≤ 2.0 |

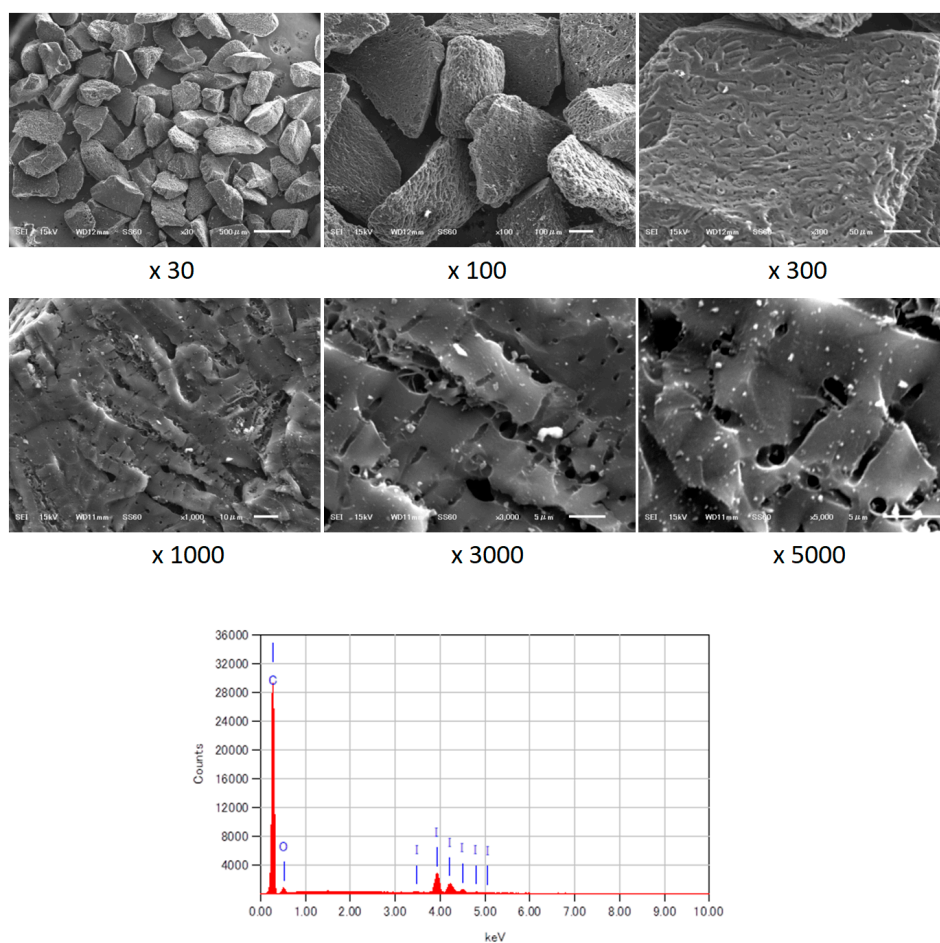


Figure 1. SEM images (various magnifications) and EDS elemental analysis of elementary iodine impregnated activated carbon (IodAC).

3.2. Batch Test Results

The batch tests, expressing the kinetics of arsenic oxidation by IodAC, were executed on 100 ml of 50 mg/L As(III) (corresponding to 5 mg of As(III) in the solution). IodAC(30) was used.

Preliminary tests were performed in distilled water in order to understand the oxidation capability of the material. The results showed that 0.1 cc of IodAC can achieve the oxidation of up to 2.5–2.9 mg of arsenic (around 50–58% of the total amount in solution) and that the process is almost completed within 3 hours. Using the amount of 0.2 cc, all the As(III) in the solution (5 mg) was oxidized (Figure 2).

Taking into account these results, new batch tests were performed, using 0.1 cc and 0.2 cc of IodAC, both in distilled water and in natural groundwater. The results showed that IodAC was also efficient on arsenite oxidation in a natural aqueous matrix.

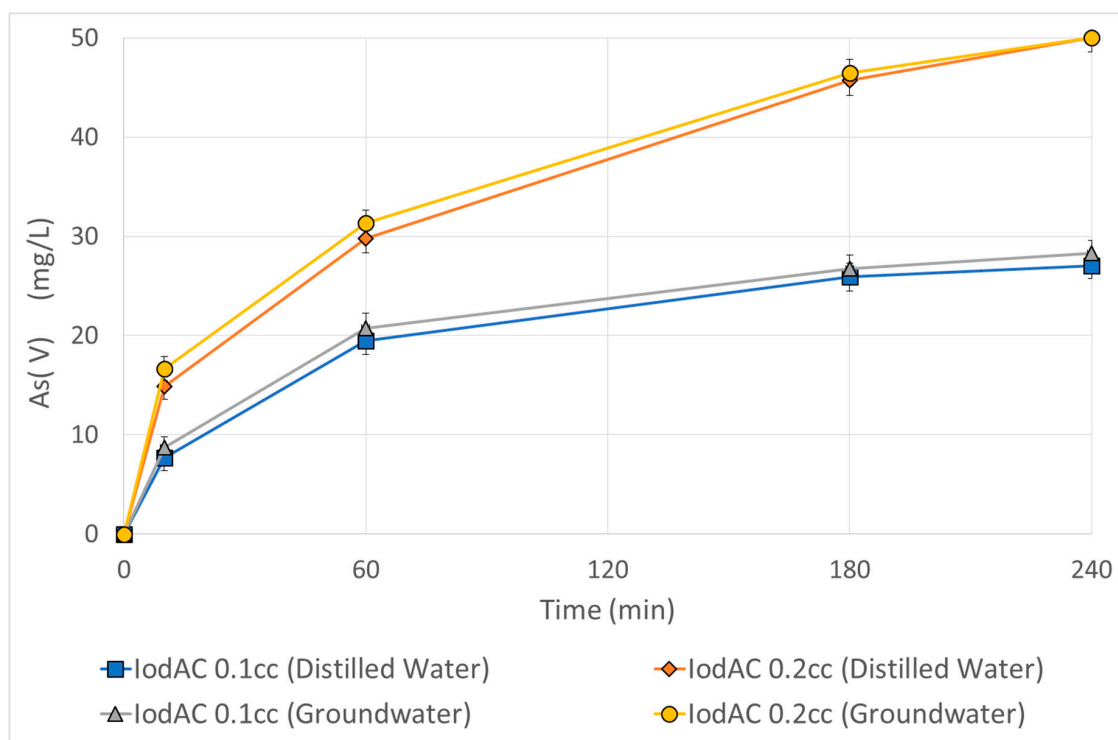


Figure 2. Kinetics of the arsenite (50 mg/L) oxidation batch test in distilled water and groundwater using IodAC(30).

3.3. Comparison with Common Oxidizing Agents

It is difficult to discuss a clear comparison between IodAC and the oxidizing agents commonly used in water treatments, since they have a different state: solid (IodAC), liquid (NaClO , KMnO_4) and gas (O_3). The consequent solid-liquid interaction between the activated carbon and the arsenic solution is affected by various factors (such as surface area), while aqueous ions in a liquid-liquid interaction tend to react faster. Therefore, it was not a surprise to notice that sodium hypochlorite and potassium permanganate showed a faster, almost instant, oxidation.

The experiments were conducted again on 50 mg/L arsenic (100% As(III)) solution (distilled water). The dose of each oxidizing agent used in the experiments, taken directly from the stock solution at the concentration mentioned in the Materials and Methods section, were calculated to theoretically achieve the oxidation of 50% of the As(III) (for the comparison with 0.1 cc of IodAC(30)), and 100% of the As(III) (for the comparison with 0.2 cc of IodAC(30)). The kinetics of arsenite oxidation are summarized in Figure 3.

The slower oxidation process of IodAC, however, is balanced by the long-term oxidation capability of the material, which will be exposed in the column experiments section. Also, the chemical hazard related to the use of concentrated liquid oxidants causes many difficulties in areas such as developing countries where specialized technical staff is rare. Indeed, a liquid-phase or a gas-phase oxidant (such as O_3 or Cl_2) needs to be regularly added, exactly at the appropriate dosage, using a dosing device; moreover, since all the above mentioned oxidants are potentially toxic and harmful, special precautions must be observed in all the handling operations. Therefore, any malfunction of the dosing device can produce an overdosage of toxic chemicals in the water.

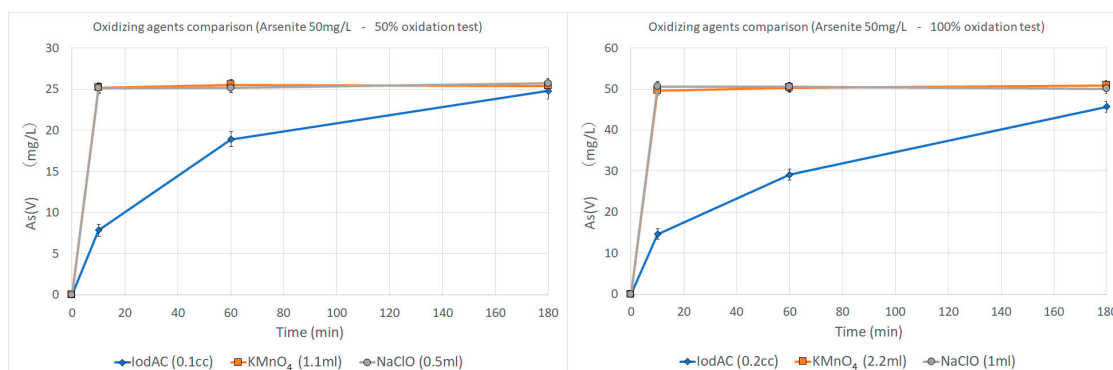


Figure 3. Comparison of the progress of oxidation by IodAC(30) and by the commonly used oxidizing agents.

3.4. Column Experiments Results

Our column experiments can be considered a “stress test”, since the amount of arsenite (50 mg/L As(III)) was considerably higher than the concentrations usually found in natural waters. Such high concentration was also used in order to not excessively protract the experiment time.

In the distilled water based experiment, and using IodAC(30), an oxidation efficiency on As(III) greater than 90% was measured up to 1.5 L (resulting in a total of about 75 mg of As(III) oxidized, and in an oxidizing capability of about 25 mg of As(III) for 1 cc of IodAC). After that, the oxidation efficiency gradually decreased and fell lower than 50% at around 2.5 L (Figure 4a).

Groundwater affected by arsenic pollution is usually characterized by an arsenic concentration in the range of ppb and shows a percentage of As(III) lower than the 100% used in our experiments. As an example, the data concerning the groundwater of central Italy shows a mean arsenic concentration of 23 $\mu\text{g/L}$ [5]; therefore, assuming a hypothetical water with a similar arsenic concentration (20 $\mu\text{g/L}$) and a 100% presence of As(III), 1 cc of IodAC(30) could theoretically oxidize about 1250 liters of such water. According to the available world data [24], even in the countries most vulnerable to arsenic occurrence in groundwater, including the regions where the incidence of reducing conditions boost the portion of As(III), the concentration of the element is still much lower than the amount used in our experiments.

No significant differences were observed when the experiment was repeated using the arsenite solution prepared with real groundwater, resulting in a very similar progress of the oxidation efficiency.

Additional similar column test was conducted on IodAC(10), to verify a proportional relation between the iodine load and oxidation capability. The amount of material used to fill the column was, also in this case, 3 cc, but the concentration of the arsenite solution was set to 25 mg/L, instead of the 50 mg/L used in the IodAC(30) experiments. The lower arsenite concentration was adopted in order to avoid an excessive rapid decrease in the oxidation power. The results showed a very good proportional comparison with IodAC(30), since the oxidizing power of IodAC(10) fell below the 90% after flowing around 900 ml of arsenite solution (Figure 4b). It corresponds to about 7.5 mg of arsenic oxidation capability each 1 cc of material (close to the expected 1/3 of the result attained with IodAC(30)).

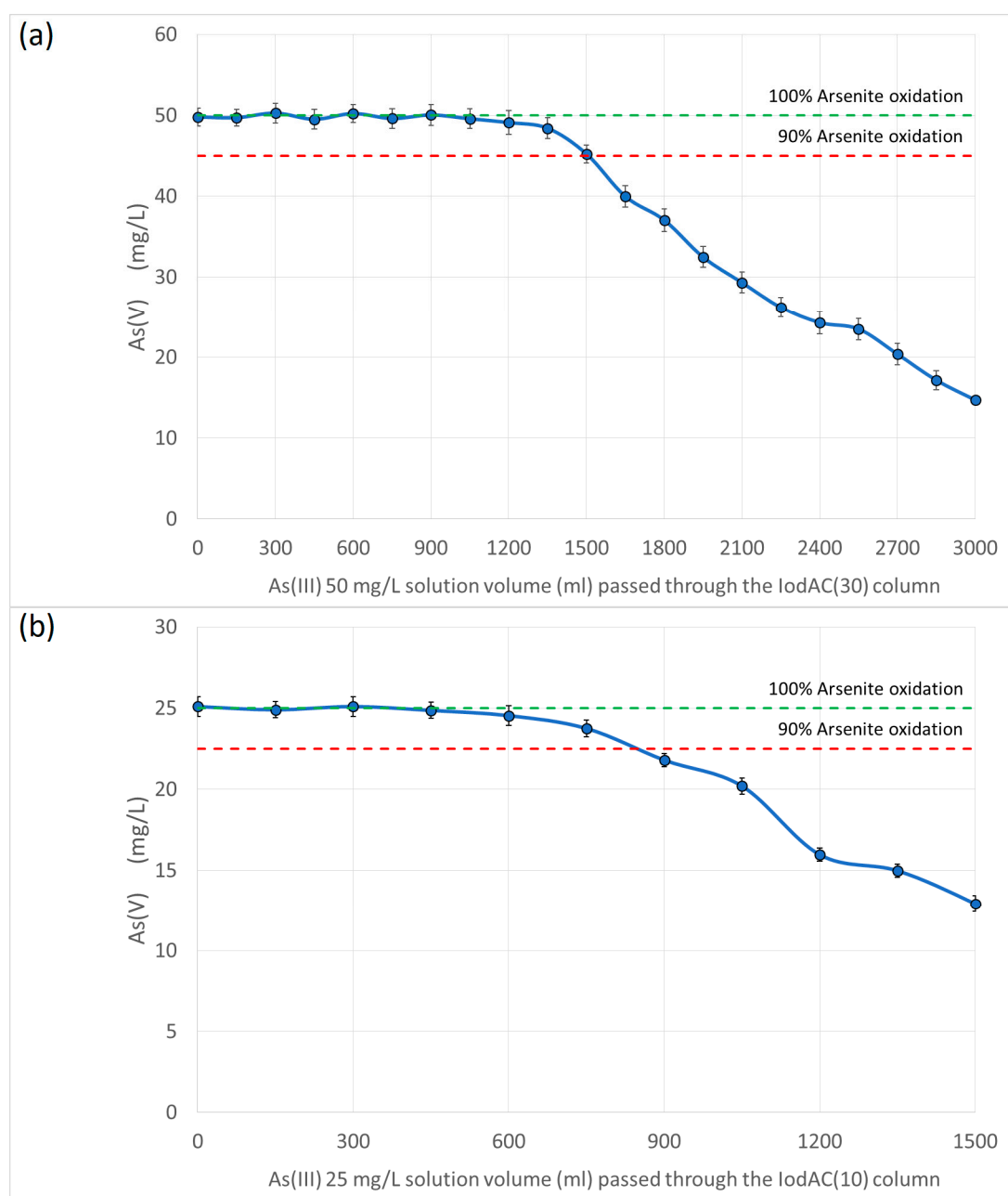


Figure 4. As(III) oxidation rate performance in column study: (a) Arsenate concentration in the output water using 3 cc of IodAC(30); (b) arsenate concentration in the output water using 3 cc of IodAC(10).

3.5. Integration of IodAC in Arsenic Adsorption System

As noticed in the introduction, the common arsenic removal techniques show better performances on arsenate. Therefore, it is important to check the effective contribution of IodAC in the improvement of arsenic removal.

A batch procedure was used as the first approach to test the integration of arsenite oxidation with arsenic adsorption. The batch experiment was carried out with conditions similar of the ones conducted to check the kinetics of arsenite oxidation. Amounts of 0.2 cc of IodAC(30) and 0.2 cc of FeAC were introduced in 100 ml of groundwater (naturally free of arsenic) spiked with arsenite (50 mg/L) and mixed for 4 hours. The quantity of IodAC(30) was decided on because the oxidation experiments revealed that such an amount allowed the complete oxidation of arsenite in 4 hours.

Aliquots of 1 ml were sampled after 10 minutes, 1 hour, 3 hours and 4 hours, filtered through a 0.45 μm syringe filter and analyzed for total arsenic. The same experiment was carried out introducing only FeAC in the arsenite spiked groundwater. The analysis results allowed calculation of the percentage of arsenic adsorption in each sampling step. The oxidation of IodAC sensibly improved the efficiency of arsenic adsorption of FeAC (Figure 5).

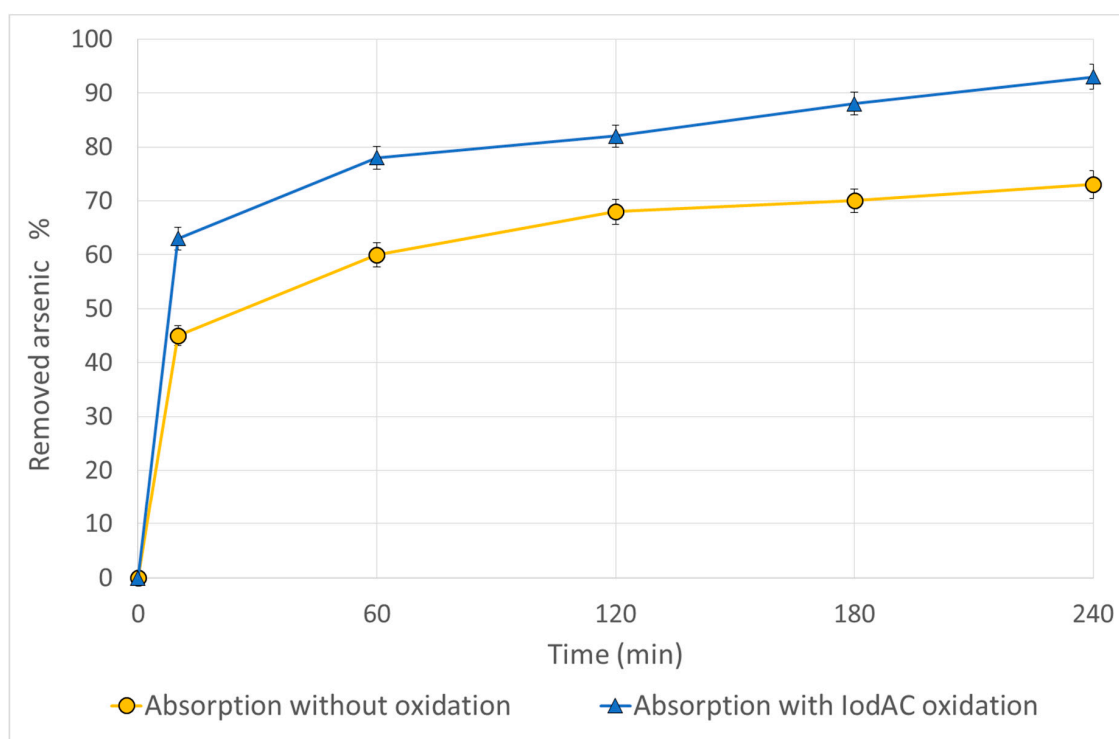


Figure 5. Comparison of the kinetic of arsenic adsorption on iron-impregnated activated carbon, with or without the use of IodAC.

As final test, a real-time column experiment was executed according to the scheme in Figure 6. A special “arsenic speciation cartridge”, purchased from MetalSoft Center (Highland Park, NJ, USA), was used to check the oxidation of arsenite to arsenate (by IodAC(30)), while a column filled with FeAC was used for arsenic adsorption. The speciation cartridge is able to remove the only arsenate and does not adsorb arsenite. Therefore, the difference between the total arsenic concentrations in the starting solution water and in the solution exiting from the cartridge, reveal the residual arsenite concentration (that is expected to be zero due to the oxidation capability of IodAC). A valve, installed on the output line of the IodAC column, allowed switching the routing of the outgoing solution in the FeAC column or in the speciation cartridge. The valve was set to continuously send the output of the IodAC column in the FeAC column; every 150 ml pumped from the starting solution and flowed in the IodAC and FeAC column, an aliquot (1 ml) was collected from the output of the FeAC columns and the valve was switched in order to collect another aliquot from the output of the speciation cartridge (the first 5 ml discarded for washing and the next 1 ml collected for analysis). Then, the valve was switched again to restore the flow in the FeAC column. The tested water was, again, the natural groundwater spiked with 100 $\mu\text{g/L}$ of As(III). The starting concentration of arsenite was set to 100 $\mu\text{g/L}$ since, according to the specifications of the maker, the speciation cartridge is designed to cope with concentrations of arsenate <500 $\mu\text{g/L}$. The results showed a great efficiency in real-time arsenic removal, and demonstrated the synergy of the oxidation and adsorption mechanisms: The oxidation of arsenite by IodAC was fully accomplished again (arsenite <0.1 $\mu\text{g/L}$ in all the steps), and the residual concentration of arsenic from the FeAC output was always lower than 1 $\mu\text{g/L}$ (ICP-MS analysis). The

test was discontinued after the completing of ten samplings, since the aim of the experiment was not to check the lasting of the removal capacity of the FeAC but only to confirm the attainment of the process and the applicability of IodAC together with an arsenic sorbent.

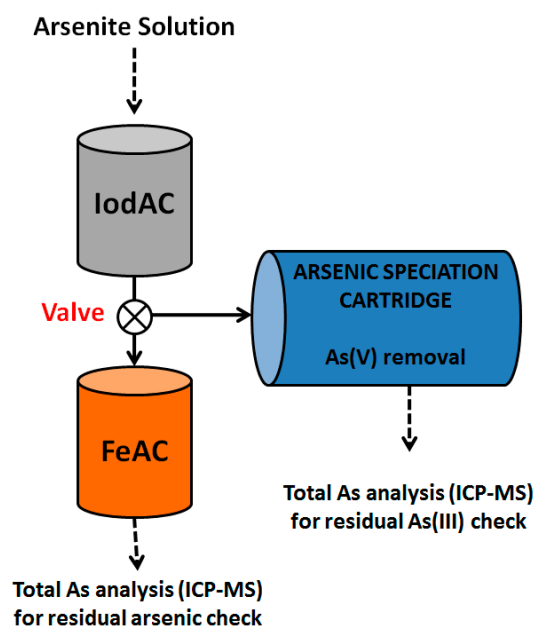


Figure 6. Scheme of the combined (oxidation and arsenic adsorption) column experiment.

3.6. Effect of Coexisting Anions

The chemical composition of natural water is the result of the dissolution of several salts. Therefore, many anions (such as Cl^- and SO_4^{2-}) coexist in water with the arsenite oxyanions. Due to the similar anionic nature, it is important to test the effect of such coexisting anions on the IodAC system and to check any lowering on the arsenite oxidation power. As an example, from the point of view of arsenic adsorption, the effect of coexisting anions on the arsenite removal performances of many sorbents is well documented [25,26].

The test was conducted using 0.1 cc of IodAC(30) in all the solutions described in the related paragraph of the Materials and Methods section. After the completing of the 4 hours agitation, the results, expressed as the percentage comparison of the As(V) concentration in the spiked solutions and in the reference solution, showed that no significant or marginal variations in arsenite oxidation efficiency occurred (Figure 7).

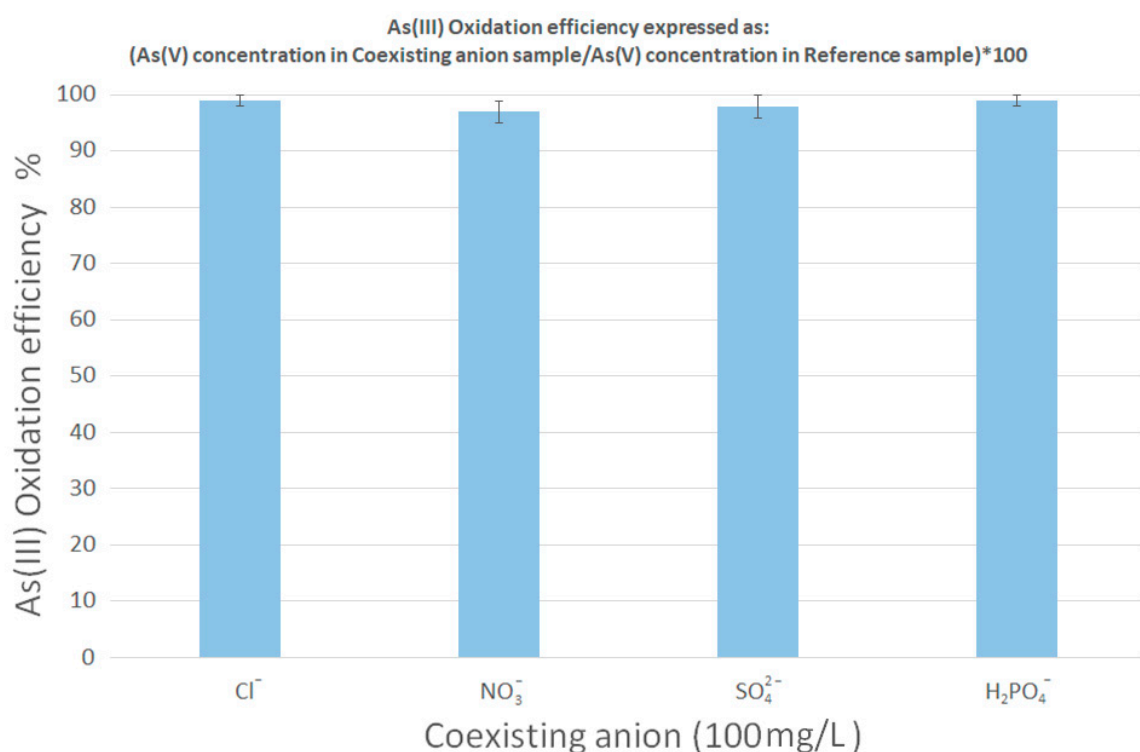
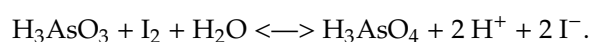


Figure 7. As(III) oxidation efficiency in presence of coexisting anions.

3.7. Mechanism of Arsenic Oxidation

The chemical mechanism related to arsenite oxidation is described in the following equation:



Therefore, iodide release and acidification must be expected as a consequence of using IodAC. Indeed, we noticed an acidification of the water during the above mentioned experiments. During the column tests with IodAC(30), the pH of the output solution sampled in the various steps was 4.36 ± 0.21 . However, it must be noted that the huge amount of arsenite, in the order of tens of mg/L, that triggered the reaction, is unlikely to be found in real groundwater. Under natural amounts of arsenite occurrence, in the order of $\mu\text{g/L}$, the acidification and release of iodide are expected to be insignificant. According to the column tests conducted on 100 $\mu\text{g/L}$ arsenite spiked groundwater (the IodAC + FeAC experiments), the pH variation was negligible ($\pm 3\%$ from the starting value of pH = 6.5) and the concentration of iodine in the output water was below 1 $\mu\text{g/L}$ (ICP-MS determination).

Indeed, the release of iodine can be easily controlled using activated carbon, such as the FeAC used in our experiments (that adsorbed both arsenic and iodine) or also using a final stage of filtration/purification through standard activated carbon. An indicative scheme of such type of system could be the one proposed in Figure 8, which includes IodAC for water sanitization and arsenite oxidation, an arsenic adsorbing media, and a final treatment such as a “guard column” made of activated carbon (to trap residual iodine) and also a neutralizing filter (if pH adjustment is required). The abovementioned components could be integrated, as example, in a multilayer cartridge for a rapid and onsite water purification.

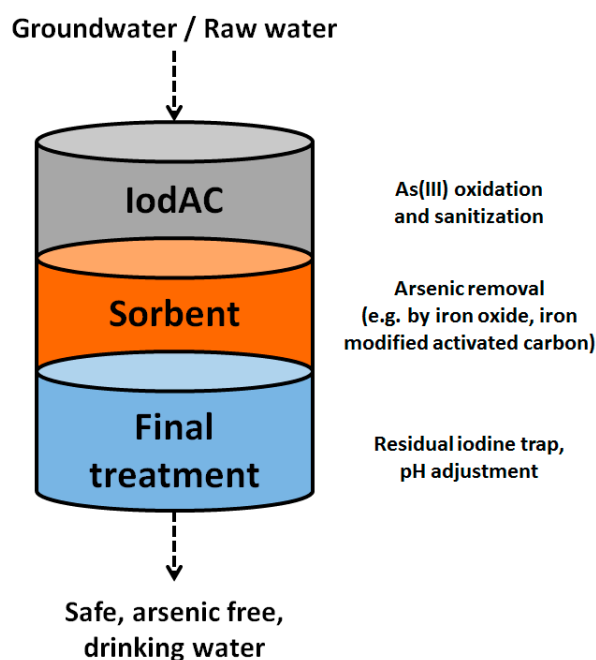


Figure 8. Model for the application of IodAC in an integrated, multipurpose, water treatment system or cartridge.

4. Conclusions

We demonstrated the applicability of IodAC in the oxidation of arsenite in groundwater. The results of the column tests showed that the two types of IodAC tested, IodAC(30) and IodAC(10), have an oxidation efficiency of over 25 mg of As(III) and over 7.5 mg of As(III) for 1 cc of material. Thus, IodAC can enhance the efficiency of arsenic adsorbing media such as iron oxide or iron-modified activated carbons.

Moreover, the results show that the main advantages of using IodAC in place of the oxidants commonly used for arsenite oxidation (such as NaClO , KMnO_4 , Cl_2 , O_3) are the long-lasting oxidizing power, the solid-state and the absence of direct chemical hazards. Such characteristics allow a simpler design of the water treatment system (no need for a dosing device for the oxidant), and easy and safe transport and handling.

Through additional experiments not exposed in this paper, we also verified the disinfectant property of IodAC related to the presence of iodine. Tea-bag like packs filled with IodAC were placed in the oxygen filter of three fish tanks, with two goldfish inside, so that the water circulating in the filter passed through the IodAC packs. A similar fish tank, without the IodAC pack, was used as a reference. After several days the biofouling appeared on the walls of the reference fish tank but not in the ones equipped with IodAC packs. Detailed results concerning these experiments will be discussed in another paper.

In conclusion, IodAC could be used as part of an integrated and multipurpose system for water treatment, such as in small or home treatment units in the developing areas of the world, aimed to achieve both the water sanitization and the arsenic-decontamination.

Author Contributions: Conceptualization, K.T.; Investigation, F.S.; Project administration, K.T.; Supervision, Y.N., Y.K., T.K. and K.T.; Writing—original draft, F.S.; Writing—review & editing, F.S.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Edokpayi, J.; Rogawski, E.; Kahler, D.; Hill, C.; Reynolds, C.; Nyathi, E.; Smith, J.; Odiyo, J.; Samie, A.; Bessong, P.; et al. Challenges to sustainable safe drinking water: A case study of water quality and use across seasons in rural communities in Limpopo province, South Africa. *Water* **2018**, *10*, 159. [[CrossRef](#)] [[PubMed](#)]
2. Huang, J.; Huang, G.; An, C.; He, Y.; Yao, Y.; Zhang, P.; Shen, J. Performance of ceramic disk filter coated with nano ZnO for removing Escherichia Coli from water in small rural and remote communities of developing regions. *Environ Pollut.* **2018**, *238*, 52–62. [[CrossRef](#)] [[PubMed](#)]
3. Schreiber, M.; Simo, J.; Freiberg, P. Stratigraphic and geochemical controls on naturally occurring arsenic in groundwater, eastern Wisconsin, USA. *Hydrogeol. J.* **2000**, *8*, 161–176. [[CrossRef](#)]
4. Rahman, M.M.; Sengupta, M.K.; Ahamed, S.; Chowdhury, U.K.; Lodh, D.; Hossain, A.; Das, B.; Roy, N.; Saha, K.C.; Palit, S.K.; et al. Arsenic contamination of groundwater and its health impact on residents in a village in West Bengal, India. *Bull. World Health Organ.* **2005**, *83*, 49–57. [[PubMed](#)]
5. Angelone, M.; Cremisini, C.; Piscopo, V.; Proposito, M.; Spaziani, F. Influence of hydrostratigraphy and structural setting on the arsenic occurrence in groundwater of the Cimino-Vico volcanic area (central Italy). *Hydrogeol. J.* **2009**, *17*, 901–914. [[CrossRef](#)]
6. Piscopo, V.; Armiento, A.; Baiocchi, A.; Mazzuoli, M.; Nardi, E.; Piacentini, S.M.; Proposito, M.; Spaziani, F. Role of high-elevation groundwater flows in the hydrogeology of the Cimino volcano (central Italy) and possibilities to capture drinking water in a geogenically contaminated environment. *Hydrogeol. J.* **2018**, *26*, 1027–1045. [[CrossRef](#)]
7. Cullen, W.R.; Reimer, K.J. Arsenic speciation in the environment. *Chem. Rev.* **1989**, *89*, 713–764. [[CrossRef](#)]
8. Yao, S.; Liu, Z.; Shi, Z. Arsenic removal from aqueous solutions by adsorption onto iron oxide/activated carbon magnetic composite. *J. Environ. Health Sci. Eng.* **2014**, *12*, 58. [[CrossRef](#)]
9. Feenstra, L.; Van Erkel, J.; Vasak, L. *Arsenic in Groundwater: Overview and Evaluation of Removal Methods*; IGRAC Report nr. SP 2007-2; International Groundwater Resources Assessment Centre: Utrecht, The Netherlands, 2007.
10. Tondel, M.; Rahman, M.; Magnuson, A.; Chowdhury, I.A.; Faruquee, M.H.; Ahmad, S.A. The relationship of arsenic levels in drinking water and the prevalence rate of skin lesions in Bangladesh. *Environ. Health Perspect.* **1999**, *107*, 727–729. [[CrossRef](#)]
11. Hughes, M.F. Arsenic toxicity and potential mechanisms of action. *Toxicol. Lett.* **2002**, *133*, 1–16. [[CrossRef](#)]
12. Payne, K.B.; Abdel-Fattah, T.M. Adsorption of Arsenate and Arsenite by Iron-Treated Activated Carbon and Zeolites: Effects of pH, Temperature, and Ionic Strength. *J. Environ. Sci. Health* **2005**, *40*, 723–749. [[CrossRef](#)]
13. Ballantyne, J.M.; Moore, J.N. Arsenic geochemistry in geothermal systems. *Geochim. Cosmo-Chim. Acta* **2008**, *52*, 475–483. [[CrossRef](#)]
14. Webster, J.G.; Nordstrom, D.K. Geothermal Arsenic. In *Arsenic in Ground Water, Chapter: 4*; Weclh, A.H., Stollenwerk, K.G., Eds.; Kluwer Academic Publishers: Norwell, MA, USA, 2003; pp. 101–125.
15. Smedley, P.L.; Kinniburgh, D.G. A review of the source, behaviour and distribution of arsenic in natural waters. *Appl. Geochem.* **2002**, *17*, 517–568. [[CrossRef](#)]
16. Liu, R.X.; Wang, Y.X.; Tang, H.X. Removal of Arsenate by a New Type of Ion Exchange Fiber. *Environ. Sci.* **2002**, *23*, 88–91.
17. Gholami, M.M.; Mokhtari, M.A.; Aameri, A.; Alizadeh Fard, M.R. Application of Reverse Osmosis Technology for Arsenic Removal from Drinking Water. *Desalination* **2006**, *200*, 725–727. [[CrossRef](#)]
18. Mohan, D.; Pittman, C.U. Arsenic Removal from Water/Wastewater Using Adsorbents: A Critical Review. *J. Hazard. Mater.* **2007**, *142*, 1–53. [[CrossRef](#)]
19. Shahrin, S.; Lau, W.J.; Goh, P.S.; Jaafar, J.; Ismail, A. Adsorptive removal of As(V) ions from water using Graphene Oxide-Manganese Ferrite (GMF) and Titania Nanotube-Manganese Ferrite (TMF) hybrid nanomaterials. *Chem. Eng. Technol.* **2018**, *41*, 2250–2258. [[CrossRef](#)]
20. Yildiz, S. Artificial neural network approach for modeling of Ni(II) adsorption from aqueous solution by peanut shell. *Ecol. Chem. Eng. S* **2018**, *25*, 581–604. [[CrossRef](#)]
21. Nicomel, N.R.; Leus, K.; Folens, K.; Van Der Voort, P.; Du Laing, G. Technologies for Arsenic Removal from Water: Current Status and Future Perspectives. *Int. J. Environ. Res. Public Health* **2016**, *13*, 62. [[CrossRef](#)]
22. Sorlini, S.; Gialdini, F. Conventional oxidation treatments for the removal of arsenic with chlorine dioxide, hypochlorite, potassium permanganate and monochloramine. *Water Res.* **2010**, *44*, 5653–5659. [[CrossRef](#)]

23. Chen, W.; Parette, R.; Zou, J.; Cannon, F.S.; Dempsey, B.A. Arsenic removal by iron-modified activated carbon. *Water Res.* **2007**, *41*, 1851–1858. [[CrossRef](#)]
24. Herath, I.; Vithanage, M.; Bundschuh, J.; Maity, J.P.; Bhattacharya, P. Natural Arsenic in Global Groundwaters: Distribution and Geochemical Triggers for Mobilization. *Curr. Pollut. Rep.* **2016**, *2*, 68–89. [[CrossRef](#)]
25. Su, C.M.; Puls, R.W. Arsenate and Arsenite Removal by Zerovalent Iron: Effects of Phosphate, Silicate, Carbonate, Borate, Sulfate, Chromate, Molybdate, and Nitrate, Relative to Chloride. *Environ. Sci. Technol.* **2001**, *35*, 4562–4568. [[CrossRef](#)]
26. Jiang, Y.; Yanagita, T.; Mitani, T. Effects of Coexisting Anions on Arsenite Adsorption of Newly Developed Iron Hydroxide. *J. Jpn. Soc. Water Environ.* **2014**, *37*, 169–176. [[CrossRef](#)]



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).