Removal of Arsenic (V) from groundwater by iron-rich material

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**Abstract:**

Recent years, arsenic poisoning has emerged as an environmental catastrophe for the human health in many countries all over the world, including Vietnam. This study aims to optimize and validate the removal of arsenic in groundwater. Experiments designed in this study have shown the model of adsorption on the column of modified iron–rich adsorbent. This follows two different models, Thomas model and Yoon-Nelson model. Besides, the investigation has set up the pilot in practical location to optimum condition for the removal of arsenic in groundwater (pH 6.5 ÷ 7.0, flow rate Q = 5 m3/day), which achieved 76 % removal of arsenic and about 85 ÷ 90% removal of iron in groundwater. The regeneration ability of the material reaches over 70%. Applying in practice at Cu Da Nursery School, Cu Khe Commune, Thanh Oai District, Hanoi city, the pilot scale can supply 5m3 water after removing arsenic per day. The results of pH, As, Fe in the water after the treatment system is consistent with QCVN 02: 2009/BYT (National technical regulation on drinking water quality). These confirm that using modified iron-rich adsorbent materials for removal of arsenic in groundwater is an effective method in terms of economic as well as amelioration of water quality.

Keywords: arsenic, groundwater, adsorption material

**1. Introduction**

In nature, arsenic exists in 200 ores including elements type arsenua, sunfua, oxide, arsenate and arsenite. Arsenic is dispersed into the environment by natural weathering process, geological activity, volcanoes, or by operation of organism dissolves minerals and ores. The transformation of arsenic from the solid phase to the liquid phase is determined by the pH, the redox potential Eh, DO, environmental temperature and water temperature [3, 5, 10, 11]

In term of human’s effect, Humans cause the major impact on arsenic contamination in groundwater through mining activities, construction of geothermal power plants, fossil fuel combustion and other industrial activities, especially, bleached wood industry. In agriculture, arsenic in the composition of herbicides, pesticides, additives in livestock feed… [3, 5]. The arsenic wastewater discharged directly into the environment without treatment. Then the wastewater will permeate the coffee rocks and finally infiltrate groundwater. Therefore, research on the treatment of heavy metals (As) in water is an important subject, that is of the great interest, and research.

Today, the use of waste materials and natural substances for the treatment of heavy metals in water is one of the most considerable research, due to the economic efficiency and the advantage in saving energy. Currently, the common methods of arsenic treatment in the world are: oxidation, deposition; adsorption on adsorbents, co-precipitation [8, 9], sand filtration [7, 8], membrane filtration [12]. The effective of methods depends on many factors such as an existent form of arsenic, arsenic concentration in water, pH, or the presence of other solvents (other metal ions, substances that may interfere with or support in the treatment)… Based on an integrated assessment of the above factors, a method or combination of methods can be selected to achieve the desired results.

In Vietnam, many studies on the absorption of arsenic in water by natural materials and rich iron materials, manganese as modified red mud [2], laterite [1, 2], combination of Zeolite and MnO2 [6]... have shown good results at the laboratory scale and are being continued to study the application in practice. The results showed that when using absorbent material from iron-rich materials, the maximum adsorption capacity was high at pH suitable for drinking water, it means the process has high processing efficiency. Therefore, this study uses iron-rich materials as the main raw material to modify the arsenic adsorption materials.

Many investigations in the world use compounds of iron to absorb arsenic. Studies on the adsorption of arsenic, with both As (III) and As (V) forms, on amorphous iron hydroxide have determined that amorphous iron hydroxide materials have a higher arsenic adsorption capacity of 5 up to 10 times the capacity of activated alumina oxide.

**2. Materials and methods**

***a) Materials and reagents***

Iron–rich materials are Iron oxide ore collected from Mirec Factory at Cao Bang and Iron (III) hydroxide, which was prepared to simulate the iron content of red hydroxide, prepared in the laboratory based on the reaction of FeCl3 with NaOH. Iron ore and hydroxide iron are prepared to a size less than 0.074mm.

Na2HAsO4.7H2O, was dissolved in deionized water for 1000 ppb As(V) stock solution.

NaCl and HCl solution was dissolved in deionized water for 0.02N solution.

All the reagents used were analytical grade and used as received.

***b) Experiments***

*\*Preparation of iron–rich modified materials*

The material components are weighed in two different mixing ratios, the major ingredient is iron ore or iron hydroxide, which was formed in two adsorbents, O1 and H1, respectively, in the following table 2.1

Table 2.1: Mixing ratio (%)

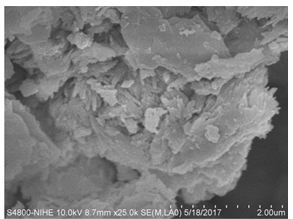
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| --- | --- | --- | --- | --- | --- | --- |
| Material | Iron ore | Ferric Hydroxide | Aluminum hydroxide | Additive 1 | Additive 2 | Kaolin |
| H1 | N/A | 40 | 10 | 7,5 | 18,5 | 24 |
| O1 | 40 | N/A | 10 | 7,5 | 18,5 | 24 |

Materials were made up of 2 cm in diameter and dried to 50 ° C for 24 hours. After that, materials were calcined at 500°C for 10h.

*\* pH effect experiment*

Adjust the pH with 0.02N HCl buffer solution, 0.02N NaOH and 0.02N NaCl solution. A mixture of 0.02N HCl (0.02N NaOH) or 0.02N NaCl solution with a volume of 45 ml + 5 ml of arsenic solution (10 ppm) and 1 gram of adsorbent material was put into a 250 ml triangular flask. This mixture was shaken at 150 rpm for 2 hours, then this solution was filtered with paper and collect the filtered solution for pH measurement and arsenic analysis after adsorption. From that result, determine the optimum pH to adsorb the arsenic of the material and select the pH to proceed with the next experiment.

*\*Investigation the effect of material’s size*

The column adsorption experiment was set up to investigated factors and conditions in removal arsenic process by adsorption. The arsenic adsorption capacity of the modified material was investigated by changing material size: size L from 8 ÷ 9.5 mm, size M and S from 3.75 ÷ 4.75 and 1.0 ÷ 2.0 respectively. Thus, the results are used to evaluate the adsorption capacity of the material based on the Thomas adsorption model and the Yoone - Nelson model.

**b)**

**a)**

***c) Installation and adjustment pilot scale at practical location.***

From the results of the adsorption column study, the system at the practical location was set up to evaluate the effective of arsenic removal in practice.

An adsorbent column, which was 760mm diameter and 1,500mm, was used with 300 mm crystal sand for sand filtration and 120mm modified adsorption material for arsenic adsorption. The system also supports arsenic removal processes such as aeration, settling. Sample was taken at valve, which was installed for sampling after each processing step in the system.

Water samples were measured pH right at the study location when taking sample, after that, analysis of the iron and arsenic concentration in water proceeded in the laboratory.

**3. Result and discussion**

***a) Morphology***

The microstructure and morphology of both the adsorbents from ferric hydroxide and ferric oxide were characterized by SEM. Figure 3.1(a) shows SEM micrographs of adsorbent from ferric hydroxide (H1), and Figure 3.1(b) show the result of modified material from ferric oxide (O1).

Results capture the surface of materials (H1, O1) showed morphology with cavities O1 unclear. The surface is mainly large sheets that fold together to create a small foam for the material. Morphology of the H1 material has a crystalline material surface, large cavities interspersed with walls, walls made of layers stacked by long plates, surface characteristics show that the H1 material has High porosity, and large surface area of contact.

O1’s morphology has sheet, more fissures folded, stacked plates, the holes, pores; H1 materials with multiple cavities, larger pores than those observed at the O1, high porosity, multiple creases and overlapping layers, are characterized by higher surface area and friction.

*b)- adsorbent from ferric oxide*

*(a)- adsorbent from ferric hydroxide*

*Figure 3.1: Comparison of SEM images of synthesized adsorbent from different materials*

The X-ray diffraction pattern of adsorbent materials

The results of the X-ray diffraction of the materials show that the peak formation of the Quartz (SiO2), Hematite (Fe2O3), Maghemite (γ - Fe2O3) here is obvious, but remains impure. The highest accuracy of the Quartz H1 of 55.08%, and Hematite, Maghemite have the accuracy of 39.67% and 18.91%, respectively. The O1 has a Quartz accuracy increase of 98.03% and Meghamite of 31.48% higher than that of the H1 material.

The results of analysis and activation of the material samples have the conversion from iron oxide to Maghemite, the accuracy of Hemaite is almost the same. However, the O1 Quartz and Maghemite materials have increased precision, Quartz as a binder and high Maghemite are also an advantage for arsenic adsorption.



**a)**

**b)**

*Figure 3.2: XRD pattern of synthesized adsorbent from different materials*

*O1 – adsorbent from ferric oxide*

*H1 – adsorbent from ferric hydroxide*

***b) Effect of pH***

The correlation between the pH change of the adsorption solution and the efficiency of arsenic adsorption is shown in Figure 3.3. Preliminary studies carried out at the original pH mixtures (without acid or base addition) showed that there are variations in As adsorption on absorbent from ferric hydroxide and oxide at the pH range 2.3 – 7, the adsorption capacity increased rapidly in H1, O1 materials with the highest adsorption capacity: 0.04 mg As/g, high efficiency: 99.90% at optimum pH 6.81 of H1 material; O1 material has the lowest adsorption capacity with maximum adsorption capacity of 0.037 mg As / g, adsorption efficiency is 90.17%. At this pH, the adsorption capacity of O1 tend to increase slowly, the highest adsorption capacity is 0.039 mg As /g, adsorption efficiency is 93.50%.

When pH is increased from neutral to alkaline medium in the pH range 7,5 – 12, the decrease in the adsorption yield was occurred, the O1 material tends to decrease quickly and the H1 decrease slightly with the lowest adsorption capacity at pH 11 ÷ 12 with only 0.0329 mg As/g, the adsorption efficiency was low at 80.39%.

*Figure 3.3: Effect of pH on adsorption of As by synthesized adsorbent from different materials*

*O1 – adsorbent from ferric oxide*

*H1 – adsorbent from ferric hydroxide*

In a study carried out at comparable conditions with the present study, it has been reported the results that the optimum pH for the adsorbent material of the materials was found to be in the range of 5 ÷ 8. pH > 8, the possibility of material adsorption decreases. Compared to other results of the research group, other As absorbable materials such as kiln (optimum pH in the range of 4 ÷ 6), bentonite red mud and red kaolin modified (optimum pH) about 4.2 to 6.5), showed that the optimum pH range of the modified materials in this study is the largest. Typically, the pH of groundwater is 6.0 to 8.5, so material H1, O1 have optimum pH range suitable for removal of arsenic in groundwater.

***c) Effect of particle size on the removal of arsenic***

Sample solution As (V), with concentrations approximately 1,000 ppb, was used in the experiments has a pH in the range of 2 optimized materials used in the experiment, suitable for use in adsorption experiments with continuous flow running through the column. Recognizing that the pH of the solution after removal from the adsorption column changes, due to arsenic adsorption of iron-rich material [9] (O1 and H1), this reaction increases the pH slightly.

The increase in percentage removal of all arsenic species with the decrease in particle size (from 8 ÷ 9.5 mm to 1 ÷ 2 mm) of the adsorbent is very different. Among the particle sizes considered in the experiment, the percentage removal is maximum for the particle size of 1 ÷ 2 mm for both O1 and H1.By application of the O1 and H1 adsorbent, Figure 3.5 and 3.6 shows the smaller the material size gets, the greater the adsorption capacity is, this trend occurred during the investigation.

However, the difference in concentration of As in the water sample from 200 minutes to 1.000 minutes of reaction time is not significant, which is most apparent in the first 100 minutes of the experiment. After 5 minutes from the start of the continuous flow through column analysis, the concentration of As in the water sample of the 3 OL, OM, OS columns was 326, 222 and 8 ppb respectively. At the time of the experiment was 800 minutes, the difference was not clear when the concentration of As in the sample water output of 3 columns OL, OM, OS is 447, 367 and 367 ppb respectively.

With the H1, at 90 minutes, the arsenic concentration in the water sample of the 3 columns containing the HS, HM, HL materials was 180, 400 and 470 ppb, respectively. After 660 minutes of experiments, the concentration of As in the outlet water in the 3 columns of materials ranging from small to large, HS, HM and HL, respectively, 150, 408 and 584 ppb. At the last sample, the difference in removal of As is still very different between the three sizes of the material.

Both of Figure 3.4 - a and 3.4 – b also shows that, as the experiment duration is extended, the adsorption efficiency decreases, which is precisely the theory of adsorption.

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| a) |
| b) |

*Figure 3.4: The As concentration in the solution after adsorption of O1 (a) and H1 (b) materials at 3 different sizes*

*(HL & OL – (8,0-9,5 mm);HM & OM – (3,75-4,75 mm); HS & OS – (1,0-2,0 mm))*

***d) The Thomas and Yoone - Nelson model***

Experiment using two common adsorption models. The Thomas model was fitted to the column data to investigate the breakthrough curve of As(V) adsorption onto O1 and H1. Application of the Thomas model to the data in the concentration (Ct) range of 1 ppb to 1,000 ppb with the fixed column height, initial As(V) concentration and flow rate helped in the determination of the Thomas’ kinetic coefficients for this system. The coefficients were determined from the slope and intercepts obtained from the linear regression performed on each set of transformed data. Analysis of the regression coefficients indicated that the regressed lines provided excellent fits to the experimental data with R2 values ranging from 0.7098 to 0.7453 (Table 3.2). Table 3.2 also presents the values of KT and q0. The bed capacity q0 increased and the coefficient KT decreased with increasing particle size for both modified adsorbent materials O1 and H1.

The simple Yoon–Nelson model was applied to investigate the breakthrough curve of As(V) onto O1 and H1 fixed bed. This model introduces the parameter τ, which shows the treatment time taken for Ct (effluent exit concentration) to be half the initial concentration (C0/2). The values of the model parameters KYN (rate constant) and τ were determined from the slope and intercepts of the linear plots of ln[Ct/(C0 −Ct)] versus time “t” with respect to bed depth, initial As(III) concentration and flow rate and are presented in Table 3.5.

The experimental data exhibited good fits to the model with linear regression coefficients ranging from 0.7089 to 0.7453 (Table 3.2), similar to KT of Thomas model. As is evident from the table the experimental and the calculated τ-values are very close to each other indicating that the Yoon–Nelson model fits excellently to the experimental data. Comparison of breakthrough curves obtained experimentally with those predicted using the Yoon–Nelson model are shown in Table 3.2. The results in the figures clearly indicate that the model proposed by Yoon–Nelson provided a very good correlation with the particle size of adsorbent materials.

Predicted parameters show that Thomas (as well as Yoone-Nelson) adsorption models are relatively similar for all material in this investigation, the values also suggest that the adsorption capacity is maximized, such as time to absorb 50% As(V) in distilled water of the same size from O1 (major component of iron oxide) is shorter than H1 (the material from ferric hydroxide).

The results also indicated that in most cases, decrease the size of adsorption materials would bring an increase in maximum adsorption capacity as well as a longer time to absorb 50% As in distilled water (V). From the above results, it seems that two types of adsorption (simulator of red mud) have the ability to adsorb arsenic, so it can be applied to pilot experiments at larger scale to continue, and calculation maximum capacity of two materials in practice in study location.

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| |  |  |  |  |  |  |  | | --- | --- | --- | --- | --- | --- | --- | |  | Thomas | | | Yoone - Nelson | | | |  | R2 | KT | qo | R2 | KYN | τ | | OL | 0.718 | 0.000967 | 39.7183 | 0.718 | 0.0009 | 616.000 | | OM | 0.742 | 0.001294 | 20.7630 | 0.742 | 0.0011 | 454.273 | | OS | 0.760 | 0.001985 | 50.8752 | 0.760 | 0.0019 | 1056.316 | | HL | 0.710 | 0.001315 | 21.9686 | 0.710 | 0.0012 | 312.417 | | HM | 0.638 | 0.000353 | 73.2814 | 0.638 | 0.0003 | 1344.000 | | HS | 0.745 | 0.000573 | 129.105 | 0.527 | 0.0005 | 2857.400 |   *Table 3.5: Predicted Thomas and Yoone - Nelson model parameters and their deviation from experimental values for As (V) adsorption onto O1 and H1 at different particle size* |

***e) Evaluating the change of pH, the arsenic and iron removal capacity of two adsorbents with pilot scale and 5 m3.day-1 volume flow rate.***

*Iron removal*

Figure 3.5 shows that the process of aeration, sedimentation and sand filtration is highly effective, iron removal performance reached about 85 ÷ 90% (from 8 to 12 mg / L to 2 ÷ 3 mg / L after sand filtration).

With the pilot using O1 material, after the water flows through the main absorbing material (O1), the iron in water is almost unchanged from that obtained at the outlet valve after exiting the sand filter (V2), indicating that the material O1 persistent, non-corrosive adsorbent (corrosion causes iron from adsorbed material to be released back into water).

With H1, after water flows through the primary adsorbent (H1), the iron in water increases slightly, compared to the sample taken at the valve after exiting the sand filter (V2), which may be explained by the corrosion of the H1’s surface for a short initially operated. Therefore, O1 material is more durable and wear – resistant in operation than H1.

However, days later, the Fe concentration at the V4 and V5 sampling valves was not significantly different from the sample, which is taken at the V3 valve (after exiting the sand filter layer and before running through for the H1 material layer), indicating that the material was no longer corroded.

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| **a)** |
| **b)** |

*Figure 3.5: Concentration of Fe after each stage of process a) O1 b) H1*

*(O1 – adsorbent from ferric oxide; H1 – adsorbent from ferric hydroxide, V0 – groundwater, V1 –water after aeration, V2 – water after sedimentation; V3 – water after sand filtration; V4 – water after flow through adsorbent O1; V5 – water was taken from the outlet of process)*

*Arsenic removal*

Figure 3.7 – a and 3.7 – b shows that the results of the groundwater samples in Cu Da village in all sampling days have an average concentration of As from 250 to 400 ppb. In the process, when water runs through aeration, settling and sand filtration, As levels are significantly reduced. After sedimentation, Concentration of As decreases considerably to about 150 ppb (approximately 50% of arsenic concentration in groundwater) due to As adsorption, producing FeAsO4 and trapping arsenic at this tank. When water flows through the sand filter layer, Fe(OH)3 continues to precipitate here, of course, the As adsorption process continues to occur, resulting in a concentration of As of about 55 to 90 ppb. After this stage, the water is adjusted to flow to the main adsorption layer in the opposite direction of gravity.

Two separated pilots were setup to test the ability O1 and H1 to remove arsenic. With the O1 – the adsorbent material with the major components of the material from iron ore, water continues to flow through the main adsorbent layer O1. At this stage, the iron in the water has been removed to a very low level, so that the O1 (the iron-rich adsorbent material) plays a key role in removal arsenic in the water. The concentration of arsenic in water after this stage ranging from 12 to 50 ppb, so the O1 material has shown the effectiveness of arsenic removal in groundwater.

Overall assessment showed that water samples taken at the outlet of the system, after all treatment stages, have concentrations of As below 0.05 mg/L (50ppb) and reach the QCVN 02: 2009/BYT standards, arsenic in groundwater has been successfully removed.

With the H1 - the adsorbent material with the major components of the material from ferric hydroxide, after the water flows through this material, the As in water output is from 12 to 50 ppb, this means that the H1 adsorption layer exhibits an important role in arsenic adsorption, similar to what the O1 has shown.

During the first two days of sampling, water samples taken at the exit of the system had an concentration of As of less than 10 ppb, which could explain the original H1 material layer with very porous and water-absorbing surfaces, so the contact area of this iron-rich adsorption material increases, this means the As processing capacity is risen too. Overall assessment showed that water samples taken at the outlet of the system, after all treatment stages, have concentrations of As below 0.05 mg / L (50ppb) and reach the QCVN 02: 2009 / BYT standards, arsenic in groundwater has been successfully removed.

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| --- |
| a) |
| b) |

*Figure 3.7 – a): Concentration of As after each stage of process*

*b): Concentration of As after each stage of process*

*(O1 – adsorbent from ferric oxide; H1 – adsorbent from ferric hydroxide, V0 – groundwater, V1 –water after aeration, V2 – water after sedimentation; V3 – water after sand filtration; V4 – water after flow through adsorbent O1; V5 – water was taken from the outlet of process)*

**4. Conclusion**

Experiments designed in this study have shown the model of adsorption on the column of modified iron–rich adsorbent. This follows two different models, Thomas model and Yoon-Nelson model. Parameters from two kinematic models, such as absorption capacity, saturation time found in the laboratory, are used to predict the process occurring during actual operation. Besides, the investigation has set up the pilot in practical location to optimum condition for the removal of arsenic in groundwater (pH 6.5 ÷ 7.0, flow rate Q = 5 m3/day), which achieved 76 % removal of arsenic and about 85 ÷ 90% removal of iron in groundwater. The regeneration ability of the material reaches over 70%. Applying in practice at Cu Da Nursery School, Cu Khe Commune, Thanh Oai District, Hanoi city, the pilot scale can supply 5m3 water after removing arsenic per day. The results of pH, As, Fe in the water after the treatment system is consistent with QCVN 02: 2009/BYT (National technical regulation on drinking water quality). These confirm that using modified iron-rich adsorbent materials for removal of arsenic in groundwater is an effective method in terms of economic as well as amelioration of water quality.

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Xử lý Asen (V) trong nước ngầm bằng vật liệu giàu sắt

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**Tóm tắt**

Những năm gần đây, ảnh hưởng của asen đến sức khỏe con người đang được coi như là một thảm họa môi trường, và là vấn đề cấp bách ở rất nhiều nước trên thế giới, trong đó có Việt Nam. Nghiên cứu này hướng đến các quá trình hiệu quả để loại bỏ asen trong nước ngầm. Những thí nghiệm trong nghiên cứu này chỉ ra các mô hình hấp phụ cột trong điều kiện dòng chảy động theo 2 mô hình động học là Thomas và Yoone-Nelson, với vật liệu hấp phụ sử dụng là vật liệu giàu sắt đã qua biến tính. Thêm vào đó, sử dụng các kết quả có được trong phòng thí nghiệm, nghiên cứu được áp dụng với quy mô pilot tại địa điểm thực tế, với mục đích tối ưu các điều kiện của quá trình hấp phụ loại bỏ As trong nước ngầm (pH từ 6,5 đến 7,0, lưu lượng qua hệ Q = 5 m3/ngđ). Tại nơi đặt hệ xử lý – Trường mầm non Cự Đà, xã Cự Khê, Thanh Oai, Hà Nội – mô hình được lắp đặt có công suất cung cấp nước sau khi loại bỏ As là 5m3/ngđ. Nước sau xử lý đạt kết quả tốt khi loại bỏ được 76% asen, đồng thời loại bỏ được 85-90% sắt trong nước, các thông số như pH, As, Fe đều đảm bảo QCVN 02: 2009/BYT (Quy chuẩn kĩ thuật Quốc Gia về chất lượng nước sinh hoạt), khả năng hoàn nguyên của vật liệu đạt tới hơn 70%. Các kết quả về khả năng loại bỏ asen trong nước ngầm của vật liệu giàu sắt biến tính cho thấy vật liệu này là một giải pháp hữu ích cả về kinh tế cũng như ứng dụng trong cải thiện chất lượng nguồn nước.

Từ khóa tìm kiếm: asen, nước ngầm, vật liệu hấp phụ

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