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Biosorption of Arsenic from Contaminated Water: Composite of Algae and Fe-Nano Particles

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Abstract: *Arsenic poisoning has been a great concern for the last two decades because of its extensive problems. Coagulation, ion exchange, ultrafiltration, and adsorption systems have been used for the removal of arsenic from polluted water. In this study, the biosorption process was used owing to eco-friendly, simplicity, biodegradable, and cost effectiveness. The uptake of arsenic from contaminated water by Ulva lactuca, Spirulina platensis, Chlorella vulgaris, orange peel, NZVI, zeolite, and composite of NZVI and algae was studied. This study observed that Chlorella vulgaris and iron coated Chlorella vulgaris absorb arsenic more effectively than other algae and iron coated algae. After 24 hours, Chlorella vulgaris and Fe-coated Chlorella vulgaris absorbs 23% and 67% arsenic from contaminated water, respectively. The presented findings depicted the great potential of using bio-sorbents and the Fe-algae bio-composite for the removal of arsenic from contaminated aqueous solutions.*

Keywords: Algae, Arsenate, Biosorption, Composite, NZVI

1. INTRODUCTION

Arsenic is an extremely poisonous heavy metal contained in the earth's crust that poses a serious threat to the environment and human health [1,2]. Arsenic is called the king of poisons. Marine sedimentary deposits, volcanic eruptions, natural erosion mechanisms, geothermal processes, and tectonic activity all contribute to its natural distribution into groundwater [3,4]. Moreover, arsenic adulteration occurs in groundwater due to human reckless doings, such as the use of agricultural pesticides, the extraction of excessive minerals from mining, and other anthropogenic activities [5,6]. Continuous exposure to arsenic contaminated water in human everyday life, such as preparing food, swallowing water, and irrigation, poses a risk to human health, including cancers of the skin, bladder, lung, kidney, breast, as well as cardiovascular disease, blackfoot diseases [7], hypertension, skin diseases (hyperkeratosis) and other ailments [8–11]. Because of its high toxicity to humans, the WHO (World Health Organization) and EPA (Environmental Protection Agency) have set 10 μgL^{-1} as the maximum concentration of arsenic in drinking water.

Several removal technologies have been proposed in the literature for heavy metal removal from polluted water. Notable removal processes were precipitation, reverse osmosis, ion exchange, oxidation, permeable reactive barriers, coagulation and membrane filtration, and bio-adsorption [12–14]. Almost all these methods are extremely expensive, non-eco-friendly, and complex operative. However, biosorption processes are promising, cost effective and environment friendly. This process removes heavy metals from water using biological resources such as algae [15], fungi, plant, bacteria, and genetic engineering breeding species. Because of its high heavy metal uptake capacity, algae biomass is one of the most promising kinds of biosorbent. Different types of algae have different affinities towards the similar metal, which are determined by the internal chemical structure of the algae. Several algae for example *Chlorella vulgaris*, *Spirulina platensis*, *Caulerpa lentillifera*, *Ulva lactuca*, *Oscillatoria*, *Chlorella pyrenoidosa* are efficient to eradicate heavy metals such as As, Cd, Hg, and Cr(VI) from contaminated water. The main advantages of the algae are cost-effective, ameliorate for environment, and easy to cultivate [16].

The purpose of the study was to find the comprehensive investigation on adsorption of As(V) using algae, and other effective environment friendly biosorbent materials [17,18]. Furthermore, this study also examined the arsenate adsorption by composite materials of algae and Fe-nano particles. The biosorption of arsenate (As (V)) was measured using inductively coupled plasma mass spectrometry method (ICP-MS Agilent-7500cx). ICP-MS Agilent 7500cx helium mode removes matrix induced interference and hydrogen mode eradicates molecular ions affected by argon. It ensures the highest performances with high consistency.

Table 1. Present National Standards for Arsenic in Drinking Water and Exposure Scenario

Country	Standard value (μg/l)	Estimated affected people (millions)	Arsenic concentration in different sources (μg/l)	References
Bangladesh	50	85	Greater than 10 to less than 2000	Edmunds W.M. et al. [19], Hossain M.F. et al. [20]
India	50	50	Greater than 10 to less than 3200	Bhowmick S. et al. [21], Biswas A. et al. [22]

Nepal	50	13	Greater than 10 to less than 2600	Thakur J. et al. [23]
Pakistan	50	13	Greater than 10 and greater than 2500	Muhammad S. et al. [24]
Canada	25	1	Greater than 10 to less than 2000	Bondu R. et al. [25]
USA	10	0.3	Greater than 10 to less than 3000	Flanagan S.V. et al. [26]
Argentina	15	8	Greater than 10 and less than 2000	Marta I. Liter [4]
China	10	20	Greater than 5 to less than 500	Guo H. et al. [27], Chen L. et al. [28]
Australia	7	0.5	Greater than 10 to less than 3000	Appleyard S.J. et al. [29]
WHO	10	-	-	-
EPA	10	-	-	-

2. MATERIALS AND METHODS

2.1 Chemicals

Sodium arsenate dibasic heptahydrate ($Na_2(AsHO_4).7H_2O$, or $AsH_{15}Na_2O_{11}$, Sigma-Aldrich company limited, Germany) were considered for the formulation of arsenate (As (V)), hydrochloric acid (HCL), Sodium Hydroxide (NaOH, purity 97.0%) were procured from Sigma Aldrich (Germany) and were of analytical grade. In this experiment, deoxygenated deionized water was exploited to make all the solutions.

2.2 Materials (biosorbent)

Ulva lactuca was collected from Kagoshima university, Laboratory of Microbiology, Faculty of fisheries, Kagoshima prefecture, Japan. The biomass of *Spirulina* species was bought from Taobao marketplace, China. This fish food product comprises ten kinds of vitamins (for example, vitamin A, B_6, B_{12}) proteins, minerals, amino acids, and chlorophyll, which stimulates the growth of fish. *Chlorella* species was collected from the Agricultural Botany Department, Faculty of Agriculture, Kafrelsheikh University, Karh El-Sheikh in Egypt, as part of the project of algal biomass production for use as non-conventional fish feed; remediation of chemicals contaminated water; carbon dioxide capture; and biodiesel and omega-3 fatty acid production was funded by National Strategy for Genetic Engineering and Biotechnology Phase (III). One-gram iron nanoparticles (NZVI) was prepared using 0.092 M ferric chloride hexahydrate and 0.58 M sodium borohydride solution following the optimized synthesis approach previously reported [30–32]. Mixture was conducted under anaerobic conditions. During the synthesis process, 30°C temperature was maintained. The black particles were isolated by vacuum filtration through a 0.45 micrometer cellulose membrane filter and washed double times by deionized water. Then the sample was used in the experiments. Zeolite was purchased from Sigma-Aldrich Co., Germany.

2.3 Stock solution:

One liter arsenic (V) stock solution was prepared by dissolving 83.3 mg $Na_2HAsO_4.7H_2O$ in 100 ml deionized water. 50 ml nitric acid was added to the solution while mixing (common concentrations is 5% v/v, to keep As (V) in ionized form and to prevent transformation to As (III) or to stock on the wall of the conical flask). After that, the solution was diluted to 1000 ml with deionized water. Arsenic (V) solution was stirred thoroughly and then kept in the refrigerator. The pH of the arsenic solution was maintained to 5.5 using HCl and NaOH.

2.4 Biosorption process of metal ions (As) by Algae, NZVI, zeolite and new composite

The experiments on bio-adsorption properties were operated for the functionalized *Ulva lactuca*, nano zero valent iron (NZVI), zeolite, live and dead (powder) *Chlorella Vulgaris*, *Spirulina platensis*, orange peel (result is not included), and *Ulva lactuca*. The batch experiments were performed in conical flasks comprising 100 ml of As(V) solution in a magnetic stir shaker under 1000 rpm at 25° C and initial pH was 5.5. At various time intervals, 2 ml samples were collected over 24 h reaction time and filtered with 0.45 micrometer membrane for analysis. The following equations were used to calculate removal efficiency and adsorption capacity.

$$q = \frac{(C_0 - C_e)}{m}V \qquad (1)$$

$$\mathrm{Re}(\%) = \frac{C_0 - C_e}{C_0} \times 100 \qquad (2)$$

where q indicates the adsorption capacity, C_0 is the initial concentration of arsenic, C_e is concentration of arsenic at equilibrium states, m is the mass of the sorbent dose, and Re indicates the removal efficiency.

3. RESULTS AND DISCUSSION

Assessing and observing arsenic (V) content in drinking water is tremendously significant because 500 million people around the world are affecting by its impact [33]. Around 48% of the total population was affected by groundwater arsenic in Bangladesh [34]. Table 1 illustrated the arsenic contamination level, affected people by arsenic, and standard value in different countries. There is no known effective treatment or medicine for arsenic toxicity, affected people can recover from the toxicity by consuming healthy food and drinking arsenic free water. Proper cost-effective water treatment along with awareness about arsenic is effective to solve the present crisis. Biosorption of arsenic using algae is one of the most reliable, cost-effective, and environment friendly process.

For analyzing the biosorption of arsenic, it is an important issue to find the calibration curve in measurement procedures. This curve established the relationship between the calibration standard values and experimental data. The calibration curve for arsenic (V) was functioned using a stock solution. Ten working standard solutions was prepared with 0.25, 0.5, 1, 3, 5, 10, 30, 50, 70, and 100 ppb. Arsenic calibration curve was shown in Fig. 1. In this study, following calibration curve was used for calculation of removal efficiency of arsenic from contaminated water.

$$Y = 2349.1X + 58.654 \qquad (3)$$

For the calibration curve, we found that the R-squared value is 1 in this study. The following expression is used for finding the R-squared value.

$$R^2 = \frac{\text{Variance explained by the corresponding model}}{\text{Total variance}}$$

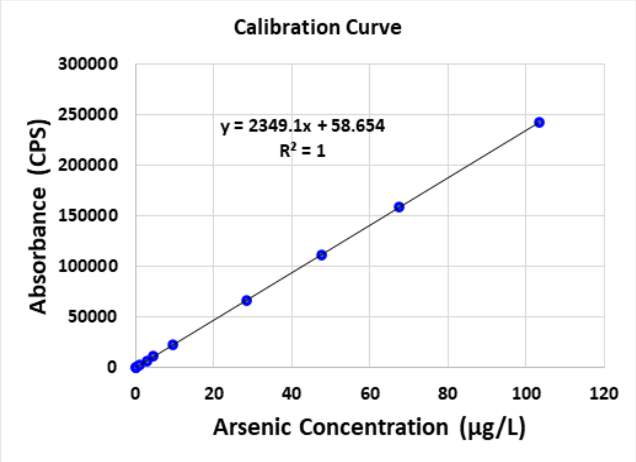

Fig. 1. Arsenic calibration curve determined using ICP-MS 7500cx.

In this study, the tests were accomplished for different dosage algae concentration. 1 g/l, 5 g/l, 10 g/l, 15 g/l and 20 g/l dosages of *Spirulina platensis*, *Ulva lactuca*, *Chlorella vulgaris*, and other composite materials with different concentration of Arsenic (V). All tests were conducted at pH 5.5. In the previous literature, it was found that if pH > 5.5, then arsenic and chromium adsorption were decreased [35]. Temperature and pH are decisive factors in biosorption investigations because they influence the solubility of metal. For biosorption study, we use different initial dosage of algae sample and other biosorbent composite materials. Fig. 2 showed the circumstances before the experiment and during the test of arsenic adsorption.

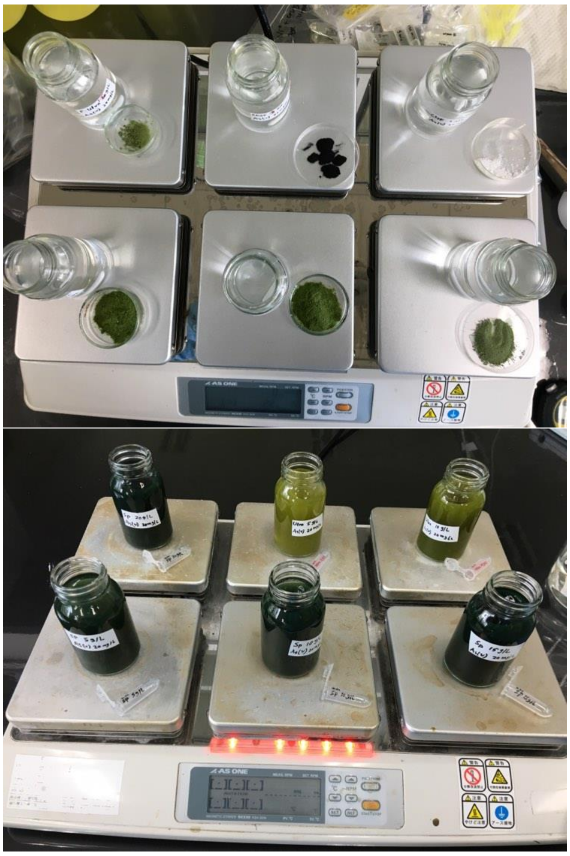

Fig. 2. Experiment of As(V) biosorption using algae before experiment (upper figure) and during experiment (lower figure).

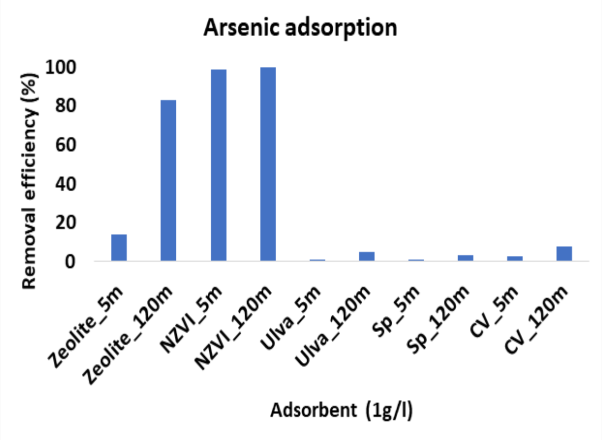

Fig. 3. Arsenic adsorption using different adsorbent materials (1g/l).

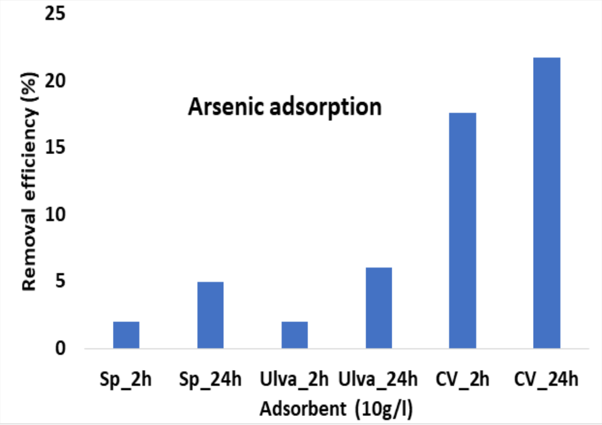

Fig. 4. Arsenic adsorption using different algae (10g/l) with different adsorption times.

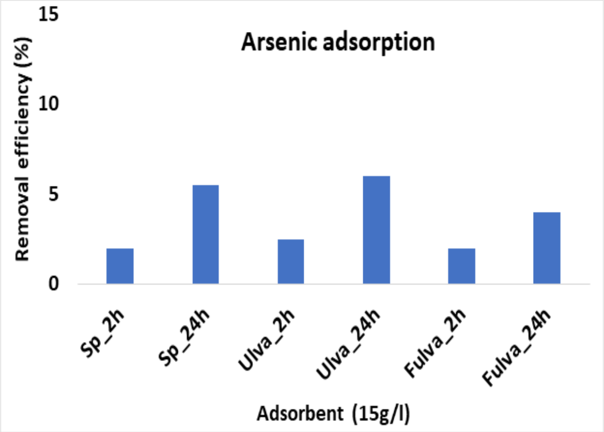

Fig. 5. Arsenic adsorption using different algae (15g/l) with different adsorption times.

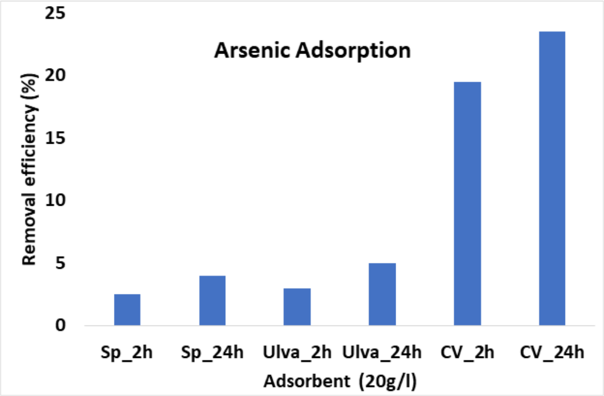

Fig. 6. Arsenic adsorption using different algae (20g/l) with different adsorption times.

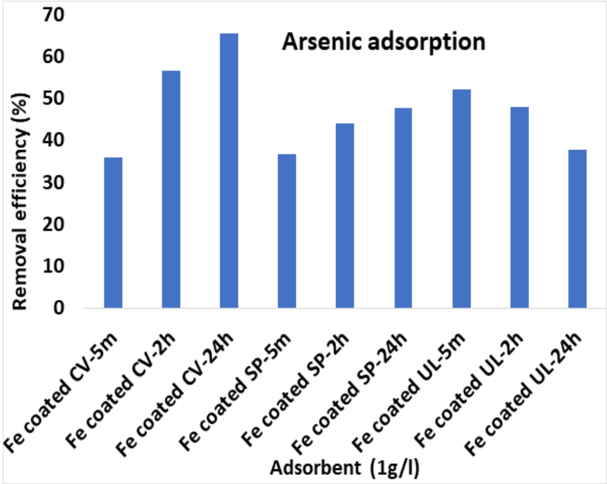

Fig. 7. Arsenic adsorption using algae composite materials (1g/l).

Fig. 3 to Fig. 6 describes the arsenic adsorption for different dosage of algae, iron nano particles, and zeolite. In Fig. 3, the usage dosage of *Ulva lactuca*, *Spirulina platensis*, *Chlorella vulgaris*, zeolite, and NZVI are 1 g/l for arsenic adsorption. It is observed that NZVI adsorbed approximately 99% arsenic after 5 minutes, zeolite adsorbed about 85% but pure algae did not show good adsorption. The main reason for the rapid and efficient removal of As(V) by NZVI is the involvement of different removal mechanisms within the reaction, including the electrostatic attraction of the negatively charged arsenate species to the positively charged surface of NZVI at pH 5.5 (point of zero charge was reported to be at pH 8.2) [36,37]. Moreover, the possible reduction of As(V) to As(III) via the released electron from the iron core, and hence the involvement of precipitation and co-precipitation of As(III) with Fe(II) and Fe(III) on the surface. While the porous structure in addition to the high ion exchange ability were the main As(V) removal mechanisms by zeolite. When we increase the dosage of algae for example 10g/l, 15 g/l, and 20 g/l (Fig. 4, Fig.5, and Fig.6), only *Chlorella Vulgaris* adsorbed about 24% arsenic after 24 hours for 20g/l. In this study, algae dosage increases due to its environmentally friendly nature, cost effective and easy to cultivate. Although the dosage of algae has been increased, the amount of arsenic adsorption has not increased significantly. NZVI absorbed approximately 100% arsenic though has some problems such as poor mobility, particles agglomeration, costly, and limited suspension stability. For this reason, we prepare composite materials using algae (for example, *Spirulina platensis, Ulva lactuca, and Chlorella vulgaris*) and NZVI in this study.

To eradicate the disadvantage of nano zero valent iron, represented in the strong tendency of the particles to aggregate, poor stability in aqueous suspensions, and the rapid corrosion of the iron core, composite materials have been made using algae and NZVI. In this research, 40% Fe-nano particles and 60% algae (*Spirulina platensis*, *Chlorella vulgaris*, and *Ulva Lactuca*) was mixing physically using mortar and shell. 1 g/l composite materials of algae and NZVI were used for this experiment to adsorb arsenic. In Fig.7, it is observed that Fe coated *Chlorella vulgaris* (Fe coated CV) increases the adsorption of arsenic corresponding with time. Fe coated CV showed 36%, 55%, and 67% removal efficiency after 5 minutes, 2 hours, and 24 hours respectively. Fe coated *Spirulina platensis* (Fe coated SP) also showed similar trends for arsenic adsorption. But Fe coated SP adsorbed about 50% arsenic from the sample after 24 hours. Fe coated *Ulva Lactuca* (Fe coated UL) showed surprising behavior for arsenic adsorption. Fe coated UL decreases the adsorption of arsenic corresponding with increasing time. Fe coated UL showed 52%, 48%, and 38% removal efficiency after 5 minutes, 2 hours, and 24 hours respectively.

4. CONCLUSION

In this research, comprehensive approach was adopted to analysis the uptake of arsenic from contaminated water by *Chlorella vulgaris*, *Ulva lactuca*, *Spirulina platensis*, orange peel, nano-zero valent iron, zeolite, and composite of algae and iron nano particles. From this study, we found that *Chlorella vulguris* and Fe coated *Chlorella vulgaris* are more efficient than other algae to absorb arsenic (V). In the future, we will observe the arsenic adsorption by algae and other efficient environment-friendly materials and their chemical compositions.

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