ARSENIC REMOVAL FROM AQUEOUS SOLUTIONS USING SODIUM ALGINATE AND PLEUROTUS OSTREATUS (OYSTER MUSHROOM) BEADS

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Abstract - Arsenic contamination in groundwater is a significant global health concern, affecting millions of people worldwide. This study focuses on the removal of arsenic ions (As(V)) from aqueous solutions using alginate beads incorporating Pleurotus ostreatus mushroom powder as a biosorbent. The effects of pH, bead composition, and bead hydration state on the removal efficiency of As(V) were investigated. The study found that pH significantly influenced the removal efficiency, with pH 5 showing a significant increase in As(V) removal percentage. This was attributed to the formation of strong bonds between the biosorbents and arsenic ions at pH 5. The chemical composition of the beads also played a crucial role, with an increase in Pleurotus ostreatus mushroom powder concentration from 4% to 6% leading to a significant increase in removal efficiency. However, increasing the alginate concentration from 4% to 6% did not show a significant difference, possibly due to the compactness of the beads at higher alginate concentrations. Additionally, the hydration state of the beads affected the removal efficiency, with dried beads showing higher removal efficiency compared to fresh beads. This was attributed to the smaller size of dried beads, providing a larger contact surface area for adsorption. FTIR analysis confirmed the presence of hydroxyl (-OH), alkyl, NH_2 and carboxyl (-COO) groups in the synthesized beads, which are assumed to be responsible for the uptake of arsenic ions. Overall, the study demonstrates the potential of alginate beads incorporating Pleurotus ostreatus mushroom powder as an effective and environmentally friendly biosorbent for arsenic removal. Further optimization of the biosorbent composition and hydration state could enhance the removal efficiency, making it a promising approach for arsenic remediation in contaminated water.

Keywords - alginate and mushroom beads, arsenic, atomic absorption spectrophotometer, FTIR.

I. INTRODUCTION

Arsenic (As) is a metalloid element that occurs naturally in the Earth's crust. The different types of arsenic such as As (V) and As (III) are commonly found in groundwater all over the world. Arsenic contamination sources can be mainly caused as natural sources and anthropogenic sources by human activities. It is commonly found in various geological formations, ores, minerals and so on. In Asia, anthropogenic sources of arsenic contamination in groundwater are mainly formed sedimentary formations 45%, mining (30%), coal (10%), petroleum (10%) and volcanic rocks (5%) [1 ,2].

Arsenic is a well-known toxic that is naturally presented in water, soil and air. [3,4]. Arsenic contamination in water sources has become a significant public health. and can cause in acute and chronic poisoning that primarily through ingestion [5]. Many studies have demonstrated that exposure to arsenic, even at low concentrations, can lead to various health issues, including skin lesions, cardiovascular diseases, and even cancer.

long-term arsenic exposure has adverse impacts on human health, causing skin lesions, cardiovascular diseases, neurological complications, reproductive and respiratory disorders, and malignant and non-malignant diseases [1] Arsenic contamination is a global issue, affecting both developing and developed countries. Shaji et al reported that arsenic contamination in groundwater affected 107 countries from globally. In addition, South Asian and South American regions were also found the natural contamination of As in groundwater.

These countries were Afghanistan, Armenia, Azerbaijan, Kyrgyzstan, Bangladesh, Cambodia, China, Georgia, India, Indonesia, Iran, Japan, Jordan, Laos, Kazakhstan, Korea, Malaysia, Mongolia, Myanmar, Nepal, Pakistan, Philippines, Russia, Saudi Arabia, Sri Lanka, Tajikistan, Thailand, Turkey, Turkmenistan, Uzbekistan and Vietnam. Among them, Southeast Asian countries, Bangladesh, India, Pakistan, China, Taiwan, Nepal, Vietnam, Myanmar, Thailand and Cambodia, are the most affected [2].

Arsenic contamination in groundwater is a pressing global issue that impacts millions of people, including those in Myanmar. The problem is particularly important in regions such as the Ayeyarwady Delta and the central dry zone, including Mandalay city.

Across Myanmar, arsenic levels exceeding the World Health Organization's (WHO) guideline of 10 micrograms per liter (µg/L) have been documented in numerous areas. Both the US Environmental Protection Agency (EPA) and the WHO have set the guideline for maximum allowable arsenic contamination in drinking water at 10 µg/L [1,3–12]. This contamination poses a significant health risk to

communities, highlighting the urgent need for remediation efforts and improved water quality management strategies.

Researchers have explored various methods to remove arsenic from contaminated water, including coagulation-flocculation, filtration, ion exchange, and remediation techniques.

However, these methods have limitations in terms of cost, efficiency, and sustainability. In contrast, biosorption and adsorption methods have emerged as cost-effective and environmentally friendly alternatives that are suitable for widespread adoption. However, further research is needed to evaluate the performance of these methods under local conditions for many communities in Myanmar.

Mushrooms are known for their excellent binding properties and tolerance to metals and adverse environments, such as various pH and temperature conditions [10,13]. They are also environmentally friendly and can quickly grow on many organic wastes or lignocellulosic wastes within 3–4 weeks [11]. Pleurotus ostreatus has shown promise in absorbing heavy metals from its surroundings.

When combined with other materials, such as alginate, mushrooms have demonstrated synergistic effects, enhancing heavy metal removal efficiency. This study aims to reduce arsenic concentrations using a biosorption method with Pleurotus ostreatus mushroom as an inexpensive biosorbent, mixed with sodium alginate to form beads known for their low cost, ease of use, and excellent removal efficiency.

The objective of this work is to evaluate the effect of Pleurotus ostreatus powder and alginate mixture beads on removing arsenic from aqueous solutions.

II. MATERIALS AND METHODS

A. Collection and preparation of mushroom powders

The fresh mushroom (Pleurotus ostreatus) samples were purchased from the local market in Kyaukse, Myanmar. The mushroom samples were thoroughly cleaned with tissue paper to remove the dust, cut into pieces with a knife and dried at room temperature for 3 days. After drying, it was ground with a blender and stored in plastic bags for later use at room temperature.

B. Preparation of bead formation

All the glassware (tubes, conical flasks, beakers, etc.) was sunk with HNO_3 for several hours in acidic water (10% nitric acid) before proceeding with a regular washing procedure and washed with deionized (DI) water before being used in the experiments.

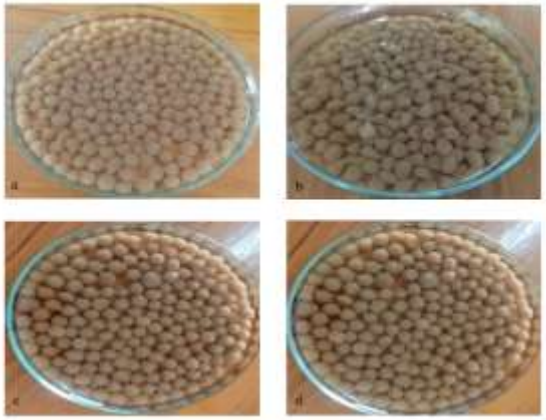


Figure 1. Bead formation of (a) 4% Alginate and 4% Mushroom Powders, (b) 4% Alginate and 6% Mushroom Powders, (c) 6% Alginate and 4% Mushroom Powders, and (d) 6% Alginate and 6% Mushroom Powders.

Then, they were dried in an oven at 60◦C for 24 hrs. Firstly, sodium alginate ($C_6H_7O_6Na$) (4% and 6%) was added to the 100 ml of DI water and stirred with magnetic stirrer at 60°C for 35 min. The mushroom powders (4% and 6%) were added to the sodium alginate solutions and stirred with a magnetic stirrer for 35 min. After cooling, this solution is dropped by a 60 ml CC syringe without a needle into the 1M calcium chloride solution and left for 5 hours to obtain strength. The diameter of fresh beads was 2 to 4.5 mm. They were washed with DI water and dried at room temperature overnight. The formation of these beads is shown in Fig. 1.

C. Preparation of Arsenic solution

Arsenic solution was purchased from the local market in Myanmar. This solution was prepared by dissolving in deionized water. The solution was stirred with a magnetic stirrer for 30 min.

D. pH adjustment

The original pH of As solutions was adjusted by using 1 M NaOH solution and 1 M HCl solution to get pH 3 and pH 5. This pH range was selected based on previous studies (Shakoor, M. B et al .2019; Abid.M. et al.2016) which demonstrated its significant impact on As (V) removal by other biosobents. Following pH adjustment, a mixture of alginate and mushroom powder beads was introduced into the solution.

E. Biosorption experiments

The bead dosage (2 g) was added to flasks containing 20 ml of Arsenic bearing solution with pH adjusted at pH 3 and pH 5. Flasks were shaken at 100 rpm at room temperature for 45 min, during which time the removal of As concentration was reached at the equilibrium stage [11]. Samples were filtered with cellulose membrane papers. Then, each sample filtrate of As was digested using the open digestion method. The metal concentrations in the filtrate were determined by AAS (7000 AA).

F. FTIR characterization

Dried metal-loaded and metal-free (control) beads were separately prepared for FTIR analysis. Firstly, the dried bead sample was ground with a mortar and pestle to fine powder. Equal amounts of 2.0 mg from each sample were taken and then mixed with approximately 200.0 mg of potassium bromide. Each potassium bromide treated control and sample were again ground to fine powder with a mortar and pestle and pressed under vacuum into pellets, which were then analyzed using FTIR.

G. Statistical analysis

The statistical analysis on the effect of the variables on the sorption capacity of the biosorbent was carried out with the statistical software R (version 3.0) by using the packages extrafont (0.19), ggpattern (1.0.1) agricolae (1.3-7), lubridate (1.9.3), forcats (1.0.0), stringr (1.5.1), dplyr (1.1.4), purrr (1.0.2), readr (2.1.5), tidyr (1.3.1), tibble (3.2.1), ggplot2 (3.5.0), tidyverse (2.0.0) and readxl (1.4.3). The data were trimmed to only the treatment variables and calculated as the percent reduction relative to the initial concentration of As.

III. RESULTS AND DISCUSSION

The effect of all combinations of the treatment variables is visualized in Fig. 2. It can be seen from the graph that the minimum removal effect was over 60 % and the maximum was nearly 90 %. The overall mean of As (V) removal was 77.4 %. Shapiro-Wilk normality test [15] of the data show approximate normality with the p value of 0.05158, indicating that the data follow the normal distribution.

A. Effect of pH

Effect of pH has significant contribution on adsorption study, especially when considering the interaction between a charged macromolecule and a charged surface [16]. pH influences the protonation state of functional groups on the biosorbent, which in turn affects arsenic (As) speciation and the overall aqueous chemistry.

This interplay is essential in determining how effectively As is sorbed onto the biosorbent[17].The acidity level of the solution influences the competition between hydrogen ions (H^+) or hydroxyl ions (OH^-) and metal ions for attachment to active sites on the biosorbent surface[18].

It had been reported that As (V) removal increased as the rise in pH from 3 to 6.5 in the experiments using charred orange peel and chemically modified wheat straw while the study using egg shell and java plum seed as biosorbant showed the maximum removal of As(V) observed at pH 4 and 5.3 [19]. Similar results were seen in this study, with the interval of pH 3 and 5, where the pH 5 showed the significant difference in increase of As(V) removal percentage with the p-value of 0.003 as shown in ANOVA analysis Table 1.

It was suggested that the increase might be due to the formation of strong bonds between the biosorbents and $H_2AsO_4^-$ and $HAsO_4^{2-}$ ions, which replace -OH/-COOH ligands on the surface of biosorbents. [20,21].

B. Effect of bead composition

The removal efficiency of arsenic ions is also influenced by the chemical composition of the beads. Variations in the amount of biopolymeric components within the beads can change the number and nature of active sites, thus affecting the adsorption process. [16].

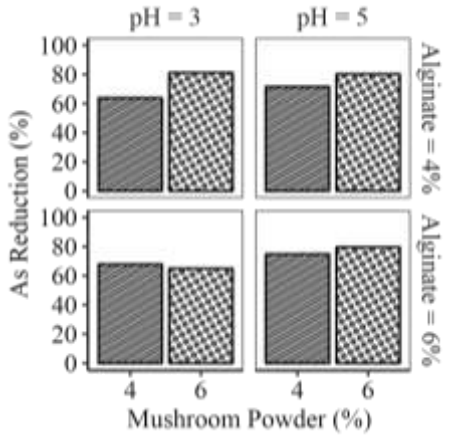
It was stated that with the increase of alginate concentration from 0.8 % to 2%, the increased soprtion of As (V) was observed. In the present study, the alginate concentration used in the formation of bead mixed with the mushroom powder were 4% and 6%, and the ANOVA analysis between these two concentrations showed no significant difference with the p-value of 0.519010 (Table 1). This might be due to the compactness of bead caused by the inclusion of the high percentate of alginate, leading to inaccessibility of the As (V) ions. Similar results had been reported by Tiwari, A. et. al [16]. and Khare, S. K.[22].

On the contrary, the composition of the Pleurotus ostreatus mushroom led to a significant increase with a p-value of less than 0.01 in the removal efficiency (Table 1) when the concentration of the powder was increased from 4% to 6% in the formation of alginate beads. This suggested that there is potential for further improvement and optimization of the removal process using mushroom powder.

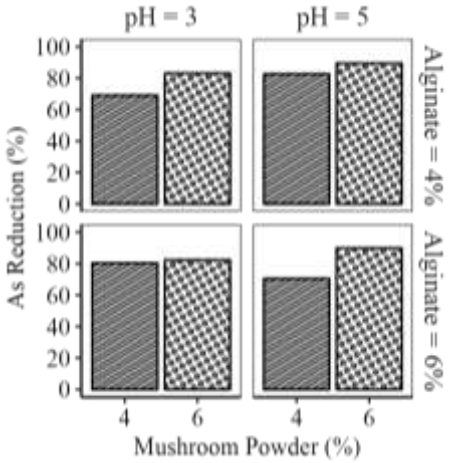
Assuming alginate within the range of 4%-6% caused no significant increase in the removal of As from the solution (Table 1), post hoc analysis was conducted for comparison and grouping of the means of the percentage of the removal efficiency of pH, the mushroom powder content and bead hydration status.

The results in the Table 2 showed that the beads with 6% of mushroom powder were in the same highest group at both pH level and bead hydration status with only a few exceptions, indicating the robustness that inclusion of the powder enhanced the removal efficiency regardless of the tested pH range and hydration status.

C. Effect of bead hydration state



(a)



(b)

Figure 2. Mean comparison of the Asensic removal percentage by (a) effect of fresh wet alginate-mushroom powder beads (b) effect of dried and swollen alginate-mushroom powder bead.

A study had highlighted the influence of hydration status on the dye removal efficiency of alginate beads incorporating zinc oxide. It was found that the removal efficiency of dry beads was higher at 78% compared to fresh beads at 64% [23]. Similar findings were observed in our study. A comparison between the effects of freshly prepared beads and dried and re-hydrated beads on As(V) removal efficiency revealed a significant difference with a p-value less than 0.01 (Table 1). This might be due to the fact that the decrease in mass due to water loss in the dried beads resulted in a higher quantity of dried beads being utilized in the treatment process. Additionally, the smaller size of dried beads provides a greater contact surface area with As(V) ions for adsorption.

	Df	**Sum Sq**	**F value**	**Pr(>F)**
Bead status (Wet/Dried)	1	406.1	16.320	0.000447 ***
PH (3, 5)	1	267.5	10.752	0.003060 **
Alginate (4%, 6%)	1	10.6	0.428	0.519010
Mushroom (4%, 6%)	1	590.6	23.735	5.19e-05 ***
Residuals	25	622.1		

Table. 1. Analysis of Variance (ANOVA) for the comparison of As removal effect of the bead status, pH, alginate percentage and mushroom percentage.

Signif. Codes: 0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘

Bead status	pH	Mushroom powder %	Removal efficiency (Mean ± sd %)
Dried	5	6	89.5953 ± 0.8554 a
Dried	3	6	82.6700 ± 0.6422 ab
Fresh Wet	5	6	79.9618 ± 0.6409 ab
Dried	5	4	76.5278 ± 7.1024 bc
Dried	3	4	74.6910 ± 6.619 bc
Fresh Wet	5	4	73.7337 ± 1.9661bc
Fresh Wet	3	6	73.0666 ± 9.3892 bc
Fresh Wet	3	4	65.2108 ± 2.4008 c

Table .2. Tukey HSD pair-wise comparison of the effect of significant factors on As removal

Band (peak) assignment for infrared spectra									
Before adsorption of arsenic (cm^{-1}) with alginate and mushroom beads	After adsorption of arsenic (cm^{-1})				After adsorption of arsenic (cm^{-1})				Assignment
	pH 3				pH5				
	B1	B3	B6	B7	B2	B4	B5	B8	
3292	3252	-	3281	3391	3385	-	3304	-	O-H stretching
2924	2924	2928	2928	2891	2924	2926	-	2905	C-H stretching
1612	1630	1634	1632	1651	1624	1638	1626	1645	N-H bending
1450	1423	1421	-	1425	1421	1421	1420	-	C-H bending
-	-	1383	1383	-	-	1383	1385	1385	C-H bending
1138	1151	1153	1151	1153	1151	1155	1150	-	C-N stretching
1093	1040	1040	1040	1042	1034	1036	1038	1084	C-H stretching
756	893,818	891	818	822	893	895	814	-	C-metal stretching
657	662		660/602	663/606	---		-	662	C-metal stretching

Table 3. Position of the main peaks in FTIR spectra of sodium alginate and mushroom bead before adsorption and after absorption of AS

B1 & B8 = alginate 4% and mushroom powder 4% beads, B3 & B4% = alginate 4% and mushroom powder 6% beads,

B2& B7=alginate 6% and mushroom powder 4% beads, B5& B6=alginate 6% and mushroom powder 6% beads.

D. FTIR characterization

A number of studies have suggested that the removal mechanism is driven by the presence of (-OH) and (-COO-) group. FTIR spectra confirms the presence of OH and COO groups in the synthesized beads, which are assumed to be responsible for the uptake of the metals. The functional groups mostly responsible for the adsorption of Pb, Cr, Cd metals were identified as hydroxyl (-OH) and carboxyl (-COO) groups [24]. Any presence of alkyl and $-NH_2$ group which are assumed to be involved in adsorption of As (III) and As(V). Gao et al. 2019 suggested the main sorption process in case of alginate beads are the exchange of the toxic metals (Pb, Cr, Cd) with the cations available on beads surface [25].

Alginate and mushroom are polysaccharide. The main FTIR character of polysaccharide is a strong and broad band at 3600 – 3000 cm^{-1} regions (Table 3). These intense band corresponds to the stretching vibration of abundant OH and NH in polysaccharide. In addition, moderate bands located at 3000 – 2500 cm^{-1} region can be assigned to the symmetric stretching vibration of abundant CH and CH_2 group [26]. At 1800 – 1500 cm^{-1} region is COO- stretching vibration of double bonds and NH bending vibration. 1500 –1200 cm^{-1} region is CH_2 and C-OH stretching vibration region [27]. At 1200–800 cm^{-1} region is CO bonding vibration for β (1-3) glucan polysaccharide.

A mixture of various bands of alginate and mushroom powder beads of FTIR spectrum have occurred the main character of polysaccharide vibration mode. After adsorption of arsenic metal solution, these

polysaccharide bead FTIR spectrum compare with pure polysaccharide bead.

The peaks of FTIR spectrum of the polysaccharide absorbed by arsenic metal solution are slightly lower than the original peak. However, it is in the finger print region on treatment with As, the hydroxyl group and NH group gets involved in metal – oxygen binding as evident by the band shift to lower wave number, indicating the role of hydroxyl group in metal binding [28,29]. Secondly chelating of As with NH functionally resulted in weakening of the NH bond strength converting it into bonded NH group and causing a downshift of the stretching as well as the bending absorption bands. IR spectrum of polysaccharide showed a band at 1612 cm^{-1} which is due to C= O stretching vibration of ester carbonyl which shift is 1634 cm^{-1} -1638 cm^{-1}. This is indicated that the COOH group is chelating with metal ions [30,31].

In the FTIR spectrum of a chelate involving arsenic and alkyl group, stretching vibration of peak appear in range at 400 -700cm^{-1}. If there are direct bonds between arsenic and carbon atom in the alkyl group. These peaks can be found in the region of 500-700cm^{-1}.

IV. CONCLUSION

The study investigated the adsorption of arsenic ions (As(V)) using alginate beads incorporating mushroom powder as a biosorbent. The effect of pH, bead composition, and bead hydration state on the removal efficiency of As(V) was analyzed. The pH of the solution significantly affected the removal efficiency, with pH 5 showing a significant increase in As(V) removal percentage. This result aligns with previous studies and suggests the formation of strong bonds between the biosorbents and arsenic ions at pH 5. The chemical composition of the beads, specifically the concentration of mushroom powder, played a crucial role in the removal efficiency. Increasing the concentration of mushroom powder from 4% to 6% led to a significant increase in removal efficiency. However, increasing the alginate concentration from 4% to 6% did not show a significant difference, possibly due to the compactness of the beads at higher alginate concentrations. The hydration state of the beads also affected the removal efficiency, with dried beads showing higher removal efficiency compared to fresh beads. This could be attributed to the smaller size of dried beads, providing a larger contact surface area for adsorption. FTIR analysis confirmed the presence of hydroxyl (-OH) and carboxyl (-COO) groups in the synthesized beads, which are assumed to be responsible for the uptake of arsenic ions. In addition, any presence of alkyl and -NH_2 group that involved in adsorption of As. This aligns with previous studies suggesting that these functional groups are crucial for adsorption. In conclusion, the study demonstrates the potential of alginate beads incorporating mushroom powder as an effective biosorbent for arsenic removal. Further optimization of the biosorbent composition and hydration state could enhance the removal efficiency, making it a promising approach for arsenic remediation in contaminated water.

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