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Application of activated carbon fiber (ACF) for arsenic removal in aqueous solution

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Abstract−The adsorption of arsenic from aqueous solution using activated carbon fiber (ACF) was investigated. Several series of experiments were conducted to investigate the effect of operating parameters such as equilibrium time, flow rate, and initial concentration that affect to the adsorption rate. Average removal efficiency was 24% for the initial arsenic concentration of 10 mg/L to 17 mg/L. Breakthrough point in ACF unit reached at 5 hours of the experimental operation for aqueous solution containing arsenic. Adsorption capacity of the filter was found to be 0.18 mg/mg of ACF. With the increase of flow rate there was a slight increase in the removal of arsenic. Field tests from thirty contaminated sites in Sonarang, Bangladesh have shown that the arsenic removal efficiency was only 13.0% in the single ACF unit, while it was increased upto 24.6% in two ACF units in series. Among the several fitting regression curves tested, three dimensional non-linear regressions gave over 90% fitting, while for other linear regression curves it was in the range of 5 to 92% depending upon the various operating parameters. Non-linear models described the relationships of C with C_0 and t better than the linear ones, and this model gives a good generalization of the kinetics of arsenic in ACF for the laboratory tested ranges.

Key words: Activated Carbon Fiber, Adsorption, Arsenic, Regression Curve, Aqueous Solution

INTRODUCTION

Arsenic contaminants in drinking water have been recognized as a serious environmental problem. Arsenic contamination in drinking water was found in the areas where water is extracted from groundwater with geological regions containing arsenic. But there are some cases of contamination from industries and mining as well [1]. Bangladesh and the part of Bengal in India are suffering from arsenic contaminated drinking water. Exposure to the concentration above its limit causes serious effects on the human body, affecting the skin, liver, nervous system, lungs, kidney, and gastro intestinal tract. Effect of arsenic in human body includes vomiting, jaundice, kidney failure and cancer [2]. Compounds of arsenic are widely used in wood preserving industries, and some percentage in agriculture, glass industries, and electronic industry [3]. There are number of treatment methods for the removal of arsenic from water and wastewater. Chemical precipitation, ion exchange, ultrafiltration, membrane techniques, lime softening and microbiological processes are the methods used for the treatment of water and wastewater containing arsenic [4]. But the anaerobic process may be inhibited in chemical precipitation. Although reverse osmosis and ion exchange methods are effective in removing such pollutants, they are expensive in the operational procedure. These factors have limited the use of methods for the removal of arsenic and other toxic pollutants from water and wastewater specially in most of developing countries [5,6].

Activated carbon fiber (ACF) has been widely applied to water purification systems in recent years [7]. ACF effectively adsorbs and removes a broad spectrum of harmful substances [8,9]. It has become popular for use in water purification because of its higher adsorption rate and capacities compared to the other granular adsorbents, such as granular activated carbon (GAC) or powdered activated carbon (PAC). These works have shown that the adsorptive capacity of ACF was ten to hundred times higher than granular activated carbon (GAC) [10,11]. ACF is especially interesting for adsorption since it has a uniform micropore structure, faster adsorption kinetics and a lower pressure drop than GAC. The adsorption kinetics and capacities for ACF are 10 to 100 times higher than these traditional adsorbents. It is thought that ACF's faster adsorption rate is due to its higher surface area compared to GAC, which arises from ACF's uniform microporous structure and graphite-like molecular characteristics [8,12,13]. ACF has been widely used in the industries of chemistry, pharmacy, and environmental protection for adsorbing toxic molecules. Its performance is particularly effective in shielding the attack of toxic chemical agents, making it ideal for nuclear, biological and chemical (NBC) protective clothing and filters [14,15]. ACF also offers a suitable system for the removal of toxic cations from different streams of water and wastewater. Due to its high microporous structure, it has a higher surface area and adsorption rate than GAC [16,17]. For the extensive use of ACF in the removal of toxic substances and arsenic, it is necessary to know the optimized level of operational parameters. In this study, several series of experiments were conducted to investigate the performance of ACF for arsenic removal and to study the kinetics of arsenic in ACF. To optimize the system, initial concentration, flow rate and operational time were taken as the parameters for finding the removal efficiency. Mathematical models for arsenic on ACFs were also conducted using Statistical Package for Social Scientists (SPSS) to simulate the experimental results performed.

EXPERIMENTAL SET-UP

In the ACF system, two series of ACF filters were used and a cartridge filter of 10 micorn was installed prior to ACFs in order to

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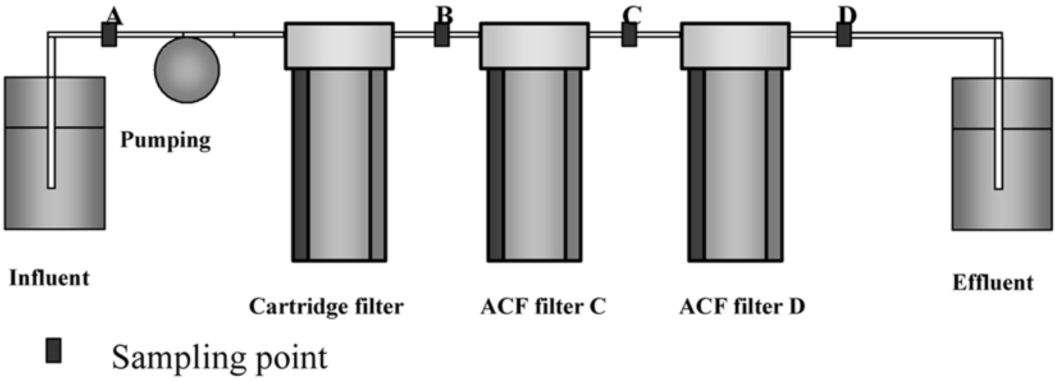


Fig. 1. Experimental set-up of ACF unit for arsenic removal from wastewater.

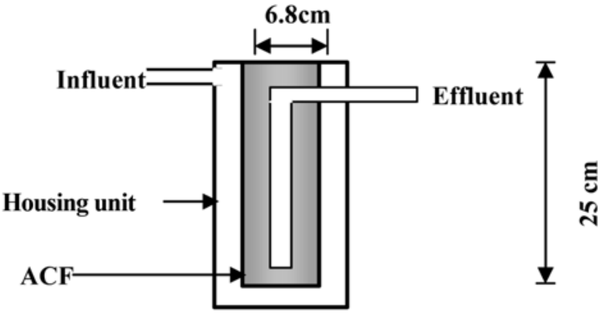


Fig. 2. Detailed apparatus of activated carbon fiber (Model FMC-250 A; Mfg: KOREA ACF).

Table 1. Characteristics of ACF

No. of fibers	103
ACF BET surface area	1,000 m^2/g
Weight of ACF	30 g /cartridge

expand the lifespan of ACFs. The sizes of used ACF are as follows: length=25.4 cm, outside diameter=6.8 cm, inside diameter=2.8 cm, thickness=2 cm. Total mass of ACF per filter was 120 mg. Fig. 1 shows the experimental setup of ACF unit for arsenic removal from wastewater. Details of the apparatus are shown in Fig. 2.

Synthetic solution of arsenic was prepared for the experimental operation. Initial concentration, flow rate and operation time were used as the parameters for optimizing the system during the experimental period. Experiments were carried out for 6 hours and the samples were collected every half-hour interval. Samples were analyzed by atomic absorption method for measuring lead contained in the effluent. SPSS and curve fitting software were used for the simulation of the adsorption and regression models [18,19]. Arsenic contaminated water was collected from thirty contaminated sites from Sonargong, Bangladesh and filtered through the ACF system in order to know the efficiency of the system in the removal of arsenic. Characteristics of membrane and used ACF in this experiment are summarized at Table 1.

RESULTS AND DISCUSSION

1-1. Effect of Operational Time on Arsenic Removal

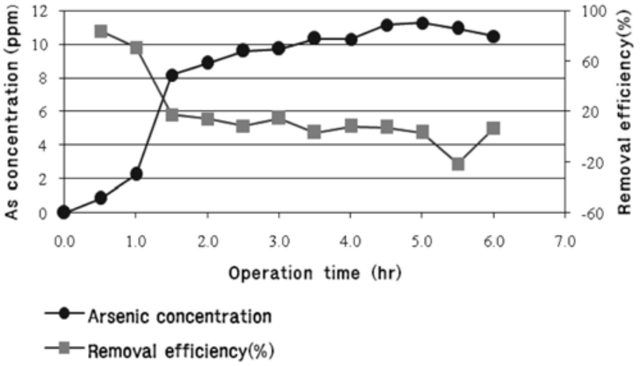


Fig. 3. Arsenic removal in ACF with time (single ACF unit).

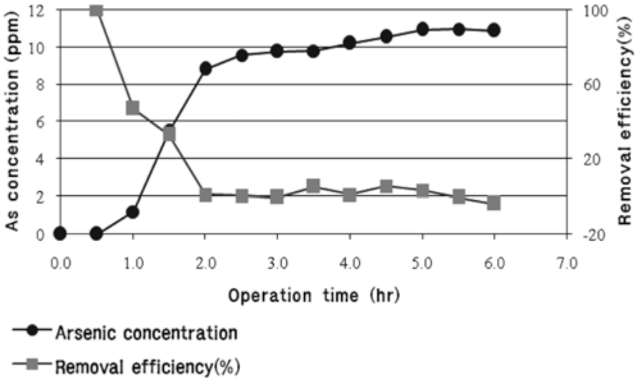


Fig. 4. Arsenic removal in ACF with time (double ACF unit).

Experimental results have shown that the breakthrough of ACF was reached at the experimental operation of 5 hours for the first ACF unit. It was oscillating adsorption and desorption for the second ACF. Maximum removal capacity of the single ACF filter was found to be 22 mg/cartridge or 0.18 mg of As per mg of ACF. In the previous study conducted at the similar experimental conditions using Kaolinite, Montmorillonite, and Illite, the maximum adsorption capacity was calculated by fitting the Langmuir equation to the adsorption isotherms and found to be 0.86, 0.64, and 0.52 mg As(V)/g, respectively [1,20]. Fig. 3 and Fig. 4 show the efficiency of ACF in arsenic removal with respect to the operational time in the single and double ACF units, respectively.

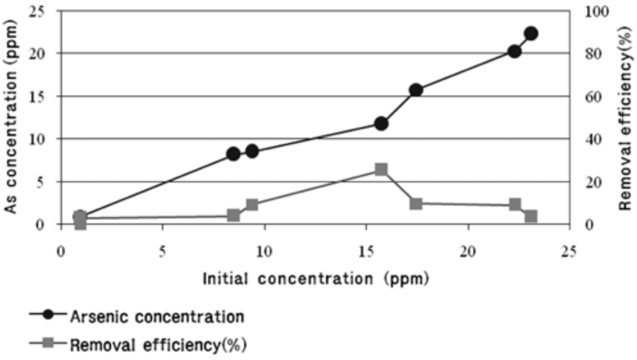


Fig. 5. Arsenic removal in ACF with C_0 (single ACF unit).

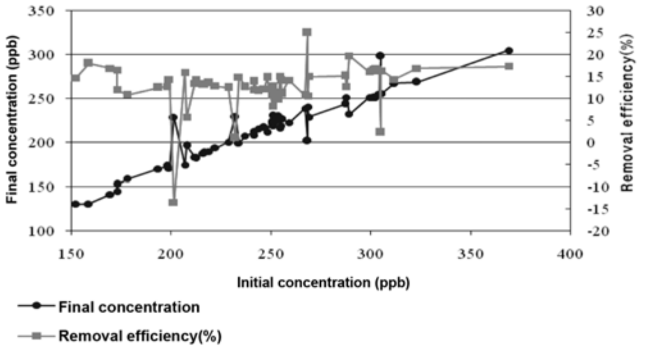


Fig. 7. Arsenic removals from groundwater samples using single ACF filter system.

1-2. Variation of Removal Efficiency with C_0

Fig. 5 indicates the arsenic removal efficiency of ACF with respective to different initial concentration. It was shown that the removal is the highest within the range of 10 to 17 mg/L. The highest removal was found at the concentration of 17 mg/L and it was 22%. In the previous study using various types of clay minerals, the arsenic removal efficiency was investigated as a function of solution pH, time, arsenic concentration and temperature. Arsenic mobility was investigated by determining the arsenic released from the loaded samples by leaching with various aqueous solutions. The kinetics of adsorption was observed to be fast, reaching equilibrium within 3 hours, while the breakthrough point in the ACF unit in this study was reached within 5 hours. Arsenic adsorption on the tested clays was pH dependent and maximum adsorption was achieved at pH 5.0 [1,20].

1-3. Variation of Removal Efficiency with Q

It was found that the arsenic removal increases steadily with the increase of flowrate, although the increase was minimal. Arsenic concentration was in the range of 8.0 to 8.9 mg/L. Fig. 6 demonstrates the relationship between initial concentrations of arsenic on its removal in the ACF system. It may be attributed to the specific properties of ACF. In fact, activated carbon fibers are recognized as efficient catalysts or supports for the catalytically active phase in heterogeneous catalysis. The use of carbon materials as supports for the active phase is favored because of their developed specific surface area, the possibility of producing the materials with homogeneous surface properties, sufficient strength, heat and chemical stability and their resistance towards the active metal phase. Among the other advantageous properties of carbon supports in comparison to commonly used mineral ones are heat conductance, which facilitates homogeneity of the temperature regime of catalytic processes, and electrical conductance capable of promoting electrocatalytic processes. The use of the fibrous forms of carbon supports has extra advantages. The catalytic activity of the element-carbon fibers depends on the relevant processing route [16,21].

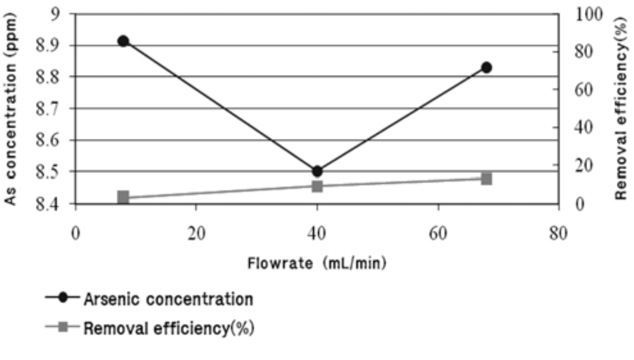


Fig. 6. Arsenic removal with flow rate in Cartridge.

2. Field Test Results

Experimental results for the water contaminated with arsenic from different sources in Sonargong, Bangladesh are presented in Figs. 7 and 8. These figures indicate that the removal was only about 13% for single ACF filter system, while it was about 24.6% for two ACF units in series. Obviously, the acceptable limit of 0.05 ppm of arsenic in drinking water was not achieved from the system.

To compare the removal pattern and efficiency of arsenic with lead (Pb), which can be easily found with arsenic in the contaminated soil, a series of experiments were conducted in a similar manner. Lead may cause serious effects on health by ingestion or inhalation as in the case of arsenic. Long-term exposure to lead can cause kidney damage, high blood pressure and birth defects [6]. Children could show delays in physical and mental growth with exposure to it. Besides inhalation and ingestion, it can enter human body through the food chain with uptake of the food contaminated with lead. In case of lead, there was 100% removal up to 8 ppm of initial concentration. But with the increases of the initial concentration the removal decreased sharply after that point. By varying the flow rate, the removal efficiency also changed. Up to the flow rate of 40 ml/min the removal efficiency was 100%. With the increase of flow

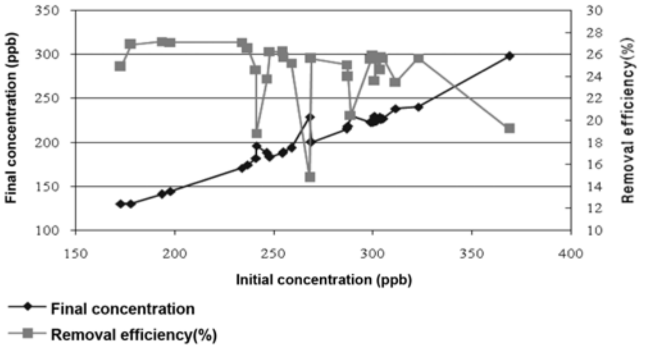


Fig. 8. Arsenic removals from groundwater samples using dual ACF filter system in series.

Table 2. Langmuir and Freundlich isotherm constants (Temp.= 298 ºK, contact time=48 hours)

Langmuir equation			Freundlich equation		
q_0	b	R_2	n	K_f	R_2
278	0.28	0.98	9.8	171	0.96

rate, however, the removal efficiency decreased sharply.

3. Adsorption Behavior of ACF on Arsenic

In the sorption technology, it is utmost essential to determine an adsorption, as it shows how the adsorption molecules are distributed in the liquid phase and with the adsorbent. For the design purpose, it is necessary to fit the isotherm data with the model. Langmuir and Freundlich isotherm equations are most widely used for the equilibrium study [6]. Following adsorption isotherm models were tested with the experimental results. Linear Langmuir equation considers the adsorption as homogeneous with negligible interaction between adsorbed species as shown in Eq. (1).

$$\frac{1}{q_e}=\frac{1}{q_0 b C_e}+\frac{1}{q_0} \tag{1}$$

Where,

q_0=Adsorption capacity (mg/g)
b=5Rate of adsorption (L/mg)

Experimental result was fitted into Eq. (1) by plotting $1/q_e$ versus $1/C_e$. Calculated isotherm constants b, q_0 and correlation coefficient R_2 are summarized in Table 2. The Freundlich isotherm equation assumes heterogeneous surface energies. It also assumes that the energy in the Langmuir equation is as a function of the surface coverage. Sites with stronger affinity are initially occupied. The second layer affinity depends upon the interaction between adsorbed molecules [14]. The Freundlich isotherm equation is given by the following Eq. (2).

$$\text{Log } q_e=\log K_f+1/n \log C_e \tag{2}$$

Where,

C_e=Equilibrium concentration (mg/L)
q_e=Amount of adsorbate adsorbed per unit mass of adsorbent (mg/g)
K_f=Maximum adsorption capacity (mg/g)
n=Dimensionless parameters

Batch experiments were conducted to determine the adsorption capacity of ACF. The Langmuir isotherm equation has shown a better correlation of the experimental results, as shown in Table 2.

Adsorption type was also evaluated using Eq. (3) as follows:

$$R_L=\frac{1}{1+b*C_0} \tag{3}$$

For low arsenic concentration the R_L value (0.012-0.023) lies in the range of 0 to 1, which implies favorable adsorption of arsenic onto ACF. For higher arsenic concentration, R_L is closer to zero indicating the possibility of irreversible fouling. Adsorptive capacity was found to be higher for the lower concentration due to its size in monomer form.

4. Mathematical Modeling

Mathematical models for arsenic on ACFs were prepared by using SPSS. The following three models were tested for determining statistically significant co-relations between different parameters and variables analyzed in the laboratory experiment [19].

4-1. Linear Two-dimensional Regression Models

R-square value for this model by using variation of t, C_0, and Q with C was found to be 0.63, 0.92 and 0.05, respectively. The result of the model is presented in Eqs. (4), (5), and (6), respectively, for time, initial concentration and flow rate variation.

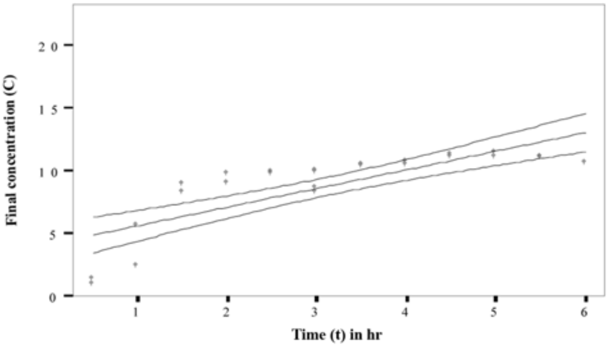
$$C=4.12+(1.48*t) \tag{4}$$

$$C=-0.86+(0.97*C_0) \tag{5}$$

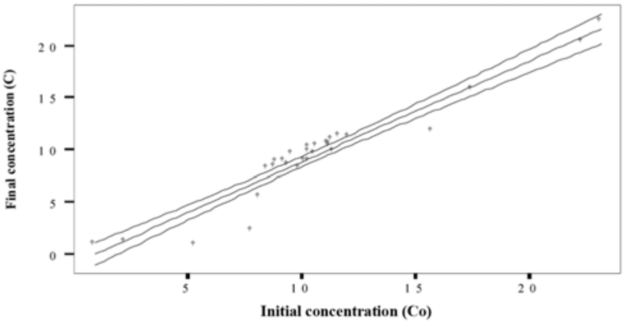
$$C=9.56-(0.01*Q) \tag{6}$$

Fig. 9 shows the result of the model for those three parameters. And for those three cases, the relationship was found to be non-linear. Linear models have been conveniently used for short time, low initial concentration and lower flow rate only.

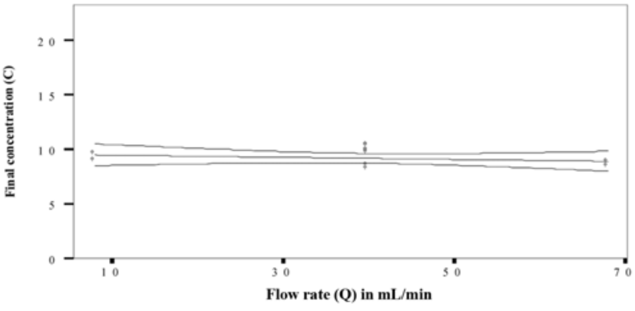
4-2. Non-linear Two Dimensional Models



(a) Variation of removal efficiency with t



(b) Variation of removal efficiency with C_o



(c) Variation of removal efficiency with Q

Fig. 9. Linear two-dimensional regression models.

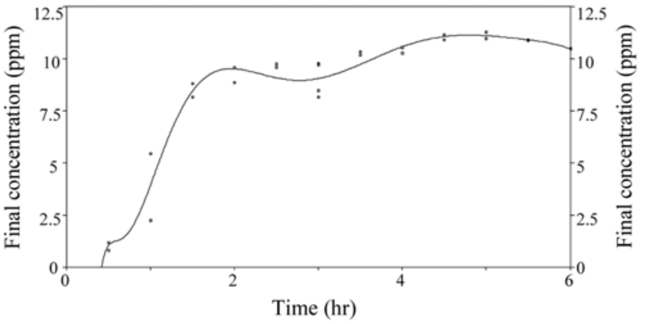


Fig. 10. Variation of final concentration of arsenic with time.

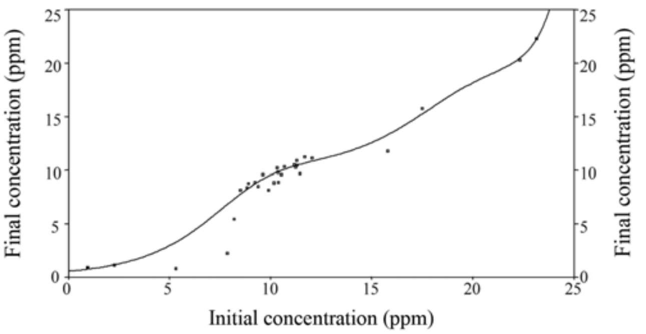


Fig. 11. Variation of final concentration of arsenic with its initial concentration.

Experimental results were tested for the non-linear two dimensional models as shown in Fig. 10 and Fig. 11. The plotted diagram clearly shows the non-linear relationship between final concentrations of arsenic with those parameters. This model is complicated and difficult to use day by day, but it can be well used for the tested range of those parameters. The result of the model for time dependent variation of final concentration is shown in Fig. 10. R-square value for this curve is found to be 0.95. Eq. (7) gives the appropriate equation for this model.

$$C=-1777.858+(11017.685*t^{0.5})-(28702.601*t)+(41030.162*t^{1.5})-(35232.401*t^{2})+(18667.411*t^{2.5})-(5981.4038*t^{3})+(1063.4861*t^{3.5})-(80.597417*t^{4}) \quad (7)$$

Testing of the non-linear two-dimensional models was also carried out for the experimental results of variation of initial arsenic concentration. Fig. 11 shows the relationships between final concentration and initial concentration of arsenic for that model. It clearly shows the non-linear relationships between them. R-square value for this curve is found to be 0.95. Eq. (8) gives the appropriate equation for this model.

$$C=1.7750125-(0.53577255*C_0)+(0.06793158*C_0^2)-(0.0042502313*C_0^5)+(0.00013004694*C_0^4)-(1.5571253*C_0^5) \quad (8)$$

Modeling result for the flow rate dependency of final concentration has shown that there is a poor fitting of the curve, so no more non-linear model is proposed in this study.

4-3. Linear Regression Models

The model was tested with 95% confidence interval of independent variables such as final concentration (C) with independent variables, initial concentration (C_0), flow rate (Q), total amount of arsenic removed in mg (X), and removal efficiency in percentage (E). For the independent variables of C_0, Q, t, and X, the R-square value is 0.94, while it is only 0.93, 0.93 and 0.90, 0.94 for the variables of C_0, t, and X; C_0 and t; C_0; C_0, t and Q, respectively. Eq. (9) to Eq. (13) was developed for those three variables from the model.

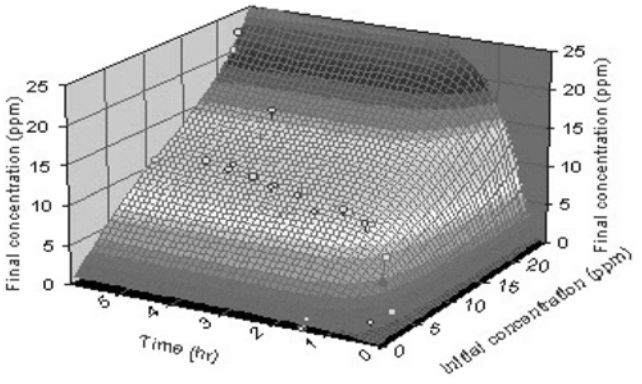


Fig. 12. Variation of final concentration of arsenic with C_0 and t.

$$C=0.21+(0.778*C_0)-(0.0294*Q)+(0.775*t)-(0.0188*X) \quad (9)$$

$$C=-0.924+(0.786*C_0)+(0.74*t)-(0.0188*X) \quad (10)$$

$$C=-0.986+(0.778*C_0)+(0.714*t) \quad (11)$$

$$C=-0.545+(0.962*C_0) \quad (12)$$

$$C=-0.148+(0.77*C_0)+(0.75*t)-(0.0294*Q) \quad (13)$$

4-4. Non-linear Three-dimensional Models

Fig. 12 shows the three-dimensional model generated to describe the final concentration of arsenic in relation to C_0 and t. This model gives good generalization of the findings in the laboratory analysis. R-square value for this model was found to be 0.98.

Eq. (14) for this model is represented by;

$$\ln C=12.17008-(9.28845*C_0)+(1.311707*C_0*\ln C_0)+(7.326476*C_0^{0.5}*\ln C_0)-3.04297/C_0^{0.5}+(0.86098*t^{0.5}*\ln t)-\{1.04763*(\ln t)^2\} \quad (14)$$

This model gives a good fitting for the laboratory and field tested data. An average of 10 percent deviation was observed for the model generated data and actual field findings. Thus, the model gives a good generalization of the kinetics of arsenic in ACF for the laboratory tested ranges. Activated carbon fiber is recognized as efficient catalyst or support for the catalytically active phase in heterogeneous catalysis [22,23]. The use of carbon materials as supports for the active phase is favored because of their developed specific surface area, the possibility of producing the materials with homogeneous surface properties, sufficient strength, heat and chemical stability and their resistance towards the active metal phase. Among the other advantageous properties of carbon supports in comparison with commonly used mineral ones are heat conductance, which facilitates homogeneity of the temperature regime of the catalytic processes, and electrical conductance capable of promoting electrocatalytic processes [21,24]. The use of fibrous forms of carbon supports has extra advantages. Catalytic activity of the element-carbon fibers depends on the relevant processing route [25,26].

CONCLUSION

The maximum removal of arsenic in tested ACF filter was 0.18 mg/mg ACF. For the initial concentration of 10 to 17 mg/L, the removal efficiency was found to be 24% in two ACF units in series, while it was only 13% for single ACF unit. ACF alone cannot be used as a stand-alone system for arsenic removal from groundwater. Non-linear models have shown higher accuracy compared to a two-dimensional linear model. They are complicated and uncertain in generalization and difficult to use day by day. With the increase of initial concentration and operational time, the effluent arsenic concentration also increased. From the various results of arsenic models, it was clear that non-linear models described the relationship of C with C_0 and t better than the linear one. Final concentration with the flow rate has shown the poor curve fittings, so no non-linear model is proposed. Raw data has shown the increase of adsorption with the increase of flow rate. For the kinetics of arsenic, three-dimensional non-linear models gave higher accuracy, and R-square value was found to be 0.98. The best model equation for the kinetics of arsenic is represented by the relevant equations.

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