

## Elimination of Nickel from Aqueous Solution Using Activated Carbon and Biofilm

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### Abstract

The main of this research was to study of the efficiency of using granular activated carbon (GAC), Biofilm and biological activated carbon (BAC) columns to treat low concentration of Nickel bearing water streams and the effects of temperature and pH on the adsorption isotherms. Studies were conducted to delineate the effect of pH, temperature, initial Ni and adsorbent concentration on adsorption of Ni<sup>2+</sup> by GAC, BAC and Biofilm. Breakthrough curves for removal of 0.5 mg/L Ni<sup>2+</sup> by GAC, Biofilm and BAC columns at two contact times were plotted. Batch adsorption and column data are compared, pH is shown to be the decisive parameter in Ni removal for GAC but not for BAC or biofilter. Lagergren plots confirms applicability of first-order rate expression for adsorption of Ni<sup>2+</sup> by GAC, BAC and Biofilm. The adsorption coefficient (K<sub>ad</sub>) for BAC were 2-3 times greater than those with plain GAC. Bed Volumes of water containing 0.5 mg/L Ni<sup>2+</sup> treated at breakthrough for GAC, Biofilm and BAC columns were 45 ml, 85 ml and 180 ml of Bed Volume respectively. BAC is more efficient than GAC in the removing of Ni from water environment.

**Keywords:** Nickel; adsorption; activated carbon; Biofilm

### Introduction

The chronic toxicity of nickel to humans and environment has been well documented, for example, high concentration of nickel (II) causes cancer of lungs, nose and bone [1].

The drinking water guideline value recommended by World Health Organization (WHO), is 0.02 mg Ni/L. Low concentration (less than 2 mg/L) of Nickel is difficult to treat economically using chemical precipitation methodologies. Ion exchange and reverse Osmosis while can guarantee the metal concentration limits required by regulatory standards, have high operation and maintenance costs [2].

Although the ability of activated carbons to remove Nickel in high concentrations from wastewater has been established by numerous researchers [3-9], very few articles are available on the use of activated carbon to remove Nickel in low concentration from contaminated surface or subsurface waters [10]. Activated carbon has been an effective adsorbent for the removal of many organics substances in water, its use for metal removal from water is rather rare. Several reports of Nickel removal from aqueous solutions by biosorption with micro-organism generated biomass have been published [11].

The underlying objective behind using GAC as a support for biofilm has been, therefore, to provide the foundation for remediation processes that can provide metal biosorption concurrently with removal of non-metal contaminants such as organic compounds. J.A. Scott and A. M. Karanjkar studied Nickel (in high concentration) adsorption on to Biofilm covered Granular Activated Carbon [7-11]. There is not any study on removal of low concentration of Nickel by Biofilm/GAC. The objective of this study was to investigate the adsorption characteristics of Nickel on to plain (non-biofilm) GAC, Biofilm and Biofilm/GAC, and also was to determine the effects of temperature and pH on the nickel uptake by plain GAC and Biofilm/GAC.

### Materials and Methods

The granular activated carbon used in this study was Darco 12-20 mesh supplied by Aldrich. Carbon was washed with double distilled water and dried in an oven at 120°C for 24 hours. All the Nickel solutions were prepared using Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and the solution pH

was adjusted with HNO<sub>3</sub> and NaOH 0.01N. Experimental data for the adsorption isotherms were obtained as follows.

The bacillus circulans is used as biofilm on this study which is isolated from wetland region on the north east of Algeria.

A predetermined mass of plain GAC and Biofilm/GAC were contacted with a fixed volume of a Nickel solution of known initial concentration. The nickel solution remained in contact with adsorbent until equilibrium was reached. Batch sorption studies were performed at an ionic strength of 0.01 (added as NaCl) at different temperature (5°C, 15°C, 25°C) and at different pH (5, 7 and 8). The contact time were selected on the basis of preliminary experiments that demonstrated that the equilibrium were established in 4 hours for GAC and Biofilm and 1.5 hours for Biofilm/GAC. For isotherm studies, a series of 250 mL Erlenmeyer flask were employed. Each Erlenmeyer flask was filled with 100 mL adjusted pH of Nickel solution of varying concentration (0.25-0.5 – 1.0 – 2.5 and 5.0 mg/L). For each concentration 4 Erlenmeyer flask were employed. A known amount of adsorbent (plain GAC and Biofilm/GAC separately) (0.05- 0.1 – 0.15 and 0.2 gr) was added in to each Erlenmeyer and agitated for the desired time periods. After this periods the solution was filtered using Glass Fiber (GF/A) filter and analysed for the concentration of the metal ions remaining in the solution by Chem Tech Alpha 4 Atomic Absorption Spectrophotometer. Conditions for the Spectrophotometer were acetylene – air flame under oxidizing conditions at 228.8 nm wavelength.

Three columns including GAC, Biofilm and Biofilm/GAC were used in this study. The length of the columns was 52 Cm and inner

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diameter of column was 14 Cm. One column was packed with 12-20 mesh sand and this column is named as biofilm column. Another column was packed with 12-20 mesh GAC.

Seeded nutrient medium (2000 mg/L Sodium acetate as the sole carbon source, 500 mg/L NH<sub>4</sub>NO<sub>3</sub>, 500 mg/L KH<sub>2</sub>PO<sub>4</sub>, 200 mg/L CaCl<sub>2</sub> and 200 mg/L MgSO<sub>4</sub>) was circulated (upflow, 25 C, pH=7) for two days through GAC and sand columns.

Biofilm samples for batch biosorption test were detached and collected from the sand media. Nickel binding isotherms were produced by measuring the amount of nickel bound by biomass from solutions containing a range of Nickel concentrations. Eighty three (83) mg samples of biomass (dry weight) were mixed with 100 mL aliquots of aqueous Nickel solutions with Ni(II) concentrations of 0.2 – 0.5 – 1.0 – 2.0 and 5.0 mg/L. The mixtures were placed for six hours on a shaker to ensure that equilibrium was attained. The mixtures were then filtered through 0.45-micrometer membrane filter to remove the biomass. The final concentration of unbound Nickel was determined by atomic absorption spectroscopy and the metal loading on the biomass calculated.

After two days circulating of culture medium through sand and GAC columns, the culture medium was replaced with a solution containing 0.2 mg/L Ni(II) for uptake studies by Biofilm/GAC, Biofilm (sand column) and plain GAC columns. Columns were operated in the upflow mode. Effluent samples were collected from the columns and acidified and the concentration of Ni(II) was determined by atomic absorption spectroscopy.

## Results and Discussion

Calculated values of correlation coefficients (R<sup>2</sup>) at different pH value are given in Table 1. According to Langmuir model, reasonable straight –line correlations (R<sup>2</sup>) were achieved for Ni(II) adsorption by GAC and Biofilm, because R<sup>2</sup> for Langmuir isotherm were greater than for the Freundlich isotherm. For adsorption of Ni(II) by GAC/Biofilm, the correlation coefficients showed that in general the Freundlich model fitted the results better than the Langmuir model.

As illustrated in Figure 1, where adsorption isotherms of plain GAC, Biofilm and GAC/Biofilm is shown, biofilm immobilized over GAC clearly enhance the uptake of Ni(II). With regards plain GAC, Ni(II) uptake is generally low, but with biofilm immobilized over GAC particles, the Ni(II) uptake level can be increased 2 or 3 fold.

Figure 2 illustrates both the effectiveness of an immobilized biofilm in taking up Nickel (0.5 mg/L), along with the influence of solution temperature on equilibrium Ni(II) loading levels. That is, the presence of the biofilm, estimated at around 80 mg (dry weight) per gram of GAC, results in a 2 to 3 fold increase in Ni(II) uptake when compared to plain (non-biofilm) GAC. Furthermore, over a temperature rise of 5-24°C, the slight increase in metal uptake indicates physical adsorption, rather than metabolic activity as the prime factor in metal accumulation by the biofilm-GAC system.

The uptake of the Nickel by plain GAC increased with an increase in temperature thereby indicating the process to be endothermic.

pH	Langmuir Model			Freundlich Model		
	GAC	Biofilm	GAC/Biofilm	GAC	Biofilm	GAC/Biofilm
5.5	0.92	0.86	0.66	0.85	0.83	0.83
7.0	0.89	0.85	0.68	0.87	0.82	0.81
8.0	0.91	0.88	0.65	0.84	0.81	0.85

Table1: Freundlich and Langmuir isotherm correlation coefficients (R<sup>2</sup>) for adsorption of Ni (II) on GAC, Biofilm and GAC/Biofilm at different pH.

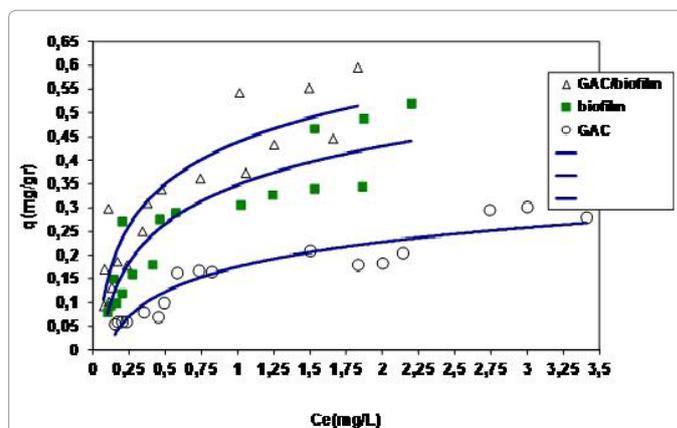


Figure 1: Isotherm plots for Ni(II) sorption by GAC, Biofilm and GAC/Biofilm.

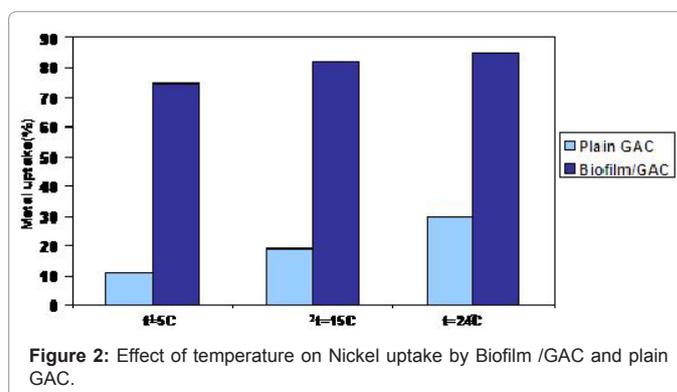


Figure 2: Effect of temperature on Nickel uptake by Biofilm/GAC and plain GAC.

Figure 3 shows the influence of solution pH on equilibrium uptake level. The experiments were carried out for pH values below the pH where chemical precipitation of the Nickel hydroxide occurs. In these condition, metal removal can be related only to the adsorption process. The adsorption of Ni(II) on the plain GAC increases with the increase in pH.

The increase in Ni(II) removal as pH increases can be explained on the basis of a decrease in competition between proton and Ni(II) for the surface sites and by the decrease in positive surface charge, which results in a lower coulombic repulsion of the sorbing Ni(II). For the Biofilm/GAC system alkaline condition (pH=8) was found to have little effect on Ni(II)

The Lagergren first-order rate equation is written as  $\text{Log}(q_e - q_t) = \text{Log} q_e - K_{ad} / 2.303 t$ .

Where  $q_e$  and  $q_t$  are the amount of metal adsorbed (mg/gr) at equilibrium and time “t” respectively. For adsorption of Ni(II) by Biofilm/GAC, a plot of  $\text{Log}(q_e - q_t)$  Vs “t” gives a straight line as can be seen in fig. 4, confirming the applicability of first-order rate expression. The adsorption coefficient ( $K_{ad}$ ) for GAC, Biofilm and Biofilm/GAC were calculated from the slope of the plots separately and the values are presented in Table 2.

The adsorption rate constants can be used for comparison between Biofilm/GAC, and GAC to adsorb Nickel from aqueous solution. The data indicates that with Biofilm/GAC, higher rate of adsorption can be achieved, because  $K_{ad}$  for Biofilm/GAC were 2-3 times greater than those with plain GAC.

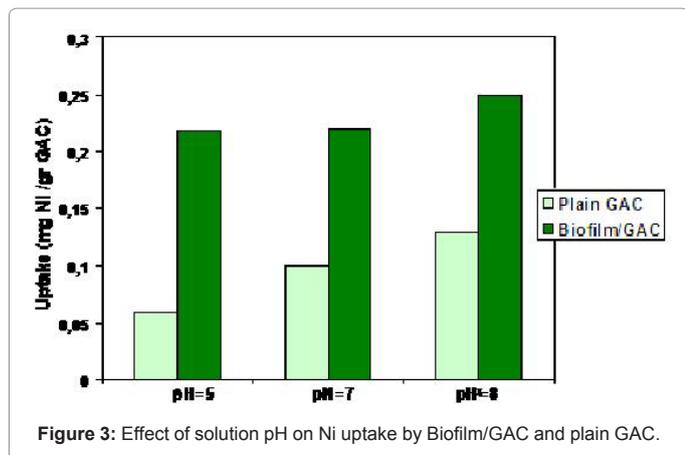


Figure 3: Effect of solution pH on Ni uptake by Biofilm/GAC and plain GAC.

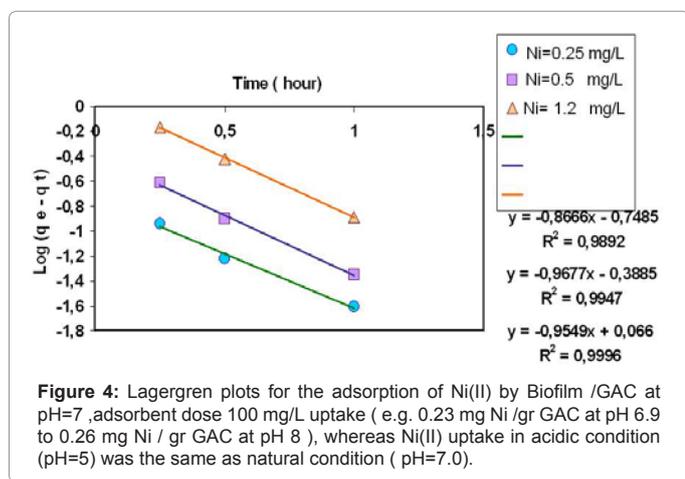


Figure 4: Lagergren plots for the adsorption of Ni(II) by Biofilm /GAC at pH=7, adsorbent dose 100 mg/L uptake (e.g. 0.23 mg Ni /gr GAC at pH 6.9 to 0.26 mg Ni / gr GAC at pH 8 ), whereas Ni(II) uptake in acidic condition (pH=5) was the same as natural condition ( pH=7.0).

Ni Concentration (mg/L)	K <sub>ad</sub> Biofilm/GAC (L/Min.)	K <sub>ad</sub> GAC (L/Min.)
0.25	2.19	1.04
0.5	2.22	1.10
1.2	1.99	0.68

Table 2: Calculated adsorption rate constants using GAC and Biofilm/GAC.

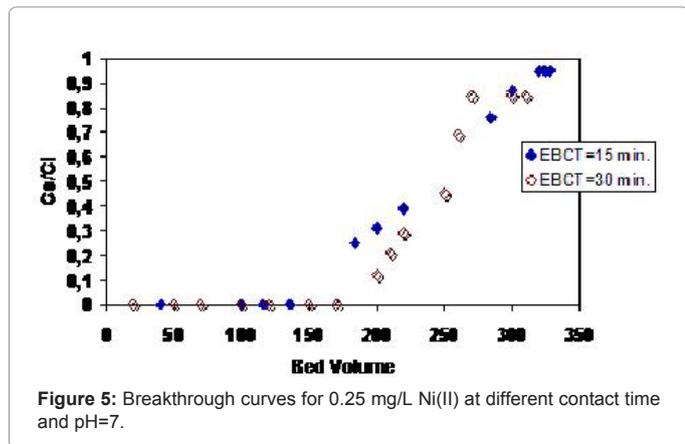


Figure 5: Breakthrough curves for 0.25 mg/L Ni(II) at different contact time and pH=7.

Normalized effluent Nickel concentration ( $C_e / C_i$ ) versus number of bed volumes (BV) treated for 0.5 mgNi/L by Biofilm/GAC column at pH=7, are presented in Figure 5.

$C_e$ : Concentration of nickel at equilibrium in mg/L

$C_i$ : Initial concentration of nickel in mg/L.

This curve will be referred to as breakthrough curve. Breakthrough was defined at  $C_e = 0.01 C_i$ . Breakthrough occurred at about 45 ml, 85 ml and 180 ml bed volume for plain GAC, Biofilm and Biofilm/GAC columns respectively.

The removal of nickel by a GAC column was increased by 400% when biofilm immobilized over GAC particles.

## Conclusion

Granular Activated carbon (GAC) is well known as an excellent adsorber of organic pollutants from contaminated water streams. GAC by itself is not in general, however, an effective adsorbent for heavy metals. Whereas, it has been shown that with a biofilm attached to the GAC surface, the uptake rate and quantity of metal ions extracted from solutions can be significantly increased. As a consequence, by immobilizing bacterial biofilms, metal removal can be combined with the adsorption of other contaminants such as organic residues.

Biosorption has the potential to provide economic metal decontamination of low concentration waste streams, but leaves the problem of metal-laden biosorbent disposal. There are, however, significant industrial and environmental process opportunities from metal impregnated over GAC surfaces, as they can usefully enhance surface activity. It is shown that it is possible to distribute metals over GAC by biosorption, through using attached biofilms. If the intention is to remove metals from contaminated streams, then ideally these biofilms should have a structure open enough not to negate the adsorption characteristics of the carbon surface for other contaminants, such as organic residues.

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