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Authors: Abdel–Gawad, Soha A, and Abd El–Aziz, Hossam M

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# Removal of Pharmaceuticals From Aqueous Medium Using Entrapped Activated Carbon in Alginate

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Soha A Abdel–Gawad[1] and Hossam M Abd El–Aziz[2]

[1]Department of Chemistry, Faculty of Science, Cairo University, Giza, Egypt. [2]Chemical Industries Development (CID) Company, Giza, Egypt.

**ABSTRACT:** The adsorption of entrapped activated carbon in alginate polymer (AG–AC) was investigated by measuring the removal of organic compounds. The general concept is that the entrapped activated carbon in alginate polymer could be used as a low–cost adsorbent for ascorbic acid and lactose removal from industrial wastewater. Ascorbic acid and lactose are the most pharmaceutical wastes that can introduce throughout the industrial process and lead to an increase in the amount of chemical oxygen demand (COD) in wastewater. Different ascorbic acid and lactose concentrations were prepared in the laboratory. The efficient removal is affected by external variables (eg, pH, contact time, adsorbent dosage, concentrations, and stirring rate). Percent removal for ascorbic acid and lactose at pH 3 using dose 30 g for 60 minutes with a fixed stirring rate at 100 rpm was about 70% and 50%, respectively. Ascorbic acid and lactose adsorption onto entrapped activated carbon in alginate polymer obey well with Freundlich adsorption isotherm.

**KEYWORDS:** adsorption study, entrapped AG–AC, pharmaceutical wastewater, removal of COD

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**CORRESPONDING AUTHOR:** Hossam M Abd El–Aziz, Chemical Industries Development (CID) Company, Giza, Egypt. Email: hossam_elywa@yahoo.com

## Introduction

Ascorbic acid and lactose are of the most used material in the pharmaceutical manufacturer, they perform to high values up to 10 times of chemical oxygen demand (COD) and biological oxygen demand (BOD),[1–3] when disposal in rivers and fields promote viruses, bacteria, or another microorganism that poses a serious potential threat to the environment or causes an increase in loading Processing Station so, it must get rid of them before dumping in sewage network.[4] Various techniques employed in wastewater treatment such as coagulation, flocculation, sedimentation, and aerobic and anaerobic biological contactor are complicated, expensive, and time–consuming. Therefore, researchers have been focused on an innovative approach that is addressed more appropriately in organic content removal from pharmaceutical wastewater.[5–7] Activated carbon has highly efficient adsorption capacity,[8–12] but the dispersion of its powder in treated media is difficult in removal. This issue can be solved through entrapment activated carbon in alginate polymer, which facilities and helps in adsorption and allows contaminated aqueous solutions to be in connection with the entrapped activated carbon.[10,13–19] It was considered one of the economical and effective process for wastewater treatment; thus, this process has aroused great interest during last years.[20–24]

The focus of this study was to investigate the feasibility of entrapped activated carbon in alginate polymer for the removal of ascorbic acid and lactose from aqueous solutions. In addition, the nonlinear Freundlich and Langmuir isotherms are used to fit the data, and it is noted that adsorption of ascorbic acid and lactose is better explained by the Freundlich model.

## Experimental

### Materials and reagents

All chemicals used were of the analytical reagent grade and of highest purity, such as activated carbon, sodium alginate ($[C_6H_7NaO_6]n$, Qingdao Bright Moon), ascorbic acid ($C_6H_8O_6$, Shandong Luwei), lactose ($C_{12}H_{22}O_{11}$, Dfe pharma), sodium hydroxide (NaOH, Riedel–de Haën), hydrochloric acid (HCl, Scharlau), nitric acid ($HNO_3$, Columbus), potassium dichromate ($K_2Cr_2O_7$, Prolabo), ammonium molybdate ($[NH_4]_6Mo_7O_{24}$, P.O.C.H., Polska, Poland), potassium hydrogen orthophosphate ($KH_2PO_4$, Fluka Garantie), stannous chloride ($TiCl_2$, Lobacheme), sulfuric acid ($H_2SO_4$, sever biotech), ethanol 96% ($C_2H_5OH$, World co. for sub & med industries), mercuric(II) sulfate ($HgSO_4$, Lobachemie), silver sulfate ($Ag_2SO_4$, LABSCAN), and calcium chloride ($CaCL_2$, Fisher Scientific). The pH value of the aqueous medium was adjusted using 0.1 M of NaOH or HCl solutions.

### Methods

*Preparation of adsorbent.* Adsorbent beads were prepared by adding 1 g of activated carbon to 2% of sodium alginate solution, stirred well. The mixture was added to a 5% $CaCl_2$ solution; the solution was filtrated and the adsorbent beads were taken out and washed for many times to be used in the adsorption, as shown in Figures 1 and 2.

*Preparation of standard solutions.* The standard concentrations were 500 mg\L against a blank prepared by dissolving separately 1 g of ascorbic acid in 1000 mL of distilled water and 0.5 g of lactose in 1000 mL of distilled water (expressed as

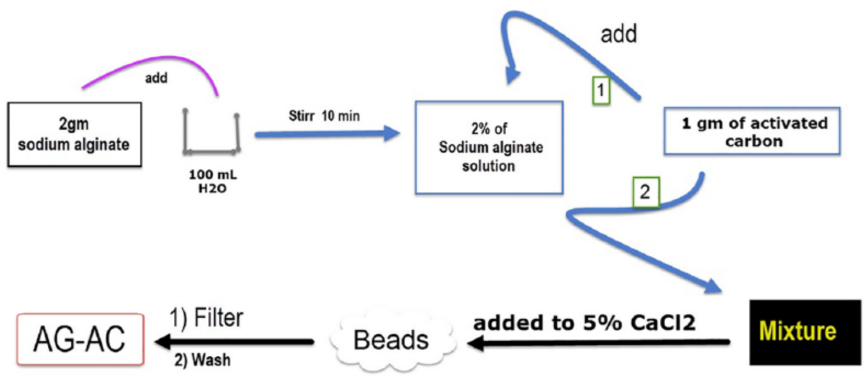


**Figure 1.** Preparation of alginate beads with entrapped activated carbon (AG-AC).

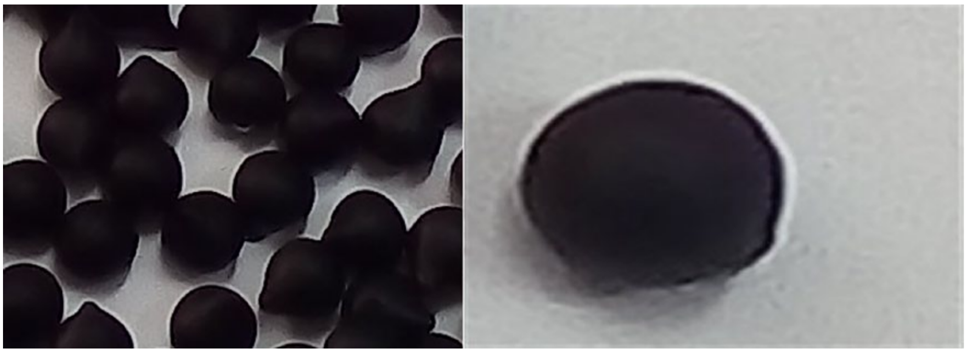

**Figure 2.** The characteristic and morphology of beads.

COD). Required concentrations of ascorbic acid and lactose solutions were prepared from the stock solution.

### *Adsorption isotherm models*

Langmuir and Freundlich isotherm models were used for describing the equilibrium adsorption data.[25,26] Langmuir assumes monolayer coverage of adsorbate over a homogeneous adsorbent surface. The linearized Langmuir isotherm model is given by the equation:

$$C_e / q_e = 1/(K_L q_{max}) + C_e / q_{max},$$

where $q_e$ (mg/g) is the mass of solute adsorbed per mass of adsorbent used, $C_e$ (mg/L) is equilibrium concentration of solute, $q_{max}$ (mg/g) is the maximum monolayer capacity of adsorption, and $K_L$ (L/mg) is the Langmuir constant related to binding sites affinity and adsorption energy. The plot of $C_e/q_e$ versus $C_e$ employed to generate the values of $q_{max}$ and $K_L$.

The Freundlich isotherm is an empirical equation employed to describe heterogeneous adsorption surface and is given by:

$$\text{Ln}\, q_e = 1/n \ln C_e + \ln K_f,$$

where $K_f$, (mg/g) $(mg/L)^{-1/n}$, and n (dimensionless) are Freundlich constants related to the adsorption capacity and adsorption intensity, respectively. $K_f$ and n are evaluated by plotting ln $q_e$ and ln $C_e$.

### *Procedure of adsorption experiment*

When entrapped activated carbon in alginate polymer is placed in a solution containing ascorbic acid or lactose which agitated for an adequate time, adsorption occurs. Aqueous solutions (100 mL) were used and the adsorption capacity was determined at different environmental conditions. According to the standard method for wastewater analysis ASTM 2005, a withdrawn sample from the solution after filtration was analyzed for dissolved ascorbic acid and lactose concentrations by spectrophotometry. Then the adsorption capacity and isothermal studies were conducted for Freundlich and Langmuir adsorption isotherm.

The percentage of removal was calculated using the following equation:

$$\text{Sorption}\left[\%\right] = \left[C_o - C_e / C_o\right] \times 100,$$

where $C_o$ is the initial concentration (mg/L) and Ce is the equilibrium concentration (mg/L) in solution. The amount of organic content adsorbed by entrapped activated carbon in alginate polymer was calculated using the following equation:

$$qe\left[mg/g\right] = \left[\left[Co - Ce\right]V\right]/m,$$

where $q_e$ is the equilibrium adsorption capacity (mg/g), V is the volume of aqueous solution (L) and m is the weight of the adsorbent (g).

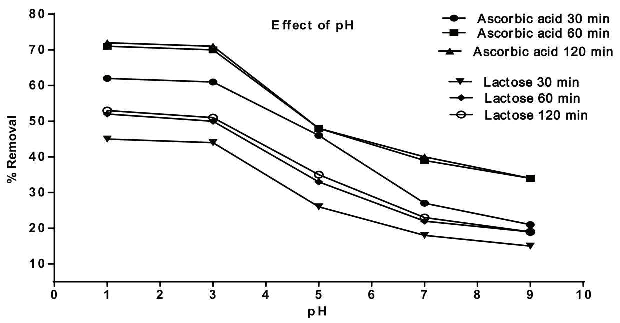


**Figure 3.** Effect of pH on ascorbic acid and lactose removal.

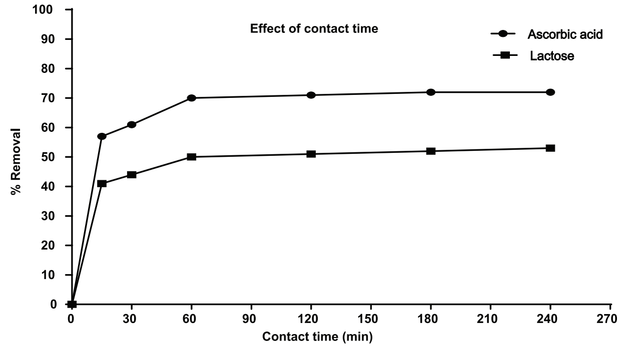


**Figure 4.** Effect of contact time on ascorbic acid and lactose removal.

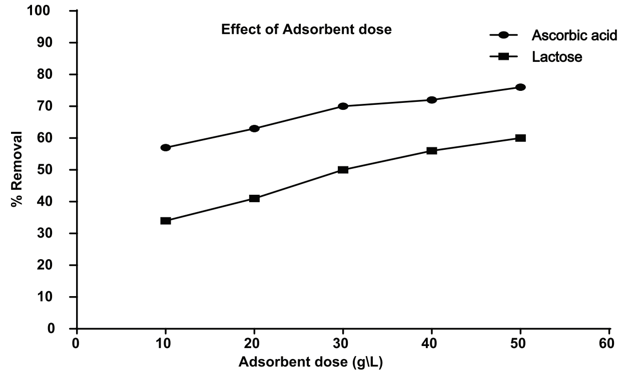


**Figure 5.** Effect of adsorbent dose on ascorbic acid and lactose removal.

## Results and Discussion

### *Effect of pH value*

The influence of pH value on the amount of ascorbic acid and lactose removed by entrapped activated carbon in alginate from the aqueous solution was estimated by carrying out experiments with different solution pH (1, 3, 5, 7, and 9) at different contact time (30, 60, and 120 min), and the conditions used were as follows: the initial ascorbic acid and lactose concentration was 500 mg\L, the adsorbent dosage was 30 g\L, and the stirring rate was fixed at 100 rpm; with ascorbic acid removal efficiencies of 62%, 61%, 46%, 27%, and 21%; 71% 70%, 48%, 39%, and 34%; and 72%, 71%, 48%, 40%, and 34%; and lactose removal efficiencies of 45%, 44%, 26%, 18%, and 15%; 52%, 50%, 33%, 22%, and 19%; and 53%, 51%, 35%, 23%, and 19%, respectively, and plots of the pH against the percentage of the ascorbic acid and lactose that was removed from the solution is shown in Figure 3. The optimum pH for the removal was 3. The pH of the aqueous solution plays a decisive role in affecting ascorbic acid and lactose adsorption. At lower pH levels, the removal sharply increased because the positively charged functional groups of organic molecules bind through electrostatic attraction to the negatively charged surface of the adsorbent. On the contrary, at higher pH the reduction in adsorption due to the increase of hindrance to organic ions diffusion because of that the abundance of $(OH)^{-}$ ions leads to repulsion between the organic molecules and the surface of the adsorbent.[27–34]

### *Effect of contact time*

The contact time is another important operational factor that affects removal efficiency. Figure 4 depicts ascorbic acid and lactose removal as a function of contact time. From Figure 2, it is shown that an increase in contact time increased removal efficiency. As shown, the ascorbic acid and lactose uptake by entrapped activated carbon in alginate was very rapid within the first 15 minutes. After 15 minutes, the uptake of ascorbic acid and lactose progressively decreased with time. As the treatment time proceeded, the adsorbent sites had the inclination toward saturation. Equilibrium was established at 60 minutes; other factors such as pH, stirring rate, and adsorbent dose were 3, 100 rpm, and 30 g, respectively, with ascorbic acid removal efficiencies of 57%, 61%, 70%, 71%, 72%, and 72%) and lactose removal efficiencies of 41%, 44%, 50%, 51%, 52%, and 53%, as shown in Figure 4. Increased in time leads to increase in the contact between the solution to the larger surface area of adsorbent as there are many adsorption sites.[35–37]

### *Effect of adsorbent dose*

Figure 5 depicts ascorbic acid and lactose removal efficiency as a function of adsorbents dosage. The adsorbent doses were varied between 10 to 50 g/L, other operational factors pH, contact time, initial concentration, and stirring rate were 3, 60 min, 500 mg\L, and 100 rpm, respectively, with ascorbic acid removal efficiencies of 57%, 63%, 70%, 72%, and 76% and lactose removal efficiencies of 34%, 41%, 50%, 56%, and 60%, as shown in Figure 5. As expected, at high adsorbent does the removal increased because of the increased adsorbent surface area and the number of available adsorption sites increased. The optimum adsorbent dose for ascorbic acid and lactose removal was 30 g, as shown in Figures 6 and 7, respectively.[35,37,38–40]

### *Effect of stirring rate*

Figure 8 depicts ascorbic acid and lactose removal efficiency by entrapped activated carbon in alginate as a function of stirring rate. The stirring rate was varied between 100 and 400 rpm, other operational factors such as pH, contact time, initial

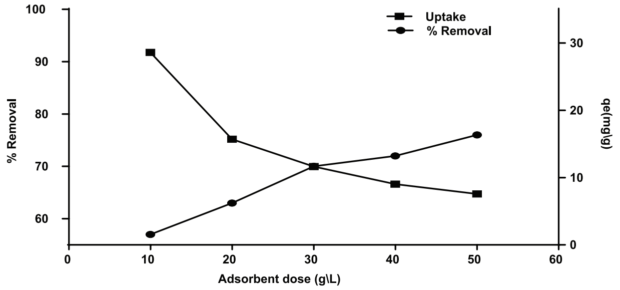


**Figure 6.** Optimum effective dose for ascorbic acid removal.

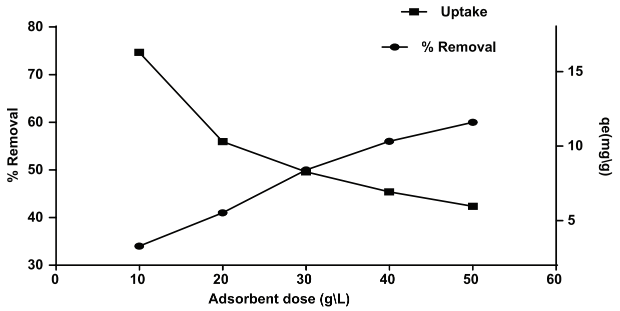


**Figure 7.** Optimum effective dose for lactose removal.

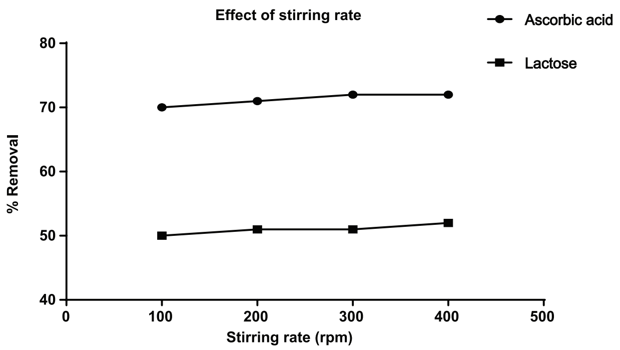


**Figure 8.** Effect of stirring rate on ascorbic acid and lactose removal.

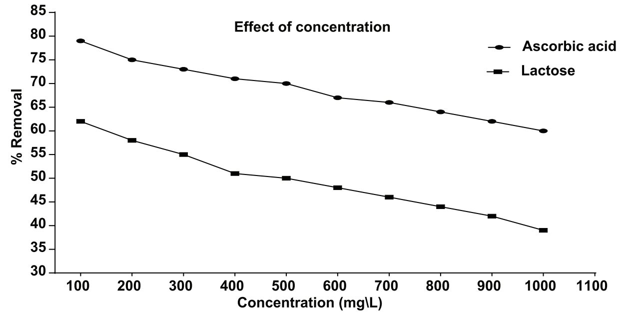


**Figure 9.** Effect of initial concentration on ascorbic acid and lactose removal.

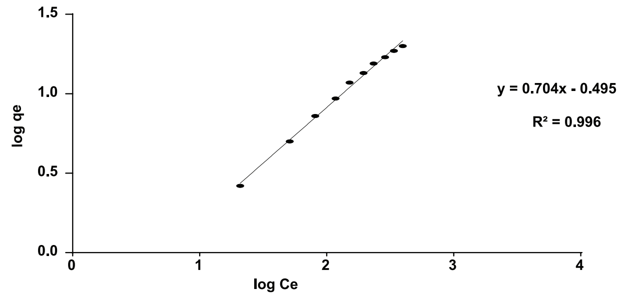


**Figure 10.** Freundlich isotherm of ascorbic acid removal.

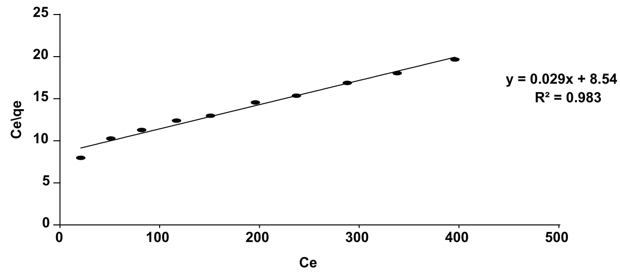


**Figure 11.** Langmuir isotherm of ascorbic acid removal.

concentration, and adsorbents dosage were 3, 60 min, 500 mg\L, and 30 g\L, respectively, with ascorbic acid removal efficiencies of 70%, 71%, 72%, and 72%; and lactose removal efficiencies of 50%, 51%, 51%, and 52%, as shown in Figure 8. The optimum stirring rate for ascorbic acid and lactose removal was 100 rpm. The increase in stirring speed resulting in increase in ascorbic acid and lactose percentage removal was due to the fact that increase in stirring rate enhanced the diffusion of ascorbic acid and lactose contaminants on the surface of the adsorbent.[41]

### *Effect of the initial ascorbic acid and lactose concentration*

The effect of concentration of the aqueous solution on the percent ascorbic acid and lactose reduction by entrapped activated carbon in alginate was studied at various initial concentrations varying from 100 to 1000 mg\L, other operational factors such as pH, contact time, stirring rate, and adsorbents dosage were 3, 60 min, 100 rpm, and 30 g\L, respectively, with ascorbic acid removal efficiencies of 79%, 75%, 73%, 71%, 70%, 67%, 66%, 64%, 62%, and 60%; and lactose removal efficiencies of 62%, 58%, 55%, 51%, 50%, 48%, 46%, 44%, 42%, and 39%, as shown in Figure 9. It can be observed that adsorption was lower at higher concentrations of ascorbic acid and lactose and vice versa.[32,42,43]

*Adsorption isotherm study.* Adsorption capacity of the adsorbent was predicted and evaluated by adsorption isotherm study.[44] The Freundlich and the Langmuir equations are the most common isotherms applications used for water and wastewater treatment.[19,45] The most commonly used model for measuring the sorption of organic compounds from wastewater is the Freundlich isotherm.[19,46] Freundlich and Langmuir isotherm has acquired vogue because of their ability to fit a variety of sorption data.[47] Freundlich isotherm model for ascorbic acid with correlation coefficient ($R^2$) around 0.996, n around 1.421, and $K_f$ around 3.12 showed better fit of adsorption data than Langmuir isotherm model with $R^2$ around 0.983, $q_{max}$ around 34.722, and KL around 0.0037, as shown in Figures 10 and 11, respectively. Freundlich isotherm model for lactose with $R^2$ around 0.992, n around 1.485, and $K_f$ around 5.22 showed better fit of adsorption data than Langmuir isotherm model with $R^2$ around 0.991, $q_{max}$ around 21.692, and KL around 0.0025, as shown in Figures 12 and 13, respectively. Freundlich model indicates that it is more appropriate than the Langmuir isotherm model.

### Statistical analysis

The effect of the variables pH, contact time, dose, concentration, and stirring rate on the removal process of ascorbic acid and lactose has been studied using the entered method, where it was found that $R^2$ = 0.966 and 0.947, and the studied variables profane occupy more than 96% and 94% of the total of the variables affecting the removal process as that the standard error of the estimate are 3.1264 and 4.34099, respectively, which means that the percentage of error in this study is very low.

Analysis of variance was applied and the data were given in Tables 1 and 2. The tables show the sum of squares and the effect of the whole model. It was observed that the *P* value is .000, where the model is considered successful if *P* value is less than .05. From Tables 3 and 4, it can be inferred that all variables had an effect on the removal process except stirring rate, where the *P* value is larger than .05, which means that it can be neglected during the removal process.

*Analysis of statistical and practical results correlation.* The effect of different operating parameters was investigated using Linear regression analysis using SPSS Statistics, where the obtained results support the practical results. The results showed that the effect of pH, adsorbent dose, stirring rate, and concentration has a significant effect on the removal process where the *P* value is less than .05. By applying the B values shown in coefficient tables, the removal equation can be deduced:

$$R\% = Bo + B1X1 + B2X2 + B3X3 + B4X4 + B5X5,$$

where X1 is the effect of pH (2, 4, 6, 8, 10, and 12), X2 is the effect of contact time (15, 30, 60, 120, 180, and 240 minutes), X3 is the effect of adsorbent dose (10, 20, 30, 40, and 50 g/L), X4 is the effect of stirring rate (100, 200, 300, 400, and 500 rpm), and X5 is the effect of concentration (100, 200, 300, 400, and 500 mg/L).

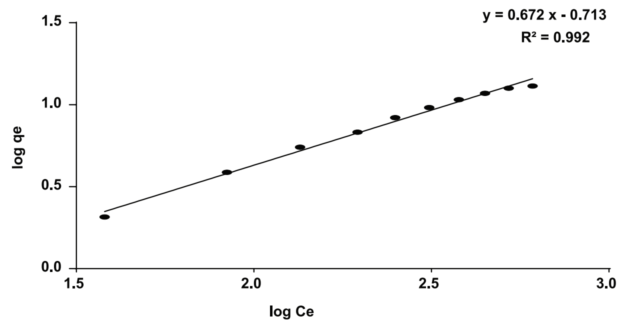


**Figure 12.** Freundlich isotherm of lactose removal.

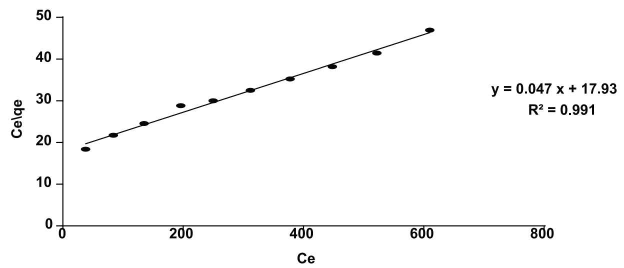


**Figure 13.** Langmuir isotherm of lactose removal.

### Application to a real sample

Sample has been analyzed before and after treatment according to Egyptian law No. 93\1962 and its decree No. 44\2000 concerning discharge final effluent to public sewer system, as shown in Table 5. The sample was collected from Chemical Industries Development Company where pharmaceuticals wastewater is characterized by high values of COD and BOD. The removal efficiency on the sample by using about 30 g\L of entrapped activated carbon in alginate, initial pH 3.4, contact time 60 minutes, and the stirring rate fixed at 100 rpm is shown in Table 6.

## Conclusions

Entrapped activated carbon in alginate polymer was successfully used as adsorbent for ascorbic acid and lactose contaminants removal in aqueous medium. Various operating factors on ascorbic acid and lactose removal efficiency were investigated and optimized. The maximum removal efficiency for ascorbic acid and lactose was at pH 3. The optimal adsorbent dose was about 30 g\L, and adsorption experiments were carried out at room temperature. The entrapped activated carbon in alginate is low–cost adsorbent and can lead to success in

**Table 1.** Ascorbic acid analysis of variance.[a]

| MODEL | SUM OF SQUARES | *DF* | MEAN SQUARE | F | SIGNIFICANCE |
|---|---|---|---|---|---|
| Regression | 7284.229 | 5 | 1456.846 | 47.648 | .000[b] |
| Residual | 1039.546 | 34 | 30.575 | | |
| Total | 8323.775 | 39 | | | |

[a]Dependent Variable (The variable we want to predict): Removal.
[b]Predictors: (Constant), Concentration, Dose, Time, pH, Stirring.

**Table 2.** Lactose analysis of variance.[a]

| MODEL | SUM OF SQUARES | *DF* | MEAN SQUARE | F | SIGNIFICANCE |
|---|---|---|---|---|---|
| Regression | 5550.898 | 5 | 1110.180 | 58.914 | .000[b] |
| Residual | 640.702 | 34 | 18.844 | | |
| Total | 6191.600 | 39 | | | |

[a]Dependent Variable (The variable we want to predict): Removal.
[b]Predictors: (Constant), Concentration, Dose, Time, pH, Stirring.

**Table 3.** Ascorbic acid coefficients.[a]

| MODEL | UNSTANDARDIZED COEFFICIENTS | | STANDARDIZED COEFFICIENTS | *T* | SIGNIFICANCE |
|---|---|---|---|---|---|
| | B | SE | BETA | | |
| (Constant) | 72.787 | 6.914 | | 10.527 | .000 |
| pH | −6.370 | 0.443 | −0.876 | −14.376 | .000 |
| Time | 0.057 | 0.021 | 0.167 | 2.743 | .010 |
| Dose | 0.470 | 0.175 | 0.163 | 2.688 | .011 |
| Stirring | 0.028 | 0.015 | 0.110 | 1.796 | .081 |
| Concentration | −0.017 | 0.006 | −0.173 | −2.848 | .007 |

[a]Dependent variable: removal.

**Table 4.** Lactose coefficients.[a]

| MODEL | UNSTANDARDIZED COEFFICIENTS | | STANDARDIZED COEFFICIENTS | *T* | SIGNIFICANCE |
|---|---|---|---|---|---|
| | B | SE | BETA | | |
| (Constant) | 49.017 | 5.428 | | 9.030 | .000 |
| pH | −5.346 | 0.348 | −0.853 | −15.369 | .000 |
| Time | 0.043 | 0.016 | 0.147 | 2.660 | .012 |
| Dose | 0.670 | 0.137 | 0.269 | 4.881 | .000 |
| Stirring | 0.020 | 0.012 | 0.090 | 1.627 | .113 |
| Concentration | −0.021 | 0.005 | −0.251 | −4.539 | .000 |

[a]Dependent variable: removal.

**Table 5.** Maximum effluent concentrations set by law 93/1962 and decree 9/1989.

| PARAMETER | REFERENCE METHODS | MAXIMUM CONCENTRATION |
|---|---|---|
| BOD | SM5210 | 400 mg\L |
| COD | SM5220B | 700 mg\L |

Abbreviations: BOD, biological oxygen demand; COD, chemical oxygen demand.

**Table 6.** Analysis of the sample.

| PARAMETER | BEFORE | AFTER | REMOVAL |
|---|---|---|---|
| | (MG\L) | (MG\L) | % |
| BOD | 870 | 302 | 63 |
| COD | 1650 | 660 | 60 |

Abbreviations: BOD, biological oxygen demand; COD, chemical oxygen demand.

treatment of wastewater and produce a good treated effluent. Adsorption isotherm studies indicate that Freundlich isotherm model is more appropriate than the Langmuir isotherm model.

## Author Contributions

Conception and design: HMAA.
Acquisition of data: SAAG, HMAA.
Analysis and interpretation of data: HMAA.
Drafting the article: SAAG, HMAA.
Critically revising the article for important intellectual content: HMAA.
Final approval of the version to be published: SAAG, HMAA.

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