

Effect of nickel on the adsorption of silver ions over activated carbon

Hossam Altaher

Sustainable Solution Group, Kuwait City, Kuwait

Yehia H. Magdy

El-Minia University, El-Minia, Egypt, and

Anwar F. Al Yaqout

Kuwait University, Kuwait City, Kuwait

Abstract

Purpose – The disposal of wastewater containing silver is an environmental concern. Due to the toxicity of silver, treatment of such wastewater is necessary. Real wastewater contains a complex matrix of pollutants. The purpose of this paper is to study the adsorption behavior of silver in single and binary systems (with nickel) onto granular activated carbon.

Design/methodology/approach – The effect of silver ions concentration and the mass of adsorbent on the adsorption behavior were analyzed. Five two-parameter isotherms (Langmuir, Elovich, Freundlich, Dubinin–Radushkevich and Temkin) were applied to investigate the adsorption mechanism. Both linear and nonlinear regressions were tested for the first three isotherms. The experimental data were also fitted to Redlich–Petersons, Sips and Toth models.

Findings – A direct relationship between the initial silver ion concentration and its adsorption capacity was observed, whereas an inverse relationship between the adsorbent mass and the adsorption capacity was documented. The Langmuir model was found to best-fit the data indicating monolayer adsorption behavior. The maximum uptake was 2,500 mg/g in the single adsorption system. This value decreased to 909 mg/g in the binary system. The adsorption was found to have an exothermic chemical nature.

Originality/value – The study of the silver adsorption in a single system is inaccurate. Real wastewater contains a complex matrix of pollutants. This research gives a clear insight into the adsorption behavior in binary systems.

Keywords Adsorption, Equilibrium, Isotherms, Wastewater, Silver, Binary system

Paper type Research paper

1. Introduction

The toxicity of silver due to wastewater disposal is an environmental concern. Silver can deposit in human bodies causing argyria. Silver may also cause skin and eye irritation, dermatitis, genotoxicity, neurotoxicity and possible carcinogenicity (Hadrup *et al.*, 2018; Jeon, 2015).

Many industries produce wastewater containing silver such as plating industry, photographic manufacturing, medicine industry, consumer products industry, electronics industries, battery industry, clothing and metal alloy industries (Ho *et al.*, 2018; Jeon, 2015). The concentration of silver in wastewater can reach a value as high as 5,000–10,000 mg/L (Ho *et al.*, 2018). Silver is included in many products including amalgam fillings, coins, jewelry, deodorants, tableware, coatings and medical devices (Hadrup *et al.*, 2018). All these are possible sources of polluting water with silver.

Many techniques including membrane separation, coagulation/flocculation, chemical precipitation, ion exchange, may be used to remove silver ions from wastewater (Freitas *et al.*, 2017). However, adsorption is the preferred method due to its various advantages over other techniques; e.g. modest operation, high efficiency, availability of adsorbents, easy recovery of adsorbent and generation of a relatively small amount of sludge (Jeon, 2015, 2017).

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The results in different literature about the adsorption of silver are inaccurate. Most of the researchers investigate the removal of silver as a single contaminant in wastewater. Real wastewater contains a composite mixture of different pollutants (Flores López *et al.*, 2018). Silver, for instance, may be found associated with nickel in some wastewater (Li, Wang, Cui, Lou, Shan, Xiong and Fan, 2018). It is, therefore, desirable and necessary to study the adsorption of silver from binary and even multi-component system (Fan *et al.*, 2008).

The aim of this current research is to analyze the adsorption behavior of silver ions in single and binary systems and to study the effect of the completion between pollutants on adsorption parameters.

2. Methods and procedure

2.1 Adsorbent

The adsorbent was commercial granular activated carbon from El-Gumhuria Company (Cairo, Egypt). The activated carbon was dried when received at 120°C and kept in sealed bottles until used.

2.2 Wastewater

All chemicals were of analytical grade. An accurately weighed amount of silver nitrate and nickel chloride were individually dissolved in bi-distilled water to prepare the working solutions. The solutions were used without any pH adjustment. Silver and nickel ions concentrations were determined calorimetrically using Scan AA4 instrument (Thermo Jarrell Ash, Franklin, MA, USA) (APHA, Greenberg, 1999).

2.3 Batch study

To determine the effect of adsorbent mass, four 125 mL Erlenmeyer flasks were washed and oven-dried at 120°C. A given mass of activated carbon (0.05, 0.1, 0.5 and 1 g) was placed in each flask. Then, 25 mL of the silver solution of predetermined concentration was poured into the flasks. The flasks were shaken at a rate of 400 rpm for 6 h. After equilibrium is attained the adsorbent was separated by filtration to determine the final adsorbate concentration. The same procedure was repeated using a different initial concentration of silver (0–500 mg/L). The experiments were repeated twice for better accuracy. The shaking procedure was operated at room temperature of 25±2°C.

For binary adsorption, mixtures of (0–500 mg/L) of silver and (0–500 mg/L) nickel were prepared. The concentration of silver and nickel was kept equal in all mixtures.

2.4 Mathematical models

The experimental data were analyzed using five 2-parameters models. These models are Langmuir, Freundlich, Temkin, Elovich and Dubinin–Radushkevich. The first three models were analyzed in both linear and nonlinear forms. In addition, the data were fitted to Redlich–Peterson, Toth and Sips as examples for three-parameters models. The equations and parameters of these models are presented in Table I. The fit of each linear isotherm model was assessed using the coefficient of determination (R^2), while the fitting to nonlinear forms was assessed using the sum of squared deviation (SSD).

3. Results and discussion

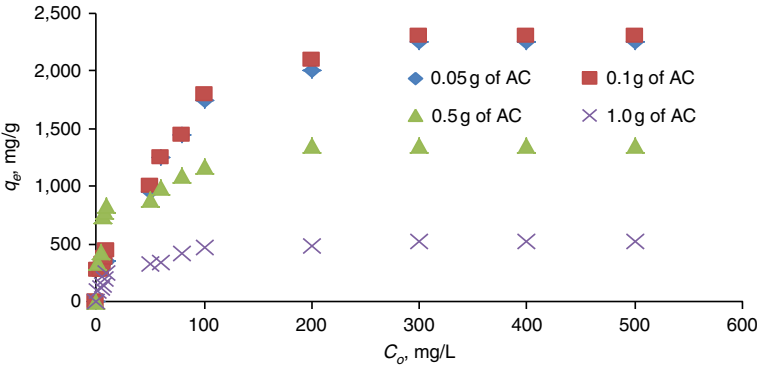
3.1 Initial concentration

Figure 1 depicts the effect of initial adsorbate concentration on the uptake of adsorbate ions in a single system. It is clear from the four plots in the figure (representing different adsorbent dosages) that the initial concentration has a significant effect on the uptake of silver ions.

Table I.
Equations and
parameter for
isotherm models

Isotherm model	Parameters	Reference
Langmuir model Nonlinear: $q_e = q_m C_e / (K + C_e)$ Linear: $1/q_e = 1/(q_m) + C_e(K_L/qm)$ $R_L = 1/(1 + K_L C_o)$	q_e and q_m (mg/g): adsorption capacity at equilibrium and maximum adsorption capacity, C_o and C_e (mg/L): initial and equilibrium concentration of the adsorbate, K_L (L/mg): constant related to adsorption rate, R_L : separation factor	Abubeah <i>et al.</i> (2018)
Freundlich model Nonlinear: $q_e = k_f C_e^{1/n}$ Linear: $\ln q_e = \ln k_f + 1/n (\ln C_e)$	q_e and C_e : same designation as Langmuir model, n : constant that identifies the adsorption intensity of adsorbate, k_f : constant related to the bonding energy	Khalil <i>et al.</i> (2016)
Temkin model Nonlinear: $q_e = B_T \ln(K_T C_e)$ Linear: $q_e = B_T (\ln K_T) + B_T \ln(C_e)$ $B_T = (R_T)/b$	q_e and C_e : same as in Langmuir model, K_T (L/mg): equilibrium binding constant, b (J/mol): Temkin isotherm constant related to the heat of adsorption, T : absolute temperature (K), R : universal gas constant	Ashour <i>et al.</i> (2015)
Dubinin–Radushkevich (D-R) Nonlinear: $q_e = q_m \exp(-Be^2)$ Linear: $\ln q_e = \ln q_m - Be^2$ $\epsilon = RT \ln(1 + 1/C_e)$ $E = 1/(2B)^{0.5}$	q_e , q_m , R , T and C_e : same as in Langmuir model, B : constant related to the mean free energy of adsorption per mole of adsorbate	Altaher <i>et al.</i> (2014), Magdy <i>et al.</i> (2018)
Elovich model Nonlinear: $q_e = q_m K_e C_e \exp(q_e/q_m)$ Linear: $\ln q_e/C_e = \ln K_e q_m - q_e/q_m$	q_e , q_m , and C_e : same as in Langmuir equation, K_e (L/mg): Elovich equilibrium constant	Farouq and Yousef (2015)
Redlich–Peterson model $q_e = ((k_R C_e)/(1 + a_R C_e^{b_R}))$	q_e and C_e : same as in Langmuir equation, K_R (L/g), a_R (L/mg) ^{b_R} and b_R are the isotherm constants	Magdy <i>et al.</i> (2018)
Toth model $q_e = ((k_t C_e)/(a_t + C_e)^{1/t})$	q_e and C_e : same as in Langmuir model, K_t and t are related to adsorbate-adsorbent pairs	Magdy <i>et al.</i> (2018)
Sips model $q_e = ((q_m(a_s C_e)^{n_s})/(1 + (a_s C_e)^{n_s}))$	q_e , q_m and C_e : same as in Langmuir equation, a_s : constant related to the adsorption equilibrium, n_s : constant that is specific to the adsorption system	Magdy <i>et al.</i> (2018)

Figure 1.
Effect of initial ion
concentration on
the adsorption
capacity of AC



It is clear from the figure that there is a direct relationship between the initial concentration and adsorption uptake. Increasing the concentration of the silver ions from 2 mg/L to 500 mg/L resulted in an equivalent increase of the uptake from 288 mg/g to 2,250 mg/g (for adsorbent mass 0.05 g). Similar behavior can be noticed for other adsorbent masses. The strong correlation between initial concentration and silver uptake may be attributed

to the strong driving force exerted by the high adsorbate concentration at the initial stages of the removal process (Qiu *et al.*, 2014).

Moreover, at low concentrations, the number of adsorbate species is low while there are many vacant adsorption sites. Accordingly, no competition occurs between adsorbent ions (Magdy and Altaher, 2018).

3.2 Adsorbent mass

The relationship between the adsorbent dose and the adsorption uptake in a single system is shown in Figure 1. The influence of adsorbent dose is obvious for the adsorption of silver. At initial adsorbate concentration of 500 mg/L, increasing the dose from 0.05 to 1.0 g resulted in a decrease in the uptake from 2250 mg/g to 525 mg/g. The same trend can be observed for other initial concentrations. This behavior may be attributed to the number of active sites as well as the surface area of the adsorbent. These two properties of adsorbents increase with increasing the adsorbent mass (Ahmadpour *et al.*, 2010). Accordingly, at certain adsorbate concentration and for a high mass of adsorbent, a very large number of active sites will be available. Accordingly, no competition between adsorbate ions will take place and consequently, the low value of adsorbate ions per unit mass of adsorbent and unsaturation of adsorption sites will be attained (Baseri and Tizro, 2017). The particle interactions (aggregation) at high doses of adsorbent may also result in a reduction in the adsorbent surface area (Subramani and Thinakaran, 2017). It is worth noting that at adsorbent masses of 0.05 and 0.1 g, the incremental adsorption capacity is very low. The reason may be an equilibrium of the adsorbate concentration on both the surface of the adsorbent and the bulk solution at these low dosages of adsorbent (Wang *et al.*, 2010).

3.3 Adsorption isotherm

Figure 2 illustrates the relationship between the concentration of adsorbate and its adsorption uptake. The shape of the curve characterized by a rapid rise in the initial adsorption stage with increasing adsorbate concentration and attaining complete monolayer coverage indicates that the adsorption belongs to Type I isotherms. The first linear region may be attributed to prompt uptake of most available active sites of adsorbent while the following region is due to the slow diffusion of the silver ions through the adsorbent pores (Quintelas *et al.*, 2008). This isotherm occurs in adsorption systems with strong attractive interaction between adsorbate and adsorbent (usually in chemisorption).

3.3.1 The Langmuir model. The Langmuir model adopts a concept of adsorbates form a monolayer on a specific energetically homogeneous surface (Abubeah *et al.*, 2018). The bonding to the sorption sites is mostly chemical in nature (Ghosal and Gupta, 2017).

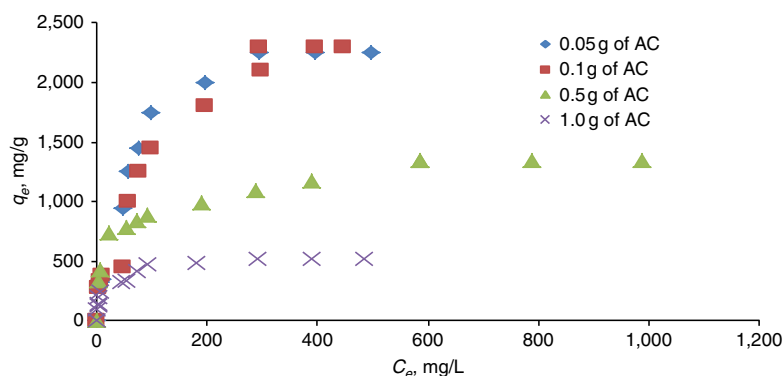


Figure 2.
Adsorption isotherm
plot for silver ions
on AC

The separation factor (R_L) is an important parameter in the Langmuir model. The value of this parameter describes the favorability of the adsorption. The adsorption is unfavorable if R_L is greater than 1. However, the adsorption is favorable for ($0 < R_L < 1$). $R_L = 1$ indicates a linear adsorption while $R_L = 0$ indicates irreversible adsorption (Rahmani *et al.*, 2018).

The parameters calculated from linear form-fitting are depicted in Table II. It is clear that the R^2 values for all tested adsorbent doses are significantly greater than that of other adsorption isotherms, which confirms the applicability of this model to describe the adsorption of silver ions on AC. The values of R_L being much less than 1 indicate that the adsorption is highly favorable. The maximum adsorption capacity as calculated from the fitting to the linear model was as high as 2,500 mg/g for AC mass of 0.05 g and as low as 526 mg/g for AC mass of 1.0 g.

The parameters obtained from both the linear and nonlinear equation for the four adsorption systems (Tables II and III) are slightly different. For instance, the maximum adsorption capacities for adsorption mass of 0.05 g of AC were 2,500 mg/g and 2,595 mg/g as obtained from linear and nonlinear forms, respectively (3.8 percent difference).

3.3.2 The Freundlich model. This model can be applied to explain adsorption in heterogeneous systems (Peruchi *et al.*, 2015). It assumes that the adsorbent surface contains various sorts of sites that act at the same time during the adsorption process. Each site has a certain energy of adsorption. The value of the exponent n is related to the favorability of adsorption. If this value is greater than 1, the adsorption is favorable (Altaher *et al.*, 2014). This isotherm is applicable in the low to moderate adsorbate concentrations (Stromer *et al.*, 2018). The Freundlich parameters are listed in Tables II and III.

The values of R^2 for the four adsorption systems are in the range 0.921–0.980. These numbers are reasonably high; however, comparing these numbers with that obtained from the Langmuir model recommends the latter to be more appropriate to fit the data. On the

Table II.
Linear adsorption
constants for
the applied
two-parameters
adsorption models

Isotherm	0.05 g of AC	0.1 g of AC	0.5 g of AC	1.0 g of AC
<i>Langmuir</i>				
R^2	0.989	0.979	0.994	0.997
q_m , mg/g	2,500	2,500	1,429	526
b	0.023	0.019	0.024	0.081
R_L	0.080	0.095	0.077	0.024
<i>Freundlich</i>				
R^2	0.950	0.921	0.980	0.936
n	2.29	2.28	4.54	4.12
K_f	183	164.8	320	134
<i>Temkin</i>				
R^2	0.934	0.859	0.968	0.907
B_T	409	408	172	59.2
K_T	0.51	0.41	0.97	2.592
b	4,857	6,043	2,554	956
<i>Elovich</i>				
R^2	0.852	0.699	0.964	0.874
q_m	833	909	227	84.7
K_e	0.13	0.08	2.59	8.07
<i>D-R</i>				
R^2	0.940	0.888	0.984	0.954
q_m	5,201	4,650	1,869	871
E	11.8	11.8	15.8	15.8

Table III.
Nonlinear adsorption
constants for the
applied 2-parameters
adsorption models

Isotherm	0.05 g of AC	0.1 g of AC	0.5 g of AC	1.0 g of AC
<i>Langmuir</i>				
SSD	1.8E5	3.7E5	2.3E5	3.8E4
q_m	2,595	3,012	1,291	489
K_L	0.017	0.008	0.041	0.150
R_L	0.105	0.2	0.047	0.013
<i>Freundlich</i>				
SSD	4.6E5	3.8E5	2.9E4	1.6E4
n	2.722	2.201	4.855	4.740
K_f	260	156	345	155
<i>Temkin</i>				
SSD	2.8E5	7.3E5	4.6E4	1.0E4
B_T	453	454	172	73
K_T	0.340	0.250	2.649	3.677
B	7,287	9,911	935	673

other hand, the values of n are in the range 2.29–4.54 and 2.2–4.74 for linear and nonlinear fitting, respectively. These values (being greater than 1) show the favorability of the adsorption; a conclusion obtained from Langmuir models.

3.3.3 The Temkin model. This model assumes that adsorption on the surface of adsorbent which has a uniform distribution of active sites having similar energies. In addition, it postulates the heat of adsorption to decrease linearly instead of logarithmically (Ashour *et al.*, 2015; Kaveeshwar *et al.*, 2018).

The parameters of the model are listed in Tables II and III. The values of R^2 are low compared with that of Langmuir and Freundlich models. However, the fit is still considerable especially in the case of the nonlinear fitting. The values of the parameter b were found to be in the range of 0.96–6.04 KJ/mol and 0.76–9.9 KJ/mol for linear and nonlinear fitting. The values of this parameter suggest a chemical nature of the process. On the other hand, the positive values suggest exothermic adsorption (Hamdaoui, 2006).

3.3.4 The Elovich model. This isotherm assumes exponential escalation of the adsorption sites with adsorption. Accordingly, the adsorption takes place forming a multilayer of adsorbates on the surface of adsorbents (Kaveeshwar *et al.*, 2018). The fitting of experimental data to the Elovich model gave a poor fit as illustrated by Table II, thus, it is clearly not appropriate for predicting the adsorption of silver on AC. This agrees with the results of the Langmuir isotherm. It also affirms the monolayer nature of adsorption.

3.3.5 Dubinin–Radushkevich isotherm. This model explains the mechanism of the adsorption process using the potential theory (Altaher *et al.*, 2014). The value of the parameter E determines the type of the adsorption, physical or chemical. If $E > 16$ KJ/mol the adsorption is chemical. If $E < 8$ KJ/mol the adsorption has a physical nature, while $8 < E < 16$ is for ion exchange (Khalil *et al.*, 2016). The values of E presented in Table II are high and indicate ion-exchange mechanism which agrees with the results of both Langmuir and Temkin isotherms.

3.3.6 Three-parameters isotherms. Redlich–Peterson model combines the properties of both the Freundlich and Langmuir models. At low concentration, the model is similar to Henry’s law while at high concentrations it approximates the Freundlich model. Toth model is usually applied in heterogeneous systems. Accordingly, most adsorption sites have adsorption energy lower than the maximum energy of adsorption. In this model, the constant (t) describes the system heterogeneity. This constant is usually less than 1 (Quintelas *et al.*, 2008). This model is usually used to evaluate the maximum adsorption

uptake in monolayer adsorption systems (Whittaker *et al.*, 2013). Sips model is suitable for describing adsorption in heterogeneous systems. It has the features of Langmuir and Freundlich isotherms, so it is also called Langmuir–Freundlich isotherm. At high adsorbate concentration, the model can predict the monolayer adsorption specific for the Langmuir isotherm while at low adsorbate concentration it is similar to the Freundlich model (Quintelas *et al.*, 2008). The parameters obtained from fitting of data to the three-parameter isotherms are presented in Table IV. The high SSD values indicate weak fitting of the data to the three models. The weak fitting may be attributed to the high concentrations of the adsorbate.

The ratio Kr/Ar obtained from Redlich–Peterson model (1,190, 371, 386 and 208 for the four adsorption systems) are not similar to the values of K_f obtained from both the linear and nonlinear fitting to the Freundlich isotherm. On the other hand, the values of b_R are in the range 0–1 which shows favorable adsorption (Hamdaoui and Naffrechoux, 2007).

The values of the maximum adsorption uptake obtained from the Sips model are greater than those obtained from fitting the experimental data to linear and nonlinear forms of the Langmuir model. These values are also much greater than that illustrated by the experimental data. The reason may be the values of the constants n_s deviated from unity (Kumara *et al.*, 2014).

The value of the constant (t) obtained from fitting the data to Toth isotherm deviates from unity (as depicted in Table IV). That means the adsorbent surface is homogeneous (Wu *et al.*, 2013); a result confirmed by the good fitting of the data to the Langmuir model. Based on the previous discussion, it can be concluded that the two-parameter isotherms better describes the adsorption data than the three-parameter models.

3.4 Effect of nickel presence on the uptake of silver

Silver ions are usually present in industrial wastewater mixed with other metal ions. The ions compete for active adsorption sites, thus decreasing the adsorbent’s removal efficiency. Therefore, the removal of silver ions in the presence of nickel ions was investigated.

In multi-adsorbate systems, many factors may affect the adsorption process. These factors include surface-charge neutralization, electrical double layer compression, cation bridging effect, competitive adsorption and salting-out (Du *et al.*, 2014). The presence of

Table IV.
Nonlinear adsorption
parameters for
the applied
three-parameter
adsorption models

Isotherm	0.05 g of AC	0.1 g of AC	0.5 g of AC	1.0 g of AC
<i>Redlich–Peterson</i>				
SSD	1.5E5	5.8E4	2.4E5	1.3E4
K_R	59.5	119	852	213
a_R	0.050	0.321	2.209	1.024
b_R	0.874	0.688	0.811	0.843
<i>Toth</i>				
SSD	1.8E5	6.6E4	2.5E4	1.1E4
K_T	4,791	296	377	150
a_T	79.6	6.53	0.586	4.5E-7
t	0.915	1.52	1.238	1.27
<i>Sips</i>				
SSD	1.2E5	3.9E4	2.45E4	8,983
q_m	3,018	4,455	3,120	1,063
a_s	0.011	0.003	0.0005	0.003
n_s	0.770	0.606	0.296	0.322

inorganic ions in the adsorption can compress the electrical double layer of adsorbents consequently affecting the adsorption capacity (Gao and Chorover, 2012).

Binary adsorption experiments were performed in this current research for the sake of comparison. As can be observed from Figure 3, no change in the shape of the adsorption curve has occurred. The adsorption of silver in the binary system is through monolayer as may be suggested by the *L*-shaped plot in the figure. The parameters obtained from fitting the data to the Langmuir and Freundlich isotherms are shown in Table V. It is clear from the values of R^2 , that Langmuir model best fits the data which means according to the postulates of Langmuir model that the adsorption of silver ions in the binary system takes place on the active sites of AC forming a monolayer. Comparing the values of R_L and n indicates that the adsorption of silver in the binary system is still favorable. The proportion of maximum adsorption uptake of silver in the binary system to adsorption uptake in the single system is 0.36. This small value indicates that the simultaneous presence of silver and nickel greatly reduced the adsorption of silver through competition for adsorption sites on AC and it explains the affinity of the individual ions for the adsorbent (Li, Liu, Liu, Zeng, Hu, Tan, Jiang, *et al.*, 2018).

4. Conclusions

The adsorption performance of silver ions on AC in both single system and the binary system was studied. The adsorption process is best defined by the Langmuir model in both single and binary systems demonstrating monolayer uptake of such ions. The adsorption

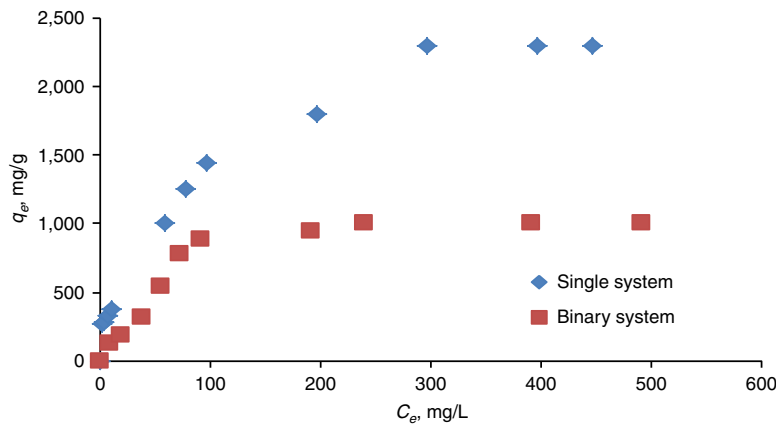


Figure 3.
Adsorption isotherms
for silver adsorption
in a binary system
with nickel

Isotherm	Single system	Binary system
<i>Langmuir</i>		
q_m , mg/g	2,500	909
K_L	0.0189	0.0166
R^2	0.978	0.963
R_L	0.021	0.062
<i>Freundlich</i>		
n	2.3	1.9
K_f	175	52
R^2	0.974	0.850

Table V.
Adsorption constants
for the adsorption of
silver in single and
binary systems

has an exothermic chemical nature. The maximum uptakes of silver, as calculated from the Langmuir model were 2,500 and 909 mg/g, respectively. A competition between silver and nickel ions is clear. However, the adsorption of silver ions in both systems is favorable.

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Corresponding author

Hossam Altaher can be contacted at: haltaher@hotmail.com