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Comparative Study of Saad Isotherm Model with Conventional Isotherm Models for Cobalt Ion Adsorption from Wastewater on Activated Saudi Clays

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ABSTRACT

The main objective of this study was to compare a newly developed isotherm model (Saad isotherm model, SIM) with five conventional isotherm models (Langmuir, Freundlich, Langmuir-Freundlich, BET and Toth) for adsorption of cobalt (Co) ion on a specified adsorbent. The Saad isotherm model fitted well the experimental data from three types of activated and non-activated Saudi clays. The Langmuir isotherm model agreed well with the experimental data of Khiber clay and Bahhah clay activated by H_2O_2, NaCl and H_2SO_4. While the Frendlich model and Langmuir-Frendlich model fitted well the experimental data of natural Tabbuk clay, activated Tabbuk clay by NaCl and the natural Bahhah clay. In conclusion, the Frendlich model fitted well the experimental data of Tabbuk clay activated by H_2O_2 and H_2SO_4. Besides, the BET model did not describe the experimental data well of the three types of activated clays.

Key words: Clay types, Tabbuk, Khiber, Bahhah, isotherm model, adsorption capacity activators

INTRODUCTION

Production of wastewater from different sources in Saudi Arabia is rising and contains a number of pollutants including some toxic heavy metals. This scenario has created interest among scientists for wastewater treatment to remove and minimize the level of toxic heavy metal ions for its reuse in industrial and agriculture sectors in order to save the precious limited groundwater sources. Among the different heavy toxic metals, Co is considered quite harmful when present above the permissible limits. The main sources of cobalt (Co) ion in wastewater are electroplating, catalytic processes, ceramic and alloys industries (Brezonik, 1974; Patterson and Passino, 1987; Emsley, 1992).

Different traditional processes such as membrane process, chemical precipitation and ion exchange process are employed for the removal of Co from wastewater. Additionally, separation of metal ion by adsorption process is another sound approach for wastewater treatment for the removal of toxic heavy metals. Presently, adsorption technology is gaining momentum as an important process in wastewater treatment. Because, it offers an attractive and cost effective solution for the separation of pollutants depending on types of adsorbents without requiring phase changes such as in the distillation process. Above all, the main advantage of adsorption process is the use of clay, a natural adsorbent which is cheap and easily available locally.

Previously different types of activators and activation techniques were examined for increasing the adsorption capacities of several types of clays (Eren, 2008; Dogan *et al*., 2008; Stathi *et al*., 2007; Unuabonah *et al*., 2007; Diaz *et al*., 2007; Adebowale *et al*., 2005; Al-Asheh *et al*., 2003; Singh *et al*., 2001; Suraj *et al*., 1998). A literature review did not reveal any scientific investigation on the activated Saudi clays for the adsorption of Co from wastewater.

Eren (2008) studied the adsorption of Cu ion by manganese oxide modified bentonite clay using a batch adsorber and found an increase in the adsorption capacity of clay for Co ion. Many investigators studied the adsorption of heavy metals on different types of clays activated by various activators such as Co and Cu ions on the modified surface of sepiolite clay by trimethoxysilane and [3-(2-aminoethylamino) propyl] (Dogan *et al*., 2008), Pb, Cd and Zn on four organic-modified montmorillonite clays (Stathi *et al*., 2007) and Pb ions on tripolyphosphate-impregnated kaolinite clay (Nigeria) by changing pH of the solution (Unuabonah *et al*., 2007). Hectorite clay from USA and vermiculite clay from Spain were converted to functionalized hectorite and functionalized vermiculite using thiol groups and used to adsorb lead ions from aqueous solution. They found that the adsorption of Pb ion by the functionalized vermiculite clay was higher than the functionalized hectorite. The adsorption capacity of clays was 33 mg g^{-1} for functionalized vermiculite and 10 mg g^{-1} for functionalized hectorite (Diaz *et al*., 2007).

Many studied investigated the adsorption of Pb, Cd, Zn and Cu ions by phosphate and sulphate modified kaolin clay (Nigeria) (Adebowale *et al*., 2005) and the methylene blue adsorption on activated bentonite clay (Al-Asheh *et al*., 2003). Furthermore, the activation of bentonite clay by sodium dodecyl sulphate enhanced the adsorption capacity better than the natural bentonite. In another study, a batch equilibrium technique was used for the adsorption of Pb on phosphatic clay from USA and the adsorption capacity of this ion was 32 mg g^{-1} (Singh *et al*., 2001). Also, the adsorption of Cu and Cd ions was carried on two kaolinites (Kerala) modified by calcination and acid. They stated that the adsorption capacity decreased by increasing calcination temperature.

This study aims to compare the relative performance of Saad Isotherm Model (a new invention) with other conventional isotherm models for the adsorption of Co ions on different types of clays.

MATERIALS AND METHODS

Materials: The adsorbents used in this research were three types of Saudi natural clays collected from different locations in Saudi Arabia.

The Co ions was used an adsorbate. The Co ion solution was prepared using cobalt (II) nitrate purified LR [$Co(NO_3)_2$] and supplied by VWR International SAS 201, Rue Camot-F- 94126, Sous bois.

Activation of experimental clays: Three types of Saudi clays were activated by sulphuric acid (H_2SO_4), hydrogen peroxide (H_2O_2) and sodium chloride (NaCl) under specific activation conditions (1 M, 1 h, 25°C).

Equilibrium experiments: Equilibrium isotherms were obtained for the natural and activated clays (Tabbuk, Bahhah and Khiber) using a constant mass of clay (1 g) in 50 mL cobalt ions solution in glass bottles placed on a constant agitation shaker. In all the isotherm runs, the cobalt ion solution concentrations ranged from 50-2000 mg L^{-1}. The particle size of clay was 0.25 mm and the temperature was 25°C to determine the isotherms.

The adsorption of cobalt ions and the clay system reached to steady of state equilibrium after 40 min as shown in Fig. 1. Therefore, the equilibrium process was run for a total period of 3 h. Then, the samples were filtered followed by dilution and measurement of absorbance by atomic absorption spectroscopy. Lastly, the absorbance of samples was converted to concentrations with the help of calibration curve for cobalt ions. The cobalt ions adsorption on the activated clay was obtained from the mass balance equation on the batch adsorber as follows:

$$q_e = \frac{V(C_0 - C_e)}{M} \quad (1)$$

where, M is the mass of clay (g), V is the volume of cobalt ion solution (L), q_e is the cobalt ion adsorption on the clay surface (mg g^{-1}), C_o is the initial concentration of cobalt ions (mg L^{-1}) and C_e is the equilibrium concentration of the cobalt ions (mg L^{-1}). The cobalt ions adsorption on natural and

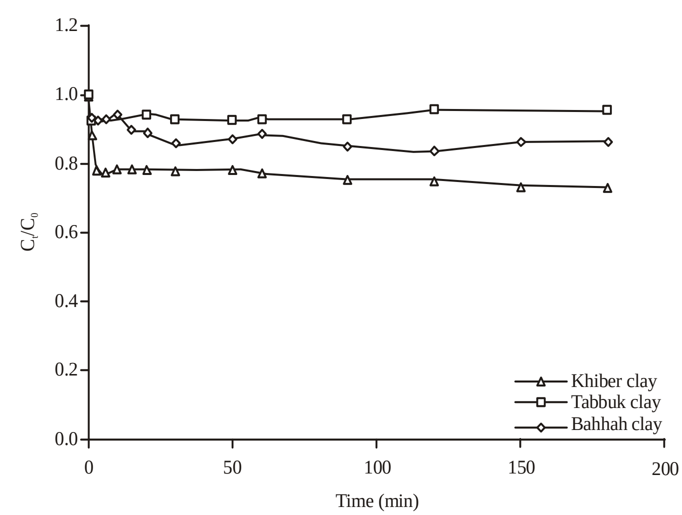

Fig. 1: Time equilibrium for adsorption of cobalt ions on different types of natural clay

activated clays versus the equilibrium concentration of the cobalt ions was plotted to obtain the equilibrium adsorption isotherm curves.

RESULTS AND DISCUSSION

Analysis of the equilibrium isothermal models: Five equilibrium isotherm models namely, Langmuir, Freundlich, Langmuir-Freundlich, BET, Toth isotherm and a new isotherm model (called as Saad isotherm model, SIM) were employed in the study. The equilibrium parameters were estimated by correlating the equilibrium equations with data from equilibrium experiments using a nonlinear regression technique (i.e., fminsearch function from MATLAB). Therefore, the equilibrium parameters were used to describe the removal of cobalt (Co) ion by three types of activated clay as adsorbents in a batch and fixed bed columns. Langmuir isotherm model.

The Langmuir equation assumed that the adsorption of Co ion on activated clay is a monolayer phenomenon and is applied to evaluate the maximum capacity of the activated clay (Mckay, 1996). The Langmuir isotherm equation is written as follows:

$$q_e = \frac{KC_e}{1+bC_e} \quad (2)$$

The Langmuir parameters K and b were obtained by using the non-linear regression technique with Eq. 2.

The equilibrium parameters K and b were calculated by non-linear regression technique as given in Table 1. The data in Table 1 illustrate that Langmuir model described the experimental data well. The dimensionless equilibrium parameter ($\acute{R}$) was obtained to decide whether the cobalt ions adsorption on three types of activated clay is favorable or unfavorable by using the subsequent form El-Geundi *et al.* (2005):

$$\acute{R} = \frac{1}{1+bC_0} \quad (3)$$

where, b is the Langmuir parameter and C_o is the initial concentration. The values of dimensionless parameter for the adsorption of Co ions on three types of activated clay were obtained as shown in Table 1. The values of the dimensionless parameter were between $0<\acute{R}<1$. This range showed that the adsorption process is favorable (El-Geundi *et al.*, 2005), thus indicating that the adsorption of Co ions on three types of activated clay is favorable.

Freundlich Isotherm Model (FIM): The Freundlich isotherm model described the experimental data for heterogeneous surface. The Freundlich form is written as follows:

$$q_e = K_F\, C_e^{1/n} \quad (4)$$

The equilibrium constants K_F and n are calculated using non-linear regression technique with Eq. 4. The n values are greater than one, as a result, cobalt ions adsorbed on the three types of activated clay is favorable (El-Geundi, 1990).

The equilibrium constants were calculated by non-linear regression technique and presented in Table 2. It is evident from the results in Table 2 that the Freundlich model is well fit with experimental data.

Langmuir-Freundlich Isotherm Model (L-FIM): The merging between Langmuir and Freundlich isotherm models created a new model named as the Langmuir-Freundlich isotherm model. The new model has three parameters K_c, b_c and m and is more suitable for heterogeneous surface (Unuabonah *et al.*, 2007). This form can be written as follows:

$$q_e = \frac{K_c\, C_e^{\frac{1}{m}}}{1+b_c C_e^{\frac{1}{m}}} \quad (5)$$

At m = 1, the model converts to Langmuir. The Langmuir-Freundlich parameters K_c, b_c and m were obtained by the non-linear regression technique with Eq. 5.

Table 1: Langmuir equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K (L g^{-1})	b (L mg^{-1})	$\bar{R}$
Khiber clay			
Natural	0.0710	0.0140	0.129
H_2O_2	0.0104	0.0025	0.418
NaCl	0.0176	0.0037	0.444
H_2SO_4	0.0098	0.0027	0.535
Tabbuk clay			
Natural	0.1510	0.0460	0.114
H_2O_2	0.0190	0.0002	0.890
NaCl	1.1360	0.3120	0.006
H_2SO_4	0.0058	0.0002	0.823
Bahhah clay			
Natural	0.0159	0.0037	0.198
H_2O_2	0.0428	0.0094	0.263
NaCl	0.1071	0.0058	0.287
H_2SO_4	0.0071	0.0007	0.629

Table 2: Freundlich equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K_F (L g^{-1})	n (-)
Khiber clay		
Natural	0.9199	4.0766
H_2O_2	0.0666	1.7782
NaCl	0.1459	2.0697
H_2SO_4	0.0687	1.8633
Tabbuk clay		
Natural	1.0550	5.5230
H_2O_2	0.0351	1.1261
NaCl	1.9005	8.9273
H_2SO_4	0.0140	1.1920
Bahhah clay		
Natural	0.3474	3.0708
H_2O_2	0.3299	2.4983
NaCl	0.5017	1.8356
H_2SO_4	0.0170	1.2488

Table 3: Langmuir-Freundlich equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K_c (L g^{-1})	b_c (L mg^{-1})	m
Khiber clay			
Natural	0.1278	0.0234	1.1867
H_2O_2	0.0431	0.0088	1.3549
NaCl	0.0693	0.0126	1.3608
H_2SO_4	0.0260	0.0053	1.2807
Tabbuk clay			
Natural	1.0558	0.0026	5.4867
H_2O_2	0.0946	0.0024	1.2877
NaCl	1.9051	0.0024	8.8986
H_2SO_4	0.1630	0.0159	1.8800
Bahhah clay			
Natural	0.2885	0.0211	2.5544
H_2O_2	0.3244	0.0146	2.3208
NaCl	0.1664	0.0090	1.0967
H_2SO_4	0.0936	0.0071	1.7414

The equilibrium parameters K_c, b_c and m were calculated by non-linear regression technique and presented in Table 3. The values of various constants indicate that the Langmuir-Freundlich equation correlated the equilibrium data well.

Toth Isotherm Model (TIM): The fourth model used was the Toth model. The Toth model can be written as follows (Unuabonah *et al*., 2007):

$$q_e = \frac{K_t C_e}{\left[1+(b_t C_e)^t\right]^{\frac{1}{t}}} \quad (6)$$

The toth parameters K_t, b_t and t are obtained using the non-linear regression technique with Eq. 6.

The equilibrium parameters K_t, b_t and t were calculated by non-linear regression technique and presented in Table 4. The analysis of data in Table 4 showed that Toth equation fits the data well.

Saad Isotherm Model (SIM): The section presents a novel isotherm model which is a modification of Langmuir model and is used instead of Langmuir model since the Langmuir model can not fit the experimental data well when the surface is heterogeneous. This model is called Saad model. This model has three parameters and it is used to fit experimental data on heterogeneous surface. The Saad model can be written as follows:

$$q_e = \frac{K_s C_e}{[1+b_s C_e]^{bb}} \quad (7)$$

where, bb = 1-s^2bs c_e.

The advantage of this model is the applicability for equilibrium experiments on homogeneous and heterogeneous surfaces. Where S is the heterogeneity parameter and when S = 0, this model converts to Langmuir model. It is an admitted fact that the Langmuir model fits the experimental data very well on a homogenous surface. But when the value of S<0, the model fits the experimental data well on a heterogeneous surface as seen from data in Table 5.

The Saad parameters K_s, b_s and S are obtained using the non-linear regression technique with Eq. 7. The equilibrium parameters K_s, b_s and S were calculated using a non-linear regression technique and presented in Table 5. The regression analysis showed that Saad equation fitted the experiment data very well.

BET isotherm model: This model describes the multilayer of solute adsorbed on the surface of the adsorbent (Sciban *et al*., 2007). The model can be written as follows:

Table 4: Toth equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K_t (L g^{-1})	b_t (L mg^{-1})	t
Khiber clay			
Natural	0.1418	0.0252	0.6627
H_2O_2	6.1915	0.0004	0.0863
NaCl	1.2439	0.0086	0.1514
H_2SO_4	0.0225	0.0026	0.4338
Tabbuk clay			
Natural	17.6593	2.8195	0.2428
H_2O_2	0.1495	0.0004	0.2448
NaCl	7.0451	1.2220	0.0002
H_2SO_4	0.2990	0.0126	0.2450
Bahhah clay			
Natural	30.5042	1.3985	0.1511
H_2O_2	6.8693	0.0902	0.1508
NaCl	2.5863	0.0037	0.1658
H_2SO_4	0.3739	0.0005	0.1428

Table 5: Saad equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K_s (L g^{-1})	b_s (L mg^{-1})	S
Khiber clay			
Natural	0.0710	0.0141	0.0000
H_2O_2	0.0104	0.0025	0.0000
NaCl	0.0177	0.0037	0.0000
H_2SO_4	0.0098	0.0027	0.0000
Tabbuk clay			
Natural	0.2670	0.0964	0.0264
H_2O_2	0.0268	0.0020	0.5711
NaCl	23.1995	8.8747	0.0030
H_2SO_4	0.0189	0.0094	0.1975
Bahhah clay			
Natural	0.0380	0.0139	0.0785
H_2O_2	0.0429	0.0094	0.0000
NaCl	0.1071	0.0058	0.0000
H_2SO_4	0.0071	0.0007	0.0000

$$q_e = \frac{K_B C_e}{(C_o - C_e)\,[1 + (b_B - 1)\,(\frac{C_e}{C_O})]} \quad (8)$$

The BET parameters K_B and b_B are obtained using the non-linear regression technique with Eq. 8.

The equilibrium parameters K_B and b_B were calculated by a non-linear regression technique and presented in Table 6. However, the data in Table 6 indicated that BET equation fitted the experimental data poorly.

Estimation the goodness of the fit: The data from the equilibrium models (Langmuir, Freundlich, Langmuir-Freundlich, Toth, Saad and BET) and the data from equilibrium experiments were evaluated by regression analysis and Chi-Square method. The adjusted coefficient of determination, $R^2_{adj.}$ was used to evaluate the goodness of the fit (Altin *et al*., 1998). In addition, Chi-square method (x^2) was applied to determine the agreements between the data from the model and the equilibrium experiments. The Chi-square (x^2) equation is written as follows:

$$x^2 = \sum_{i=1}^{N} \frac{\left(q_{i_{exp}} - q_{i_{calc}}\right)^2}{q_{i_{calc}}} \quad (9)$$

where, N is the number of data points, $q_{i_{exp}}$ is the amount of Co ion adsorption on the clay during the experiment and $q_{i_{calc}}$ is the calculated amount of Co ion adsorbed on to clay for a given data point i.

The ($R^2_{adj.}$) with (x^2) were calculated to assess the goodness of the fit between the data from the equilibrium models and the experiments. The calculated values of ($R^2_{adj.}$) and (x^2) were presented in Table 7 for all the three types of clays activated by various activators used in the experiment.

The ($R^2_{adj.}$) along with (x^2) show the most appropriate model to fit the data from different experiments. The ($R^2_{adj.}$) values varied from 0 to 1. However, if the value of $R^2_{adj.}$ is equal to one, the amount of Co ion adsorbed on the surface of activated clay is hundred percent. In the case of Chi-square method (x^2), the value of x^2 is small. This suggested that the results from the model is close to the result of equilibrium experiments and vice versa .

A review of study data showed considerable variations among these models as indicated from different values of $R^2_{adj.}$ and x^2. It was found that Langmuire model fitted the experimental data well for activated Khiber clay in all the cases. While, the Langmuir model fitted the experimental data well for Bahhah clay activated by H_2O_2, NaCl and H_2SO_4. This may be due to the fact that the Co ions adsorbed on the clay surface as a monolayer. While the Frendlich model and

Table 6: BET equilibrium parameters for the cobalt adsorption on three types of activated clay by different activators

Type of activated clay and activators	K_B (L g^{-1})	b_B (L mg^{-1})
Khiber clay		
Natural	0.0005	-1.3766
H_2O_2	0.0004	-0.6132
NaCl	0.0008	-0.6631
H_2SO_4	0.0003	-0.7040
Tabbuk clay		
Natural	0.0016	-1.4438
H_2O_2	-0.0029	0.2218
NaCl	0.0010	-0.6700
H_2SO_4	0.0001	-0.1129
Bahhah clay		
Natural	0.0004	-1.2562
H_2O_2	0.0039	-0.6544
NaCl	0.5476	-2.0141
H_2SO_4	0	1.8108

Table 7: Comparison of (R^2) and (x^2) for Langmuir, Freundlich, Langmuir-Freundlich, Toth, saad and BET models

Type of clay and activators	Langmuir		Langmuir-Freundlich		Freundlich		Toth		Saad		BET	
	x^2	R^2	x^2	R^2	x^2	R^2	x^2	R^2	x^2	R^2	x^2	R^2
Khiber clay												
Natural	0.671	0.844	1.024	0.716	0.721	0.828	0.745	0.819	0.671	0.844	516000	-5.0100
H_2O_2	0.456	0.917	0.752	0.842	0.765	0.862	0.738	0.849	0.456	0.917	23943	-0.2430
NaCl	0.523	0.907	0.962	0.813	0.844	0.855	0.900	0.830	0.523	0.907	2949	-0.9840
H_2SO_4	1.529	0.752	1.973	0.651	1.740	0.709	1.774	0.699	1.529	0.752	35240	- 0.3590
Tabbuk clay												
Natural	0.270	0.690	0.0602	0.937	0.060	0.937	0.102	0.890	0.03	0.967	192810	-11.5800
H_2O_2	2.364	0.947	1.578	0.950	1.730	0.901	1.803	0.899	1.413	0.95	-42160	0.4240
NaCl	0.857	0.286	0.586	0.526	0.586	0.525	0.673	0.448	0.226	0.823	1395000	-5.7540
H_2SO_4	1.497	0.969	0.9150	0.973	0.840	0.916	0.871	0.909	0.411	0.965	271300	0.3880
Bahhah clay												
Natural	0.710	0.797	0.185	0.926	0.181	0.928	0.197	0.916	0.154	0.940	1.72280	-3.3490
H_2O_2	0.111	0.961	0.386	0.869	0.380	0.878	0.327	0.889	0.111	0.961	10600	-1.7490
NaCl	1.710	0.924	3.177	0.857	2.159	0.909	2.921	0.871	1.710	0.924	346.67	0.3490
H_2SO_4	0.156	0.993	0.199	0.989	0.912	0.891	0.638	0.940	0.156	0.993	3030000	-0.4630

Langmuir-Frendlich model fitted the experimental data well for natural Tabbuk clay and activated tabbuk clay by NaCl. In addition, Frendlich model and Langmuir-Frendlich model fitted the experimental data well for natural Bahhah clay. Furthermore, it was found that Frendlich model fitted the experimental data well for activated Tabbuk clay by H_2O_2 and H_2SO_4. On relative basis, among all the three activated clays, the BET model proved the worst model in fitting the experimental data. This may be attributed to the absence of cobalt multilayer on the activated clays. Finally, it was observed that Saad model fitted all the experimental data well for three types of clays.

CONCLUSION

This study investigated the adsorption of Co ion on three types of clays activated by different activators to enhance their adsorption capacity. Several types of theoretical isotherms were used and evaluated. The Langmuir isotherm model agreed most favorably with the experimental data for Khiber clay in all the cases and for Bahhah clay activated by H_2O_2, NaCl and H_2SO_4. This happened, because the Co ion adsorbed on the clay as a monolayer adsorbate. The Frendlich model and Langmuir-Frendlich model fitted well with the experimental data for natural Tabbuk clay as well as Tabbuk clay activated by NaCl, H_2O_2 and H_2SO_4 and the natural Bahhah clay. Finally, a poor correlation showed that the BET model was not successful for the experimental data unsuccessfully for three activated clays. Because there was no indication of multilayer adsorption of Co ion on the surface of the activated clays. A new isotherm model, proposed by the author and called Saad, was compared with the five different isotherm models. Overall, it was found that newly developed Saad model fitted the experimental data well when compared with the other five models. Besides, when the value of S = 0, the Saad model fitted the data similar to Langmuir model as the Saad model converts to Langmuir model in case of activated and non-activated Khiber clay and the Bahhah activated clay.

NOMENCLATURE

M = Clay mass (g)
V = Volume of the solution (L)
q_e = Amount of cobalt ions adsorption (mg g^{-1})
C_o = Initial solution concentration of cobalt ions (mg L^{-1})
C_e = Concentration of the cobalt ions at equilibrium (mg L^{-1})
K = Equilibrium parameter of Langmuir model (L g^{-1})
b = Equilibrium parameter of Langmuir model (L mg^{-1})
$\acute{R}$ = Dimensionless equilibrium parameter (-)

K_F = Equilibrium parameter of Freundlich model ($L\ g^{-1}$)
N = Equilibrium parameter of Freundlich model (-)
K_c = Equilibrium parameter of Langmuir and Freundlich model ($L\ g^{-1}$)
b_c = Equilibrium parameter of Langmuir and Freundlich model ($L\ mg^{-1}$)
m = Equilibrium parameter of Langmuir and Freundlich model (-)
K_t = Equilibrium parameter of Toth model ($L\ g^{-1}$)
b_t = Equilibrium parameter of Toth model ($L\ mg^{-1}$)
t = Equilibrium parameter of Toth model (-)
K_B = Equilibrium parameter of BET model ($L\ g^{-1}$)
b_B = Equilibrium parameter of BET model ($L\ mg^{-1}$)
K_s = Equilibrium parameter of Saad model ($L\ g^{-1}$)
b_s = Equilibrium parameter of Saad model ($L\ mg^{-1}$)
S = Heterogeneity parameter of Saad model (-)

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