Batch experiments on the removal of U(VI) ions in aqueous solutions by adsorption onto a natural clay surface

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Abstract

The efficiency of the clay from Bikougou deposit (Gabon) as adsorbent for removing U(VI) ions dissolved artificially in aqueous solutions has been studied. Batch experiments have been performed for that by varying pH, U(VI) ions solution concentration, ionic solution strength, clay dosage, interaction duration and temperature. The U(VI) ions uptake per unit mass of clay, increases with the increase in pH (2-7), U(VI) ions solution concentrations and temperature. It decreases with the increase in ionic solution strength and clay dosage. The adsorption isotherm is best described by Dubinin-Kaganer-Radushkevich isotherm model. The kinetics of the adsorption of U(VI) ions on clay surface follows the pseudo-second-order kinetic model and the interaction realized spontaneously is exothermic. The mean values of thermodynamic constants $\Delta H°$, $\Delta S°$ and $\Delta G°$ obtained at 308k are respectively-62.54kJ/mol, -0.18kJ/K.mol and -8.68 kJ/mol.

Keywords: Adsorption capacity, adsorbent, U(VI) ions solution, isotherm model, distribution constant

1. Introduction

Among heavy metals presenting antagonist effects again nature and human health, uranium is known as one of the most dangerous (Agency for Toxic Substance, 1990). The reason is its chemical toxicity and radioactive nature (Francis et al., 1999)

Guanghui Wang et al., 2010). Uranium disseminated in the nature through mining and milling procedures (Krestou et al.,2004), applications of phosphate fertilizers to soils (Yamaguchi et al.,2009) and development of nuclear industries (Mellah et al., 2006) can induce a reduction of biodiversity and manifestation of various forms of cancers and kidney toxicity for its human exposure (Christine Chin Choy et al., 2006) The uranium ore deposits are scarce and unequally distributed in the world but operations such as exploitation of uranium-mine, its concentration, purification, accidents into nuclear power-stations and testing of nuclear weapons have increased the uranium presence in the environment. The knowledge and sensitization today on the environmental and human health effects of uranium have conducted to be developed methods for to reduce uranium exposure. In environmental media where the common form of uranium is U(VI) well known as being cancerigenic (Yusan and Akyil 2008), its removal in aqueous effluents is possible by using one of developed methods for water purification and recycling such as: reverse osmosis, nanofiltration, solvent extraction, ion exchange and adsorption (Parekh, 1988; McNulty, 1984; Nemerow and Dasgupta, 1991; Samuel and Osman, 1987). Each of them has its limits and advantages in application. Adsorption is low-cost method compared to the other preceding methods which are expensive. It is also the most efficient method in terms of simplicity in feasibility with high potential for the removal, recovery and recycling of metals ions from wastewaters (Imran and Gupta, 2006).

Several studies concerning the application of the adsorption method for U(VI) removal from aqueous solutions using adsorbents of various materials are reported. The U(VI) adsorption capacities of activated carbons,

zeolite, olivine rock, coir pith, smectite, biomass, goethite, polymeric materials, pure and modified clays have been evaluated (Joseph et al., 2011). These studies revealed that adsorption process depend of adsorbents characteristics and of solution properties.

Some of the preceding adsorbents are expensive to be used in routinism adsorption experiments. The tendency of research is to obtain low-cost and available adsorbents from industrial and agricultural wastes (Pehlivan et al., 2009; Lohani et al. 2008). Considered as low-cost adsorbents natural clays have been found as alternative to high-cost adsorbents .This family of materials, constituted of mixtures of clayey minerals and other adsorbent species in various rates has received much interest in adsorption application. A number of works mention the adsorption of heavy metals ions onto natural clay surfaces (Eba et al., 2010, Kamel et al., 2004). Saad Mohamed, 2010). The results obtained showed the high adsorption capacity of these clay categories and also that adsorption isotherm is described by the Langmuir and/or Freundlich isotherm models and kinetics is often obeying to pseudo-second–order kinetic model.

In this study, batch experiments have been performed to determine the adsorption capacity of the clay from Bikougou (Gabon 11°37' E and 1°58' N) for the removal of U(VI) ions in aqueous solution.

2. Materials and methods

2.1 Adsorbent characterization

The natural clay from Bikougou deposit used as adsorbent has been previously characterized. By combining the X-ray powder diffractogram results (Figures 1 and 2) and the whole-rock chemical analyses (Table 1), the modal compositions have been calculated .according a constrained multi-linear calculation (Eba et al., 2011). It indicated that the crude clay of Bikougou deposit is constituted mainly of 29.5% of kaolinite, 20.0% of albite, 16.0% of montmorillonite and 8.0% of illite and its other characteristics are presented in Table 2 (Eba et al., 2010).

2.2 Adsorption procedure

Batch adsorption experiments were realized by mixing with a constant speed in a 250 mL conical flask 0.15 mg of clay and 50mL of U (VI) ions solution of various concentrations (from 5 to 40 mg/L).

After shaking the flask for an appropriate time, the reaction suspensions were filtered and concentrations of residual U (VI) ions in the filtrates were determined by using ICP-AES.

2.3 Calculation

The amount of U(VI) ions adsorbed per unit mass of clay: q_e (mg/g) at the adsorption equilibrium is given by the equation:

$$q_e \text{ (mg/g)} = (Co - Ce)*V/m$$

Where: Co and Ce are respectively initial and at the adsorption equilibrium concentrations (mg/L) of U(VI) ions solutions. V is the volume (L) of U(VI) ions solutions and m (g) is the clay mass.

3. Results and discussion

The impact of many parameters such as: pH, initial concentrations of U(VI) ions solution , adsorbent mass, ionic strength of U(VI) ions solutions, duration of contact between the U(VI) ions solutions and the clay and the

temperature on the adsorption performance has been investigated.

3.1 Effect of the pH of U (VI) ions solutions.

The impact of the pH of the U(VI) ions solutions on their adsorption on natural clay from Bikougou was investigated by mixing 50 mL of U(VI) ions solutions of 35.5 mg/L and 0.15 mg of clay at the pH range 1-7 for 120 minutes. The results have been reported in the Figure 3.

It is shown that the amounts of U(VI) ions adsorbed increased continuously with the increase in pH from 1 to 7. The amounts of U(VI) ions adsorbed below pH 3 have been found weak compared to high values obtained above pH 3. Similar results have results have been reported by Sachs and Bernhard (2008).

This U(VI) adsorption behaviour might be explained if it is considered sides of weak and high values pH. In the low pH side (pH < 3), there is an excess of H_3O^+ ions in solution which compete repulsively with the U(VI) ions for the attainment of the available adsorption sites on the clay surface. When the pH increases, the uptake of H_3O^+ ions decreased strongly, making progressively more and more sites on clay surface available for the exchange with U(VI) ions. The hydrolysis and precipitation of U(VI) ions occur above pH 3 forming UO_2OH^+, $(UO_2)_2(OH)_2^+$, $(UO_2)_3(OH)_5^+$ complexes and a precipitate of $(UO_2)_2(OH)_2$ which increase the U(VI) removal from aqueous solutions (Parab et al., 2005).

3.2 Effect of the initial concentrations of U(VI) ions solutions

The variations of the amounts of U(VI) ions adsorbed per unit mass of clay as function of the Initial concentrations of U(VI) ions solutions have been studied by mixing 0.15 mg of clay and 50mL of U(VI) ions solutions of various concentrations remained between 5.11 and 35.52 mg/L. As it is shown in Figure 4, the amounts of U(VI) ions adsorbed increased gradually with the increase in initial concentrations of U(VI) ions aqueous solutions. This result is similar to those reported by Guanghui Wang et al. (2010) and Barger and Koretsky (2011) respectively in their studies on adsorption of U(VI) from aqueous solution on calcined and acid-activated kaolin and on the influence of citric acid, EDTA, and fulvic acid on U(VI) sorption onto kaolinite

These results correspond to the fact that at relative low U(VI) ions concentrations, the number of available adsorption sites on the adsorbent surface is relatively higher than of the number of U(VI) ions given by the adsorbate solution and consequently the amount of U(VI) ions adsorbed is weak in this case. In the opposite, when the concentration of U(VI) solutions is progressively high, the adsorption becomes more and more efficient, because of the increase of U(VI) ions number liberated by U(VI) solutions which compete to adsorption sites (Gupta et al., 2003).

3.3 Effect of ionic strength of U (VI) ions solutions.

It is possible that salts exhibit a significant effect on the adsorption process.In this study, potassium chloride (KCl) was selected as salt model for testing its effect on the adsorption capacity of the clay from Bikougou for U(VI) ions. The experiments consisted to add from 0.0001 to 0.1 mole of KCl to one litre of U(VI) solution of fixed concentration (35.52 mg /L), before mixing 50ml of the preceding solution with 0.15 mg of clay for 120 minutes. The results shown in Figure 5, demonstrated that the U(VI) uptake adsorbed on solid phase decreased by increasing the concentrations of potassium chloride. Since, the presence of KCl in the solution reduces the electrostatic interaction of opposite charges of clay and U(VI) ions. For this reason, the amount of U(VI) ions adsorbed decrease with increase of KCl concentrations (Mahir et al., 2008).

3.4 Effect of clay dosage

The adsorbent dosage is an important parameter because it is associated to the determination of the capacity of an adsorbent for a given concentration of adsorbate.

The adsorbent dosage experiments were performed by varying the mass of clay from 0.05g to 0.5 to interact with 50 mL of U(VI) (35.52 mg/L) solutions. The results reported in Figure 6 revealed that the amounts of U(VI) ions adsorbed decreases by increasing in the adsorbent mass. Similar results are reported by other authors (e.g. Pb(II), Cd(II) and Ni(II) by kaolinite and montmorillonite, Gupta and Bhattacharyya, 2008, Cr(III) by ion exchange resins, Rengaraj et al., 2003). These results may be attributed   (i) to the availability of less binding sites on the surface at higher concentrations of adsorbent for complexation of U(VI) ions (ii) and /or to the eventual formation of curds at higher values of adsorbent mass which reduce the available surface for the adsorption process (Shukla et al., 2002).

3.4 Adsorption isotherm studies

The good description, at a fixed temperature, of the adsorptive process for the removal of solute from solution onto adsorbent, depends of successful applicability of adsorption isotherm model to experimental data. An adsorption isotherm model is expressed by an equation which contains some constant parameters describing the surface properties and affinity of the adsorbent. Three kinds among several isotherm equations were tested to fit the experimental data:

Langmuir equation: $q_e = C_e*b*q_m / (1 + b*C_e)$ (Langmuir, 1918)

Freundlich equation: $q_e = k_F \ln C_e^{1/n}$ (Gupta and Bhattacharyya, 2008)

Dubinin-Kaganer-Radushkevich (D.K.R.) equation $q_e = X_m e^{-\beta*\varepsilon^2}$ (Karapinar and Donat, 2009).

where qe is the amount adsorbed at equilibrium (mg/g) and Ce is the equilibrium concentration .

The other parameters are different isotherm constants, which are calculated by regression of the experimental data. From the Langmuir equation, q_m (mg/g) and b represent respectively the maximum monolayer adsorption capacity and constant to adsorption energy. In the Freundlich equation, k_F and n indicate the maximum multilayer adsorption capacity and constant of adsorption intensity respectively. X_m, β and ε are the DKR parameters corresponding to the maximum adsorption capacity and the activity coefficient to the mean energy of adsorption. The Polanyi potential is given by the expression:

$$\varepsilon = RT \ln(1/1+C_o)$$

where R is gas constant (kJ/mol*K) and T the temperature (K).

In the Figure 7, there are presented plots comparing the variations of the equilibrium amounts of U (VI) ions adsorbed versus equilibrium concentrations of U(VI) ions solutions between experimental and isotherm models. The isotherm model constants and their correlation coefficients (R2) are shown in the Table 3. The best fit of experimental data is obtained with the isotherm model giving highest correlation coefficient value.

The results showed that the correlation coefficient value higher than 0.999 indicates there is a very strong positive relationship for the data and also that the interaction U(VI)-clay is well described by using the DKR adsorption isotherm model. The values of saturation limit X_m representing the total specific microspore volume

and the sorption energy E, calculated through the following equation:

$$E = -(\,2\beta\,)^{-0.5}$$

are reported in the Table 3. The E value (7.9 kJ/mol) was found in the range of that from the adsorption of Mn^{2+} (8.8 kJ/mol at 303 K) by natural zeolite with moderate strong U(VI) ions-clay bonding for the chemical controlled process (Saeed et al., 2003).

The Freundlich isotherm parameters k_f and n (Table1) obtained with correlation coefficient higher than 0.96 illustrate the good applicability of the non specific adsorption of U(VI) ions on the heterogeneous clay surface. The Freundlich constant: n found higher than unity corresponds to favourable adsorption of U(VI) on the clay surface.

The description of the adsorption behaviour of the U(VI) ions on clay has been tested by using Langmuir isotherm , but data were not found in good agreement as those of the DKR and Freundlich models. The Langmuir monolayer maximum capacity: q_m.

And the Langmiur energetic constant b have been obtained with correlation coefficient of 0.59 (Table1).

Among the three isotherm models tried, the best fit of experimental isotherm adsorption data was obtained through the application of DKR and Freundlich isotherm models.

3.5 Determination of the kinetic order

The results found on the variations of the amounts of U(VI) ions adsorbed per unit mass versus time for the adsorption of U(VI) ions onto natural clay of Bikougou were: 1.65 and 3.47 mg/g after 10 minutes of interaction, 1.68 and 3.47 mg/g after 20 minutes of interaction, 1.68 and 3.50 mg/g after 40 minutes of interaction, 1.68 and 3.52 mg/g after 60 minutes of interaction, 1.69 and 3.53 mg/g after 120 minutes of interaction respectively for 5.11 mg/L and 10.79 mg/L of initial solution of U(VI) ions.

The amounts of U(VI) ions adsorbed increase by the increasing of interaction duration. Similar results have been reported in the literature (Anirudhan et al., 2010). The process is fast reason why that more of the half of the initial U(VI) ions in solution has been consumed after 10 minutes of equilibration. The equilibrium of the adsorption has been obtained after 120 minutes of interaction.

The experimental variations of the adsorption capacities as a function of time were modelized for testing their adhesion to the pseudo-first-order kinetic (Ho, 2004) and pseudo-second-order kinetic ((Ho et al., 2001) models. The pseudo-first-order kinetic model is expressed by the equation:

$$Ln(\,q_e - q_t\,) = Lnq_e - k_1 t$$

And the pseudo-second-order kinetic model is given by the expression:

$$t/q_t = \tfrac{1}{2} * k_2 * q_e^2 + t/q_e$$

Where k_1 (min^{-1}) and k_2 (g/mg.min) are respectively the pseudo-first order kinetic and the pseudo-second order kinetic model rate constant s. The applicability of a kinetic model to the adsorption process is supported by (i) an obtainment of kinetic linear plots model with highest values of correlation coefficients (ii) a good agreement between experimental and modelized values of equilibrium adsorption capacity and (iii) plots going through to the origin.

Figures 8 and 9 illustrate the applicability respectively of the kinetics of pseudo-second- order and pseudo-first-order models.

The kinetic parameters are reported in the Table 2. The pseudo-first-order kinetic rate constant k_1 (min^{-1}) and the adsorption equilibrium amount of U(VI) ions adsorbed have been determined with correlation coefficient varying from 0.84 to 0.96 while the initial concentrations of U(VI)ions solutions were remained from 5.11 To 35.53 mg/L. The rate constant values were remained between 0.200 and 0.086 min^{-1}. The modelized values of equilibrium amounts of U(VI) ions adsorbed were found varying from 0.04 to 1.04 mg/g and several time weaker than of the corresponding adsorption experimental data. It is notified that plots do not pass through the origin.

The straight-line plots related to the kinetics of second order have been realized with correlation coefficients ranged between 0.98 and 1.00. The values of the second order rate constant were found remained between 0.022 and 0.218 g/mg.min. The calculated values of the adsorption capacities (1.69 to 11.64 mg/g) were close to the values of experimental data. The percentage of deviation was found varying between -0.081 to -0.360. It is observed that these plots go through the origin.

Therefore, the adsorption of the U(VI) ions on the clay surface followed better the pseudo-second order kinetics than the pseudo-first order kinetics. The pseudo-second order kinetics assumes that the rate limiting step may be chemical adsorption. In this case generally, the pseudo-second-order rate constant (k_2) decreases with the increasing metal concentration, as it was reported in certain studies before (Anirudhan and Radhakrishnan, 2007; Anirudhan et al., 2010). Furthemore, the activation energy E_a, calculated from slope of Arrhenius equation:

$$\text{Ln}k_2 = -5895.4/T + 17.628 \quad (R^2 = 0.99)$$

was found as 49.21kJ/mol. This value of activation energy is without the energy range (0–40 kJ/mol) of physical process (Mellah et al., 2006; Ho et al., 2000). Then, the type of adsorption of U(VI) ions onto clay from Bikougou according to the value obtained is chemical.

3.6 Thermodynamic studies

The adsorption studies were carried out at different temperatures: 25°C, 35°C, 45°C and 55°C.The adsorption standard free energy was calculated from:

$$\Delta G° = -RT\text{Ln}K_d$$

where k_d, the distribution constant of U(VI) ions between liquid and solid phases is given as :

$$K_d = q_e/C_e \text{ (L/g)}$$

R is the gas constant (8.314 J/K.mol) and T is temperature
The standard enthalpy ($\Delta H°$) and standard entropy ($\Delta S°$) changes related to the adsorption of U(VI) ions on the clay surface were calculated from Van't Hoff equation

$$\text{Ln}k_d = -\Delta H°/RT + \Delta S°/R$$

as respectively   the slope and intercept of linear plot Ln k_d versus T^{-1}(K) (Table 5).

The negative mean value of ΔH^o (-62.538 kJ/mol) is in accordance with the exothermic nature of the interaction. Its level could be considered as corresponding to the case of moderate strong U(VI) ions-clay bonding. The negative value of the entropy change (-174.89 J/mol.K) expressed the fact that the U(VI) ions in the solid state are at more ordered distribution than they are in aqueous solutions.

The negative Gibbs energy values indicated the spontaneous nature of the adsorption of the U(VI) ions on the clay surface. The ΔG^o values increased from –10.424 to –5.17 kJ/mol by increasing in temperature from 298 to 328 K. Thus, the surfactant species have more affinity towards U(VI) ions at lower temperature than at related elevated temperature as it was reported by Donat (2010) for the removal of uranium (VI) from aqueous solution onto natural sepiolite (negative values of thermodynamic parameters: $\Delta H° = -126.64$ kJ/mol, $\Delta S° = -354.88$ J/mol and $\Delta G° = - 21.14$ kJ/mol). Negative values of thermodynamic parameters have been also obtained by Mellah et al.,(2006) from the study of the adsorption of U(VI) from aqueous solution on activated carbon. The enthalpy and entropy changes determined by Guanghui Wang et al., (2010) after the adsorption of U(VI) ions on modified kaolin were positive except the free energy which was found negative ($\Delta H° = 3.68$ kJ/mol; $\Delta S° = 27.48$ J/K.mol; $\Delta G° = -4.664$ kJ/mol).

4. Conclusion

The results show that the natural clay from Bikougou exhibits a high adsorption capacity to be used as adsorbent for the removal of U(VI) ions in aqueous solutions. The amounts of U(VI) ions adsorbed increase with the increase both in pH and initial concentrations of U(VI) ions, and decrease with the increase in ionic strength of the U(VI) ions solution and mass of the adsorbent. The experimental adsorption data are fitted both by DKR and Freundlich isotherm models. The pseudo-second-order kinetic model has shown a best adhesion to experimental data for determining both the order and the mechanism of the adsorption process. The adsorption process is spontaneous between 298 and 328 K. It is accompanied by an exothermic thermal exchange.

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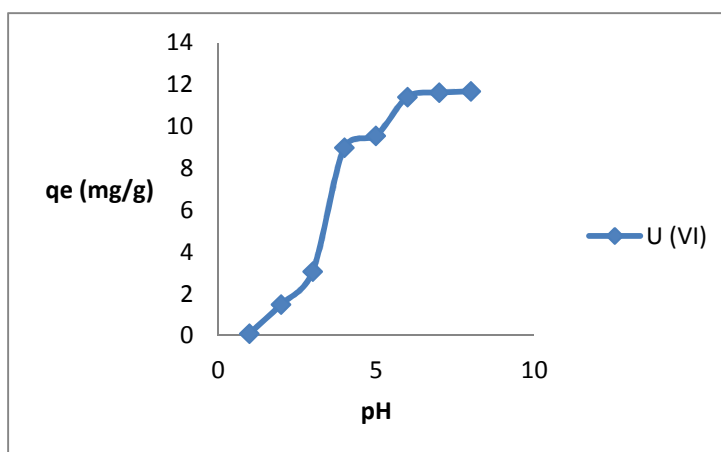
Figure 3: Amount of U(VI) ions adsorbed per unit mass (qe) on clay at different pH (clay mass 0.15g; C_0 = 35.525mg /L and interaction time 120 min   at 25°C)

Table 4: Kinetic models: parameters and correlation coefficients for the determination of order

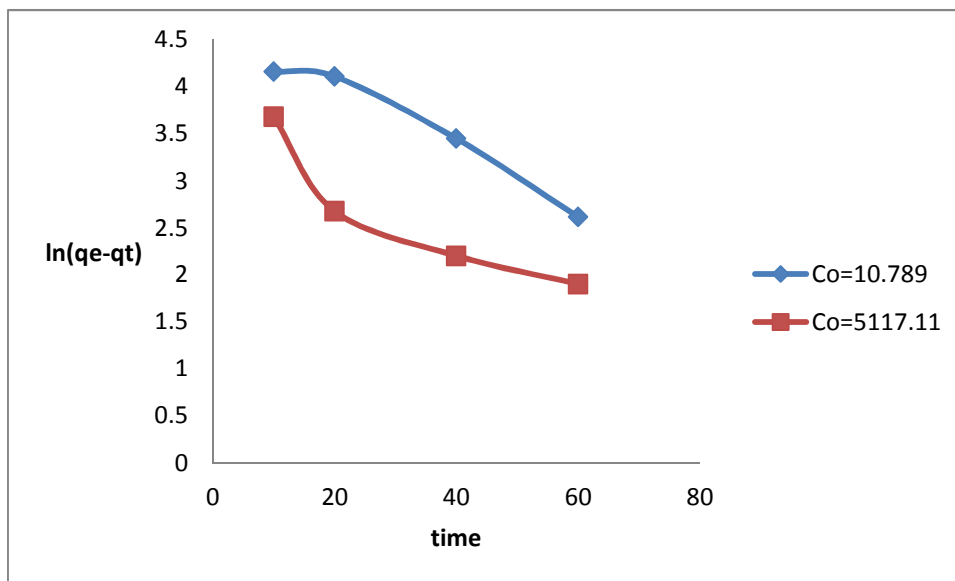| Initial concentration (mg/L.K)	q_e experimental (mg/g)	Pseudo-first order			Pseudo-second order		
		q_e (mg/g)	k_1	R^2	q_e (mg/g)	k_2	R^2
5.1117	1.69024	0.04	0.0322	0.838	1.6934	0.218	1
10.789	3.5316	0.1016	0.032	0.964	3.5410	0.080	1
19.9193	6.4808	0.094	0.0208	0.92	6.4893	0.0667	1
35.525	11.5986	1.04	0.0859	0.943	11.6414	0.0217	0.99

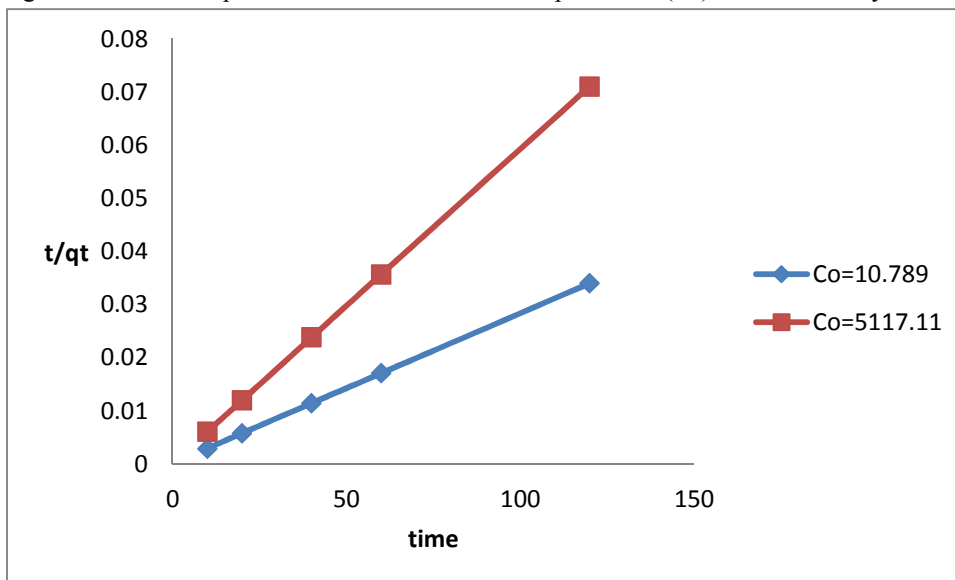Figure 9: kinetics of pseudo-first order for the adsorption of U(VI) ions on the clay surface.

Figure 8: kinetics of pseudo-second-order for the adsorption of U(VI) ions on the clay surface.
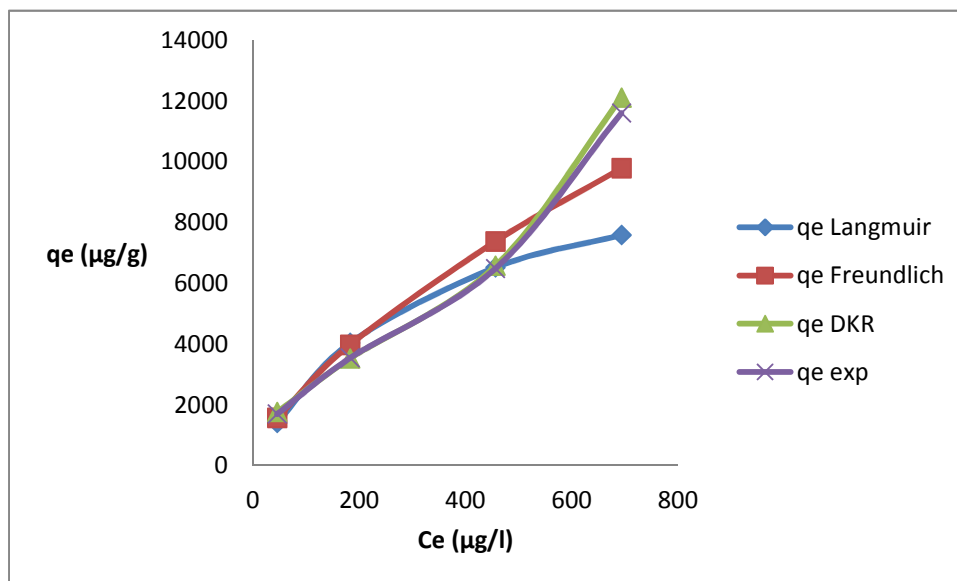
Figure 7: Amounts of U(VI) ions adsorbed per unit mass on clay versus equilibrium adsorbate solution: comparison between the equilibrium experimental data and those calculated by using the Langmuir, Freundlich and DKR models.

Table 3: Parameters and statistical of Langmuir, Freundlich and DKR isotherm models (pH=5; 0.15 g of clay; 308K; 50 ml of 35.52 mg/L of U(VI) solution interaction time 120 min)

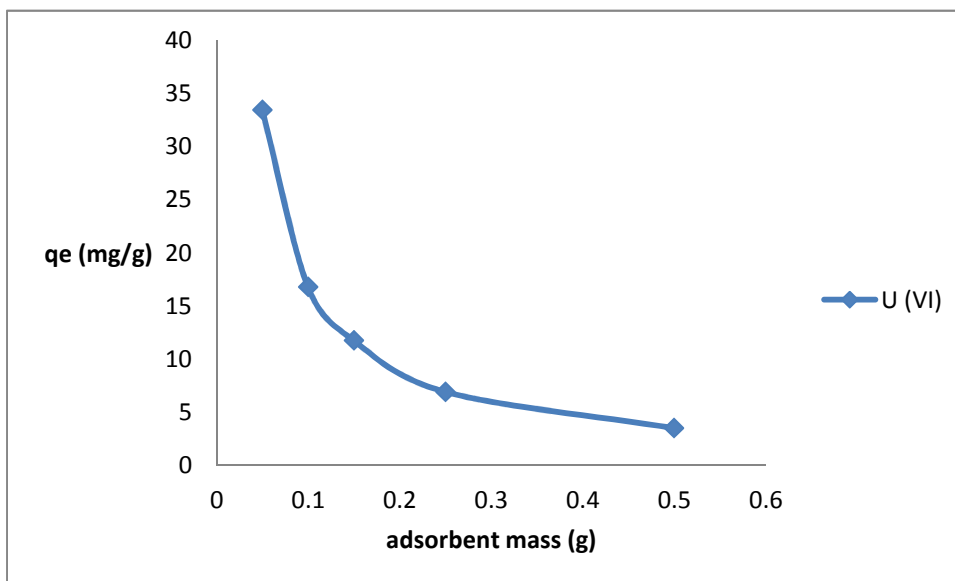| Langmuir model			Freundlich model			DKR model		
qm (mg/g)	b (L/g)	R^2	K_F	n	R^2	Xm (mg/g)	E (kJ/mol)	R^2
11.111	3.1	0.59	116.268/1000	1.476	0.968	38.168/1000	7.9	0.9999

Figure 6: Variations of amount of U(VI) ions adsorbed per unit mass as a function of adsorbent dosage (equilibrium time 120 min , initial concentration 35.52 mg/L at 25°C)
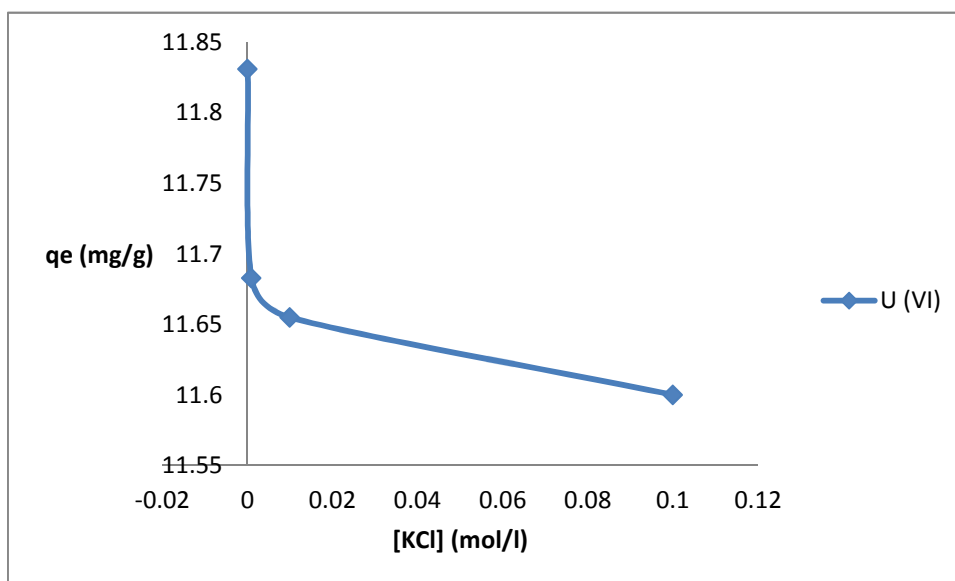

Figure 5: Amount of U(VI) ions adsorbed per unit mass of clay for four values of ionic strength (0.001, 0.01, 0.025, 0.1 mg/L; equilibrium time 120 min; adsorbent dose 3 g/L and temperature 25°C)
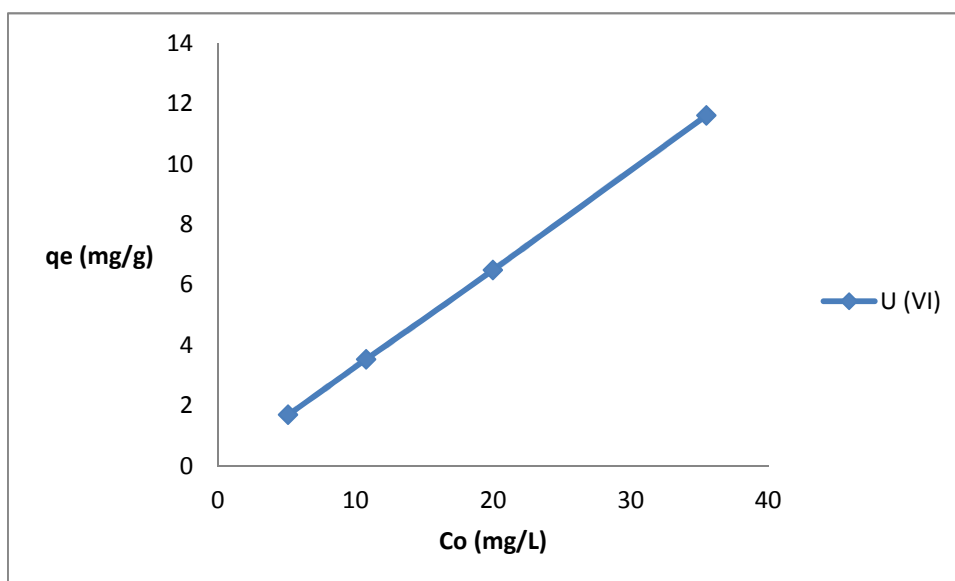
Figure 4: Amount of U(VI) ions adsorbed per unit mass (qe) on clay for four different initial uranium concentrations (5.11, 10.78, 19. 92, 35.53 mg/L; clay mass 0.15g, interaction time 120 at 25°C min)

Table 5: Thermodynamic parameters and statistical for adsorption of U(VI) (pH =5; 0.15g clay of clay mass and 120 min of interaction)

Co (mg/L)	$\Delta H°$ (kJ/mol)	$\Delta S°$ (J/mol.K)	$\Delta G°$ (kJ/mol)				R^2
			298	308	318	328	
5.117	-60.575	-168.05	-10.51	-8.82	-7.14	-5.45	0.988
10.789	-64.501	-181.735	-10.34	-8.53	-6.71	-4.89	0.943

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