



# Sorption behavior of cesium on Ain Oussera soil under different physicochemical conditions

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## ABSTRACT

In the present study, the sorption behavior of cesium was investigated in Ain Oussera soil around the Es-Salam reactor facility. This study was conducted using batch method under different physicochemical conditions including contact time, ionic strength, pH, solid/liquid ratio and temperature. The results showed that sorption followed pseudo-second-order kinetics with a good regression coefficients ( $R^2 = 0.999$ ). The activation energies were 11.26 and 15.21  $\text{kJ mol}^{-1}$  which correspond to ion-exchange-type sorption mechanism. The adsorption was favored at low temperature and it was exothermic ( $\Delta H^0 < 0$ , with average value of  $-1.97 \text{ kJ mol}^{-1}$ ) and spontaneous ( $\Delta G^0 < 0$ , with average value of  $-11.97 \text{ kJ mol}^{-1}$  at 23 °C and  $-13.2 \text{ kJ mol}^{-1}$  at 60 °C). The presence of competing cations such as  $\text{K}^+$  and  $\text{Ca}^{2+}$  ions in groundwater can significantly reduce the Cs adsorption onto soil. Desorption reaction was also investigated using three reagents with different ionic strengths (deionized water, groundwater and 0.1 M KCl solution). The results showed that Cs ions were preferentially distributed onto high affinity sorption sites.

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## 1. Introduction

In Algeria and precisely in the south of Algiers, about 200 km near the town of Ain Oussera, there is a nuclear research reactor (Es-Salam reactor). Therefore, there is always a risk of soil contamination around this facility in case of an accidental release of radioactive waste. Among major fission products, cesium is one of the main constituents of wastewater effluent. It is also of concern from an environmental standpoint because of its high solubility and long half-life of its radioactive isotopes, cesium-137 (30 years) and cesium-135 ( $2.9 \times 10^6$  years) [1]. The mobility of Cs can be substantially reduced by interaction with clay minerals, sediments and rocks [2,3]. This mobility is also sensitive to soil type and its components such as clay, organic matter etc. The behavior of Cs in soils is particularly important in selecting sites of nuclear facilities and waste repositories where the essential characteristic sought must be its ability to retard the radionuclide migration.

The main objective of the present work is to study, using a batch method, the sorption behavior of Cs in Ain Oussera soils near the Es-Salam reactor and the factors that control Cs sorption. Adequate determination of Cs sorption mechanics on soil is important for

understanding the fate of such probable pollution and establishing a groundwater surveillance program and will facilitate successful environmental remediation procedure [4].

The Ain Oussera soil in the vicinity of the Es-Salam reactor has approximately the same physicochemical properties within a radius of 2 km. It is a sandy soil poor in organic substance with 10% of clay minerals consisting of montmorillonite, illite and kaolinite. In this work, a series of batch experiments were conducted to investigate Cs sorption under different physicochemical conditions. In order to examine the degree of sorption reversibility and the Cs-soil adsorption mechanism, desorption reaction was investigated.

## 2. Materials and methods

### 2.1. Soil and chemicals

The adsorption studies were carried out on four soil samples with a superficial depth of 50 cm, collected from the south, north, east and west of Es-Salam reactor facility then air-dried at room temperature during 48 h and finally sieved at 2 mm.

Particle size analysis was made using Robinson's pipette method [5] and soil pH was measured in 1/5 soil/solution suspension both for pH ( $\text{H}_2\text{O}$ ) and pH (KCl) [6]. Organic matter and carbonate have been determined using Walkly-Black wet digestion method [7] and Bernard Calcimeter method [5], respectively. The mean for  $n = 3$

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measurements were reported in all sorption/desorption experiments.

## 2.2. Adsorption/desorption batch experiments

The soil sorption capacity was investigated using batch method. Sorption experiments were performed in screw cap centrifuge tubes at room temperature (23 °C), except in the case of sorption evaluation as function of temperature. The soil/solution ratio was 0.1 g/50 mL throughout the sorption experiment (except where otherwise specified). 0.1 g of soil was added into 50 mL of 1 mg L<sup>-1</sup> CsCl in deionized water solution spiked with appropriate amount of radioactive <sup>134</sup>Cs tracer. The mixture was shaken up for 1 h (except where otherwise mentioned), centrifuged using a centrifuge (Bioblock scientific 2-5, SIGMA) at 3500 rpm then filtered through a 0.2 μm cellulose nitrate filter (Sartorius). The radioactivity of the solution before and after mixing was counted with high-purity germanium detector (GL 0515R, Canberra). The counts were corrected for adsorption loss of the radionuclides on the walls of the tubes by running blank experiments (without soil added). The samples were counted for 5000 s, to reduce counting error.

Generally, the equilibrium distribution coefficient ( $K_d$ ) is defined as:

$$K_d = \frac{[Cs]_s}{[Cs]_l} \quad (1)$$

where  $[Cs]_s$  is the amount of cesium sorbed per unit mass of adsorbent (mg g<sup>-1</sup>) and  $[Cs]_l$  is the equilibrium solution concentration (mg mL<sup>-1</sup>).  $K_d$  can also be formulated in terms of activities using the following expression:

$$K_d = \frac{A_i - A_e}{A_e} \frac{V}{m} \quad (2)$$

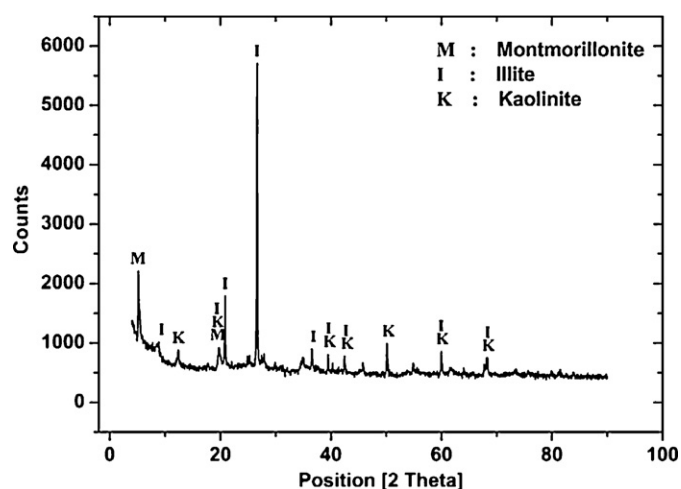
where  $A_i$  and  $A_e$  are the activities of radionuclide in solution at the beginning and at the end of sorption respectively,  $V$  is the volume in mL of the solution used for equilibration and  $m$  is the mass of adsorbent in grams.

The kinetic adsorption experiments were conducted according to shaking time ranging from 1 min to 50 h. It was performed at two different temperatures (23 and 60 °C) and for two different initial concentrations of Cs (1 and 100 mg L<sup>-1</sup>). To study the effect of ionic strength, a series of batch experiments were conducted with Cs loading (CsCl) of 1 mg L<sup>-1</sup>. The concentration of background electrolyte of four individual cations (Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>) was in the range of 10<sup>-5</sup>–1 M. The pH effect has been studied using a series of experiments and pH was carefully adjusted (from 2 to 12) by adding HNO<sub>3</sub> or NaOH solutions without altering the solid/liquid (S/L) ratios. In order to evaluate the effect of S/L ratio on cesium adsorption onto Ain Oussera soil, 0.05–2 g of soil was added into 50 mL of 1 mg L<sup>-1</sup> CsCl DW solutions. The suspensions were collected after 1 h shaking.

For desorption experiments, samples of Cs loaded soil, were separately mixed with three extracting reagents (DW, GW and 0.1 M KCl solution). Analysis of the eluted Cs was performed at 1 h, 3 h, 20 h, 50 h, 70 h, 100 h, 120 h and 7 days. The S/L ratio used in these experiments was 1 g/50 mL.

**Table 1**  
Physicochemical properties of Ain Oussera Soils.

| Sample | Location vs. nuclear facility | pH (H <sub>2</sub> O) | pH (KCl) | OM (%) | CaCO <sub>3</sub> (%) | Sand (%) | Slit (%) | Clay (%) |
|--------|-------------------------------|-----------------------|----------|--------|-----------------------|----------|----------|----------|
| 1      | North                         | 8.5                   | 7.9      | 0.51   | 3.3                   | 80.5     | 9.4      | 9.9      |
| 2      | South                         | 8.4                   | 8.0      | 0.53   | 3                     | 82       | 7.9      | 10.05    |
| 3      | East                          | 8.3                   | 7.9      | 0.53   | 2.95                  | 81       | 7        | 11       |
| 4      | West                          | 8.4                   | 7.9      | 0.55   | 3.0                   | 78       | 12.8     | 8.9      |



**Fig. 1.** XRD spectra of the fine fraction (<2 μm) of Ain Oussera soil (devoid of organic matter).

## 3. Results and discussion

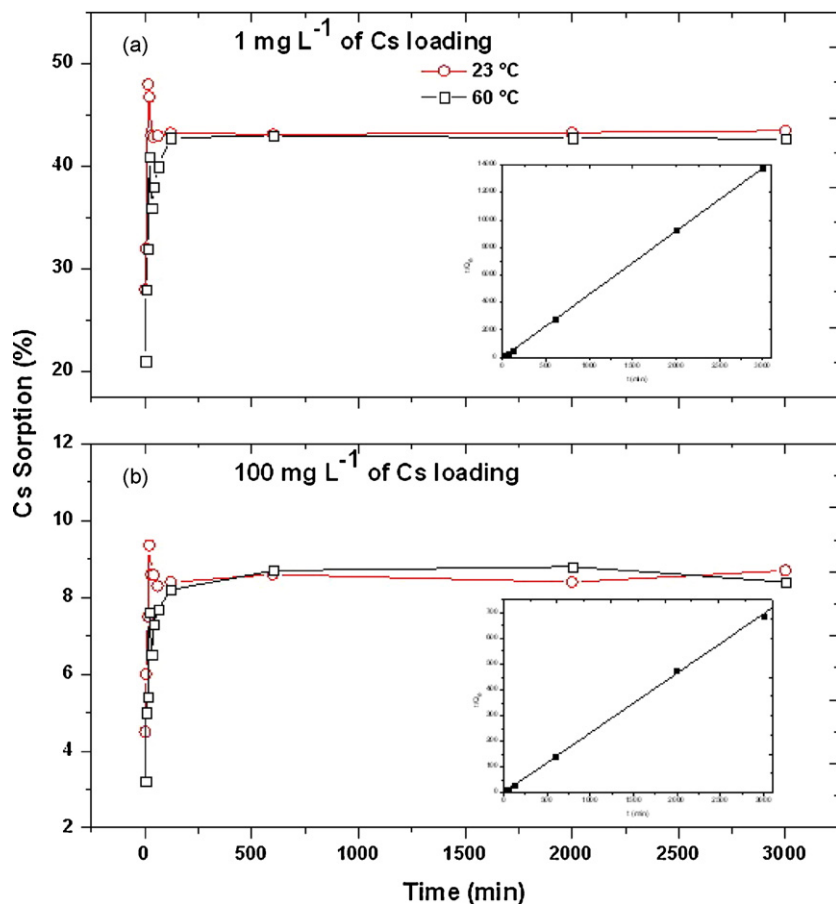
### 3.1. Soil analysis

A feature of the region soil is its extreme homogeneity; the composition remains markedly unchanged even at a distance of some kilometers. The results of physicochemical characterization of the four samples are shown in Table 1.

To determine the type of clay in this soil, its fine fraction (diameter <2 μm) was analyzed using X-ray diffractometer (PW1710) and the spectrum is shown in Fig. 1. According to this analysis, three clay minerals are mostly found in soil samples: montmorillonite, illite and kaolinite.

### 3.2. Kinetic analysis

The sorption was performed as function of contact time (Fig. 2). The rate of Cs sorption, on Ain Oussera soil, is fast for both initial concentrations and equilibrium is reached after mixing of 1 h. No significant change in sorption percentage was observed up to 50 h. Therefore, equilibrium time of 1 h was selected for all subsequent studies. This behavior is a typical adsorption feature of Cs onto minerals such as biotite, illite, hornblende, calcite and montmorillonite described by its speed and also by its partial reversibility [8]. This finding is in agreement with XRD analysis of Ain Oussera soil which revealed the existence of montmorillonite and illite. It is noticed that this phenomenon is related to the adsorption taking place at the accessible sorption sites on external surface of these minerals [2,3]. The instantaneous uptake of cesium by this soil indicates that ion-exchange may be the predominant sorption mechanism. On the other hand, the data indicate fast accumulation of the cesium ions at the adsorbent surface followed by a partial desorption leading to equilibrium attainment. A sorption reaction is believed to pass through three steps: film diffusion, particle diffusion, and finally exchange reaction at the sorption site [9]. Similar results were also reported by Shahwan et al. in a study including Cs retardation by



**Fig. 2.** Cs sorption as function of time at two distinct temperatures (23 and 60 °C). (a) Cs loading of 1 mg L<sup>-1</sup> and (b) Cs loading of 100 mg L<sup>-1</sup>. The inset figures give the corresponding linear fit using the pseudo-second-order equation.

natural kaolinite [10], therefore this suggests that three steps sorption is a typical behavior of the cesium sorption on kaolinite. We know that the XRD analysis revealed the existence of Kaolinite in Ain Oussera soil. Concerning the effect of rising temperature, it is noted that at high temperature (60 °C) sorption takes relatively more time to reach equilibrium.

In order to calculate the “apparent” rate constant, the following pseudo-first-order and pseudo-second-order equations [10] were used:

$$\frac{1}{[Q_e]} = \left( \frac{K_1}{[C_e]} \right) \left( \frac{1}{t} \right) + \frac{1}{[C_e]} \quad (3)$$

$$\frac{t}{[Q_e]} = \left( \frac{1}{K_2 C_e^2} \right) + \left( \frac{1}{C_e} \right) \quad (4)$$

where  $Q_e$  is the concentration of Cs<sup>+</sup> sorbed on the solid at time  $t$  (mg g<sup>-1</sup>),  $C_e$  is the concentration of Cs<sup>+</sup> sorbed at equilibrium (mg L<sup>-1</sup>),  $K_1$  is the pseudo-first-order equation rate constant (min<sup>-1</sup>), and  $K_2$  is pseudo-second-order equation rate constant (g mg<sup>-1</sup> min<sup>-1</sup>). The graph obtained using Eq. (3) shows that Cs adsorption kinetics does not agree with the pseudo-first-order equation at any concentration or temperature. However, applying

equation Eq. (4), the adsorption of Cs follows perfectly the pseudo-second-order equation with correlation coefficient close to unity. The insets in Fig. 2a and b show the change in  $t/Q_e$  versus  $t$ . The values of the linear correlation coefficient  $R^2$ , rate constant  $K_2$ , Cs concentration  $C_e$  and activation energy of sorption  $E_a$  at two distinct initial concentrations and at two temperatures are shown in Table 2. The activation energy was obtained using the following equation:

$$\ln \frac{K_2(T_2)}{K_2(T_1)} = \frac{E_a}{R} \left( \frac{1}{T_2} - \frac{1}{T_1} \right) \quad (5)$$

where  $R$  is the perfect gas constant (8.314 J mol<sup>-1</sup> K<sup>-1</sup>). The values of  $E_a$  were obtained as 11.26 and 15.21 kJ mol<sup>-1</sup> (for the Cs loading of 1 and 100 mg L<sup>-1</sup>, respectively). The activation energy can be conceived as the minimum kinetic energy required for a given reaction to take place. Indicating that the energetic barrier against sorption on Ain Oussera soil is easier to overcome, leading to rapid absorption. On the other hand, the  $E_a$  values are much below the energies corresponding to chemisorptions barriers (usually >40 kJ mol<sup>-1</sup>), suggesting that physical sorption is operating where the attractive forces are of Van Der Waals type in addition to weak electrostatic forces accompanying most ion-exchange reac-

**Table 2**

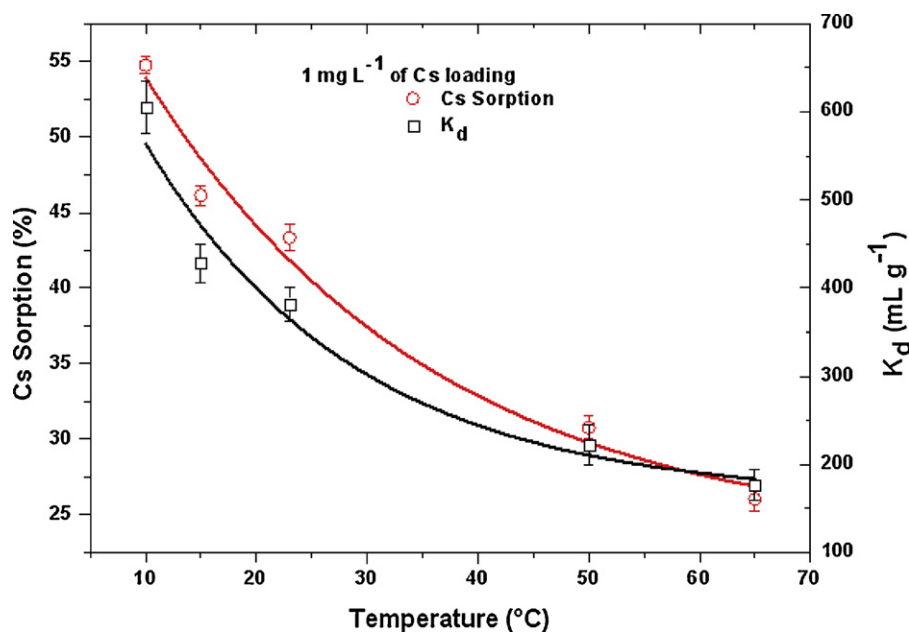
The values of  $R^2$ ,  $E_a$ ,  $C_e$  and  $K_2$  obtained from the linear fits of experimental data of second-order rate equation.

| Cs loading (mg L <sup>-1</sup> ) | T (°C) | $C_e$ (mg g <sup>-1</sup> ) | $K_2$ (g mg <sup>-1</sup> min <sup>-1</sup> ) | $R^2$ | $E_a$ (kJ mol <sup>-1</sup> ) |
|----------------------------------|--------|-----------------------------|---|-------|-------------------------------|
| 1                                | 23     | 0.217                       | 5.57  | 1.000 | 11.26                         |
|                                  | 60     | 0.211                       | 9.26  | 0.999 |                               |
| 100                              | 23     | 4.31                        | 0.075   | 0.999 | 15.21                         |
|                                  | 60     | 4.27                        | 0.150   | 0.999 |                               |

**Table 3**

Thermodynamic parameters calculated from the sorption data of Cs on Ain Oussera soil.

| Cs loading ( $\text{mg L}^{-1}$ ) | $K_d$ ( $\text{mL g}^{-1}$ ) (23 °C/60 °C) | $\Delta H^\circ$ ( $\text{kJ mol}^{-1}$ ) | $\Delta G^\circ$ ( $\text{kJ mol}^{-1}$ ) (23 °C/60 °C) | $\Delta S^\circ$ ( $\text{kJ mol}^{-1} \text{K}^{-1}$ ) (23 °C/60 °C) |
|-----------------------------------|--|---|---|---|
| 1                                 | 371.19/333.33                              | -2.38                                     | -14.56/-16.08   | 0.041/0.041   |
| 100                               | 45.25/41.71                                | -1.80                                     | -9.38/-10.32  | 0.025/0.025   |

**Fig. 3.** Distribution coefficient  $K_d$  and Cs sorption percentage as function of temperature.

tions [10]. When comparing the activation energies corresponding to the two initial concentrations of Cs (1 and 100  $\text{mg L}^{-1}$ ), it is obvious that at high concentrations, sorption is much less important due to the limited number of sorption sites compared to the high concentration of ions present in the suspension. This reflected by a higher value of energetic barrier against sorption (15.21  $\text{kJ mol}^{-1}$ ). In contrast, at the low initial concentration, lower value of  $E_a$  (11.26  $\text{kJ mol}^{-1}$ ) leads to higher sorption (Fig. 2a) because of availability of sorption sites. The  $K_2$  values also indicate that sorption with 1  $\text{mg L}^{-1}$  of Cs initial concentration is more effective than with 100  $\text{mg L}^{-1}$  (Table 2).

### 3.3. Thermodynamic study

The  $K_d$  value corresponding to sorption of Cs on soil at 23 and 60 °C and at a mixing period of 50 h are shown in Table 3. These values were used in the calculation of the enthalpy change of sorption ( $\Delta H^\circ$ ), the Gibbs energy of sorption ( $\Delta G^\circ$ ) and entropy of sorption ( $\Delta S^\circ$ ), by applying the following equations:

$$\Delta H^\circ = R \frac{T_1 T_2}{T_2 - T_1} \ln \frac{K_d(T_2)}{K_d(T_1)} \quad (6)$$

$$\Delta G^\circ = -RT \ln K_d \quad (7)$$

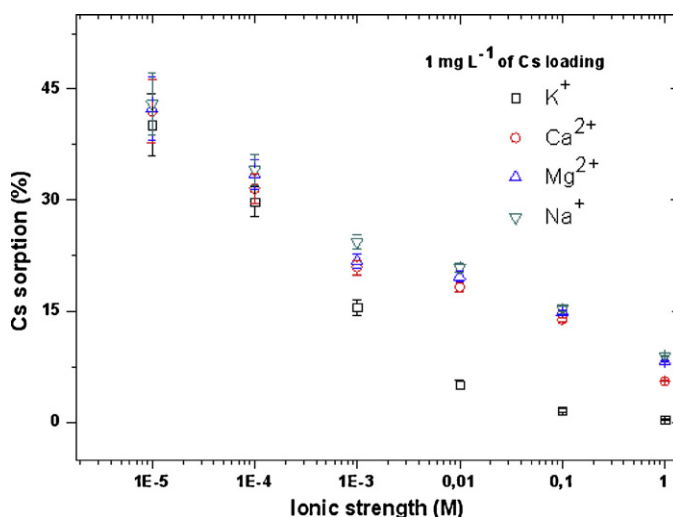
$$\Delta S^\circ = \frac{\Delta H^\circ - \Delta G^\circ}{T} \quad (8)$$

According to the values of  $\Delta H^\circ$  and  $\Delta G^\circ$  (Table 3), the sorption is exothermic, spontaneous and is favored at low temperatures. In order to elucidate this behavior, a study of sorption as a function of temperature was performed at 10, 20, 30, 50 and 60 °C. The result indicates that sorption increases with decreasing temperature (Fig. 3). This behavior could originate from thermal stabilization leading to an increase of the mobility of the  $\text{Cs}^+$  ion on the surface of the solid as the operating temperature is increased, thus

enhancing the desorption steps [10]. For Ain Oussera soil, the values of activation energies (11.26 and 15.21  $\text{kJ mol}^{-1}$ ) are within the energy range 8–16  $\text{kJ mol}^{-1}$ , corresponding to ion-exchange-type sorption mechanism [11], which facilitates the Cs desorption at high temperature.

### 3.4. Effect of ionic strength

To study the effect of ionic strength on Cs adsorption in Ain Oussera soil, a series of batch experiments were carried out with competing cations of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  with concentration ranges from  $10^{-5}$  to 1 M. The four cations were selected due to

**Fig. 4.** Cs sorption percentage as function of ionic strength.

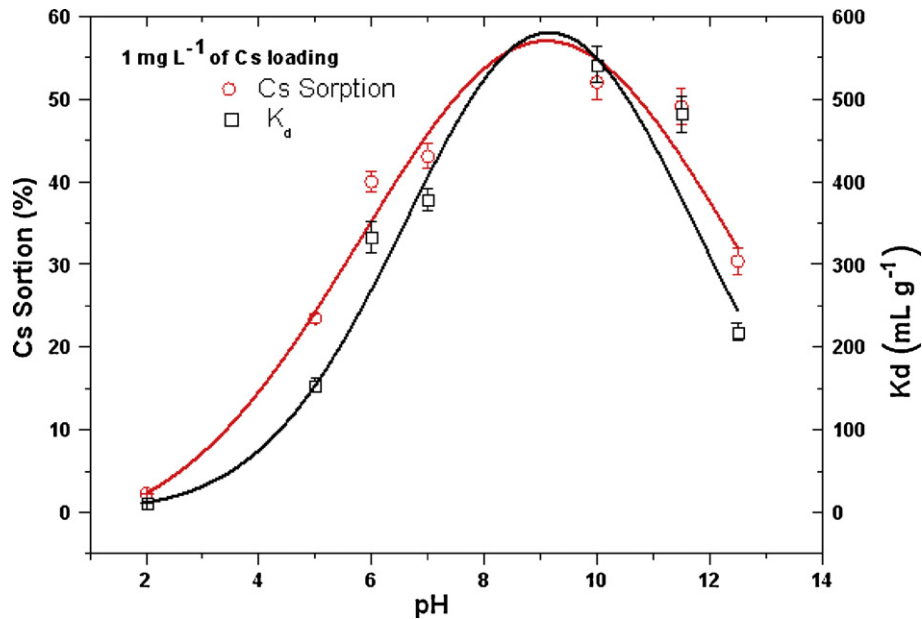


Fig. 5. Variation in  $K_d$  and Cs sorption percentage as function of pH.

their abundance in natural water. As shown in Fig. 4, it is noted that adsorption of Cs decreases significantly when ionic strength increases. Usually in ion-exchange processes, the greater is the ionic potential ( $z/r$ ) of the added cation, the greater will be the ability to depress the sorption, but in certain cases size and hydration, rather than the valance of the competing cation become the determining factors. The presence of divalent cation,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and also the monovalent cation  $\text{Na}^+$  inhibits significantly Cs adsorption. Among these cations, the presence of  $\text{K}^+$  strongly constrains the Cs adsorption, especially when potassium concentration exceeds  $10^{-3}$  M (Fig. 4). This is in agreement with other studies that Cs adsorption is the result of cation-exchange reactions, in which

cations, with similar radius and hydration energy, compete more effectively against  $\text{Cs}^+$  ions on the mineral surface [12]. So this explains the behavior of potassium, because  $\text{K}^+$  has the same characteristics that  $\text{Cs}^+$  (ionic radius of 138 pm in comparison with 167 pm of  $\text{Cs}^+$  ions, and similar hydration energy) [13]. While  $\text{Ca}^{2+}$  as well as  $\text{Na}^+$  also affects the Cs adsorption to a certain extent, this is due to their high contents in GW solutions. These results show that the ability of the cations to depress the sorption of cesium in Ain Oussera soil can follow this order:

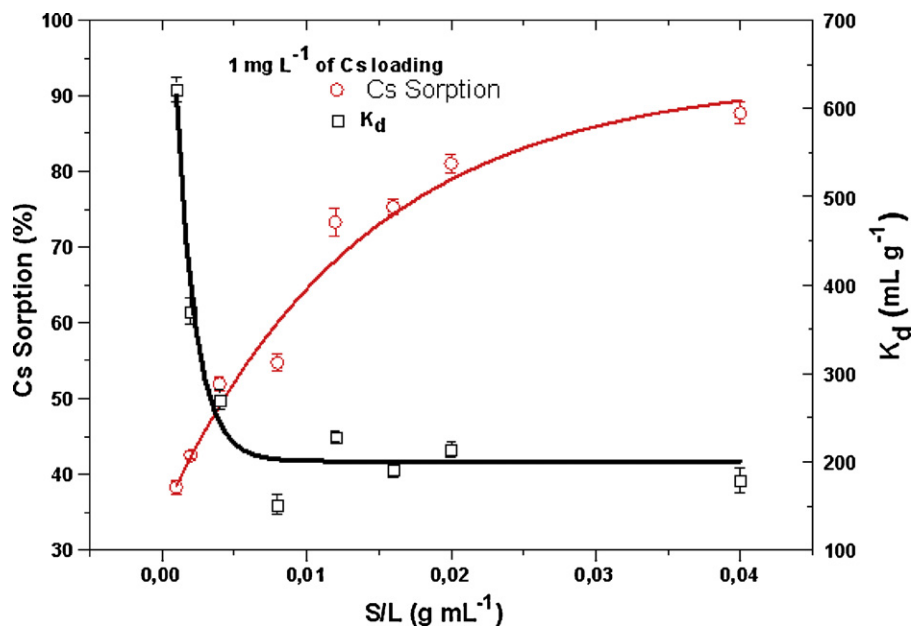
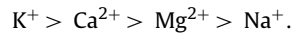


Fig. 6.  $K_d$  and Cs sorption percentage as function of S/L ratio.



### 3.5. Effect of pH

The effect of pH was studied at room temperature by varying the pH of cesium-soil solution suspension from 2 to 12.5; results are shown in Fig. 5. The sorption of cesium in Ain Oussera soil increases with increasing pH until the value of 10, then decreases clearly with increasing pH. The same behavior was also reported by Xu et al. in an evaluation study of Pd(II) adsorption on bentonite as a function of equilibrium pH [14]. At low pH, the decrease in sorption is both due to competition with hydrogen ions for exchange sites and to dissolution of a portion of soil. The greater increases in  $K_d$  and sorption percentage values are observed when the suspension pH varies from 5 to 10. The effect was more noticeable at equilibrium pH greater than 7, while it was maximum at pH 10. As analysis results indicate, Ain Oussera soil is basic; its average pH value is 8.4 so it is included within the pH range corresponding to the maximum of Cs adsorption. At pH greater than 10, the cesium hydroxides begin to form and some of them is negatively charged  $\text{Cs}(\text{OH})_2^-$  which results in the decrease of Cs adsorption due to electrostatic repulsion [14,15]. On the other hand, precipitation phenomenon cannot contribute to increase sorption due to the solubility of cesium hydroxides [16].

### 3.6. Effect of S/L ratio

To evaluate S/L ratio effect, sorption experiments were carried out with S/L ratio ranging from 0.001 to  $0.40 \text{ g mL}^{-1}$  at room temperature and Cs loading of  $1 \text{ mg L}^{-1}$ . Visibly, the sorption percentage and  $K_d$  values vary markedly with the S/L ratio.  $K_d$  decreases sharply and sorption percentage increases with increasing S/L ratio as plotted in Fig. 6. The behavior of  $K_d$  is justified by its mathematical formulation which is inversely proportional to S/L ratio (Eq. (2)), whereas the increase of the sorption percentage is due to the greater availability of exchanging sites at higher quantities of the sorbent [17].

### 3.7. Desorption

Desorption studies were carried out by carefully decanting the remaining solution after adsorption. 1 g of the loaded soil was resuspended in 50 mL ( $0.02 \text{ g mL}^{-1}$ ) of extracting reagents (DW, GW and 0.1 M KCl) separately for different time intervals at room temperature. After centrifugation and filtration the supernatant was counted for percent desorption calculations. In this series of experiments, we used DW solution to quantify sites of weakly adsorbed surfaces, whereas we adopted 0.1 M of KCl (Potassium chloride) solution to estimate amounts of high affinity ion-exchangeable sites and GW was also conducted to simulate the Cs adsorption behaviors in the real environment. From the results of particle size analysis, Ain Oussera soil contains around 10% of fine fraction mainly of clay type: montmorillonite, illite and kaolinite providing several faces with either different crystallines or imperfections (edges and corners) which will form sorption sites with different energies, resulting in different affinity sites [18]. Desorption results are shown in Fig. 7. In terms of these results, it is obvious that Cs ions were preferentially distributed onto high affinity sorption sites because about 90% of Cs was desorbed with 0.1 M KCl solution. By contrast, with DW less than 10% of Cs ions were desorbed. This means that cesium ions were probably weakly bounded and come from outer-sphere complexes, which is the consequence of electrostatic associations of hydrate Cs with anionic surfaces within the basal plane or interlayer and dissociated edge hydroxyl groups [19]. In contrast, the strongly sorbed Cs ions, which were desorbed using 0.1 M KCl solution, were generated by inner-sphere complexes, involving formation of strong electronic bonding of Cs ions to the frayed edge sites, external basal plane or within the interlayer [20].

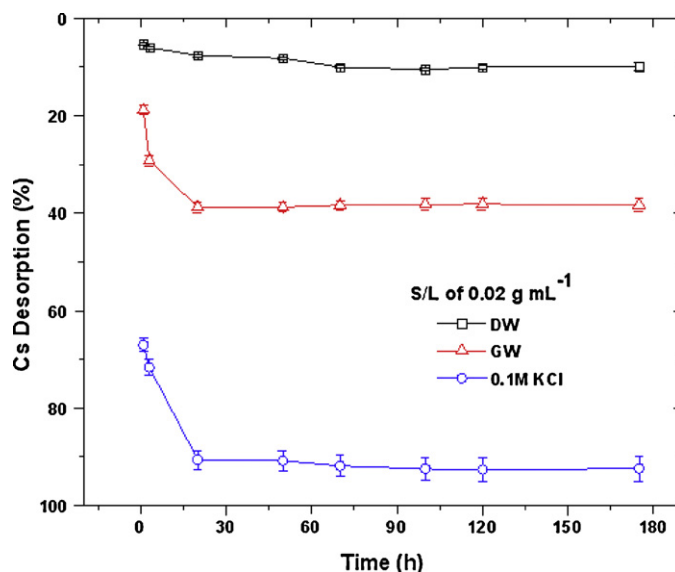


Fig. 7. Cs desorption percentage from Ain Oussera soil under 0.02 S/L ratio and at room temperature (23 °C).

In the case of groundwater, where about 40% of Cs was desorbed, it should be noted that the groundwater used in this investigation is comes directly from aquifer. The natural simulation conditions used with groundwater indicate that Cs was partially reversibly sorbed on Ain Oussera soils containing illite, kaolinite and montmorillonite clay mineral. The short-term sorption/exchange of  $\text{Cs}^+$  often exhibits reversible behavior [21,22], and has been described by multi-site, equilibrium exchange models [23,24]. Moreover, desorption investigation was conducted at fixed value of S/L ratio (0.02), according to the study of sorption as function of S/L ratio shown in Fig. 6, Cs desorption should decrease with increasing S/L ratio.

## 4. Conclusion

The adsorption kinetic was well simulated by the second-order kinetics equation, with the rate constant indicating faster sorption, with also a lower activation energy barrier toward  $\text{Cs}^+$  fixation. The Cs adsorption onto Ain Oussera was an exothermic ( $\Delta H^\circ < 0$ ), spontaneous reaction ( $\Delta G^\circ < 0$ ) and the forward reaction was favorable at low temperatures. Results of Cs adsorption under different ionic strengths indicate that the presence of potassium remarkably reduces Cs adsorption. Desorption results shows that Cs ions were preferentially distributed onto high affinity sorption sites. According to this investigation, it is reasonable to expect that much of the Cs ions released intentionally or accidentally would be strongly adsorbed by the geological substance present in Ain Oussera soil. Considering the fact that Ain Oussera region has a low rainfall, a basic soil and a low average annual temperature, the threat of groundwater contamination by radioactive nuclides is considerably reduced.

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