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STUDY OF BATCH ADSORPTION OF URANIUM IONS BY MCM-48 MATERIALS SYNTHESIZED AT ROOM TEMPERATURE

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Abstract

The mesoporous MCM-48 was synthesized at room temperature with different templates such as hexadecyltrimethylammonium bromide (C₁₆TABr), hexadecyltrimethylammonium chloride (C₁₆TACl) and dodecyltrimethylammonium chloride (C₁₂TACl). The obtained materials were characterized by XRD, N₂ adsorption/desorption, FTIR and SEM techniques, and their performance as uranium adsorbent were investigated. In this framework, the effect of contact time, pH of solution, initial uranium concentration and temperature were investigated in batch reactor.

The results showed that the adsorption process was favorably fitted with the second-order kinetic model and the isotherm obeys to Langmuir model. Moreover, the maximum adsorption capacity of MCM-48 for uranium (VI) was of the order of 160 mg/g. The thermodynamic parameters indicated that the sorption of uranium ions on MCM-48 materials was a spontaneous and endothermic process.

Keywords: adsorption, characterization, MCM-48, synthesis, uranium

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1. Introduction

In the last decade, the problem of metals has become an important subject, especially in contaminated waters. Because they are toxic and non-biodegradable, and both their accumulation and transfer pose not only a huge risk to the environment, but also to human health, even trace levels. Uranium is a toxic radioactive element arising from nuclear industry (mining, production of nuclear fuels, laboratory etc.). It is usually found in the environment in the form hexavalent (VI⁺), the mobile, aqueous uranyl ion UO₂²⁺ (Krestou et al., 2003). Its concentration in drinking water, which is recommended by the United States Environmental Protection Agency (USEPA) is 0.03 mg/L. The permissible discharge levels for nuclear industries range from 0.1 to 0.5 mg/L (Anirudhan et al., 2010). Several methods were used for the removal of uranium ions from aqueous solutions,

such as chemical precipitation (Krestou et al., 2004), solvent extraction (Shen et al., 2011), membrane filtration (Chellam and Clifford, 2002), flotation (Prasada Rao et al., 2006) and ion exchangers (Lee et al., 2002). Among these methods, adsorption is the most attractive and effective way to remove uranium from aqueous solution, due to its highly removal efficiency, low cost and simplicity of implementation (Reza et al., 2018; Zhang et al., 2015). Several works have explored the adsorption ability of a several materials such as resin, zeolitic materials, carbon, carbon nanotube composites, biomass, mesoporous materials, for removing uranium ions from aqueous solutions (Chen et al., 2018; Dan et al., 2016; Elabd et al., 2014; Mellah et al., 2006). Nevertheless, poor selectivity, low surface area and capacity restricted their use (Selvam et al., 2001).

Since 1992, a new family of ordered mesoporous silica named M41S was synthesized by Kresge et al.

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(1992) and Beck et al. (1992). These solids are prepared by hydrothermal transformation of basic silicate or aluminosilicate gels in the presence of quaternary ammonium surfactants, $C_nH_{2n+1}(CH_3)_3 N^+X^-$, with different alkyl chain lengths and counter ion ($n=12, 14, 16, 18$; $X=Br, Cl$ and OH). The M41S family includes hexagonal MCM-41, cubic MCM-48, and unstable lamellar MCM-50 mesostructures (Matei et al., 2016). The characteristic properties of these materials are of a high surface area and an ordered pore distribution, with homogenous sizes in the range of 2-10 nm. Their pore size can be tailored through the proper choice of surfactant used as template, auxiliary chemicals and reaction conditions (Koyano and Tatsumi., 1997).

The MCM-48 is the most attractive material from M41S family, owing to its three-dimensional, interconnected channels, providing more advantages including fast diffusion and resistance to pore blocking of coming molecules over the one-dimensional pores of MCM-41 (Maneesuwan et al., 2013). Moreover, due to its long-range order, large surface area, and narrow pore size distribution, MCM-48 has been used as an adsorbent, catalyst, and catalyst support.

Generally, to obtain MCM-48, the most widely used raw materials are hexadecyltrimethylammonium bromide (CTAB) as a surfactant, sodium hydroxide (NaOH) as a catalyst, and tetraethylorthosilicate (TEOS) as a silica source, using conventional autoclave heating for several days (Longloiler et al., 2011; Wang et al., 2009).

The aim of this study is to synthesize MCM-48 materials under accessible operating conditions (room temperature and short reaction time), using three directing agents, namely hexadecyltrimethylammonium bromide ($C_{16}TABr$), cetyltrimethylammonium chloride ($C_{16}TACl$) and dodecyltrimethylammonium chloride ($C_{12}TACl$), NH_4OH as catalyst and TEOS as silica source.

In an intent to use it as adsorbents to remove uranium from contaminated waters, the effects of various operational conditions, such as contact time, solution pH, initial uranium concentration and temperature were determined.

2. Experimental

2.1. Chemicals used

$C_{16}TABr$ (96%, Fluka), $C_{16}TACl$ (25% in H_2O , Aldrich), $C_{12}TACl$ (98%, Fluka) were used as the structure directing agent and TEOS (98%, Aldrich) as source of silica. Ethanol absolute C_2H_5OH (analytical grade, Merck), ammonia solution NH_4OH (29%, Prolabo) and deionized water were used as reagents for the synthesis.

Uranyl nitrate hexahydrate $UO_2(NO_3)_6 \cdot 6H_2O$ (99%, Merck) was used for the preparation of uranium stock solution at 1 g/L. The working solutions were prepared by proper dilution from the stock solution. The pH of each solution at the start experiment was adjusted to the required value with diluted nitric acid (HNO_3) and sodium hydroxide (NaOH) solutions.

2.2. Synthesis of mesoporous silica

The MCM-48 materials were prepared individually using three structure directing agents: (a) $C_{16}TABr$, (b) $C_{16}TACl$, (c) $C_{12}TACl$. In a typical synthesis 2.65 g, 9.09 g and 1.92 g of each template $C_{16}TABr$, $C_{16}TACl$, $C_{12}TACl$, respectively were added to 120 ml of deionized H_2O and 50 mL of ethanol under stirring. After complete dissolution, 12 mL of NH_4OH were added, and after 5 min 3.6 mL of TEOS. The sample is stirred vigorously during 16 hours at room temperature. The obtained solid was recovered by filtration, washed with deionized water and dried overnight at room temperature. The templates were removed by calcination at $540^\circ C$ for 6 hours.

2.3. Adsorption batch experiment

The batch uranium adsorption experiments were carried out in polypropylene flasks containing 21 mg of the different MCM-48 adsorbents (a; b; c) suspended in 15 mL of uranium solution at the selected pH. The flasks were agitated on a shaking table at 160 rpm for different contact times, initial pH of uranium solution, uranium concentration and temperatures. The solution was separated from the adsorbents by centrifugation. Then the initial and residual uranium concentrations were measured on a UV-spectrophotometer (JASCO V-530) using Arsenazo III method at a wavelength of 652 nm.

The amount of the adsorbed uranium q (mg/g) on the different MCM-48 and distribution constant (K_d) (L/g) were calculated using the following equations (Eqs. 1-2):

$$q = (C_i - C_{eq}) \cdot \frac{V}{m} \quad (1)$$

$$K_d = \frac{(C_i - C_{eq}) \cdot V}{C_{eq} \cdot m} \quad (2)$$

where C_i (mg/L) is the initial uranium concentration, C_e (mg/L) is the residual uranium concentration at equilibrium; V (L) is the volume of uranium solution; m (g) is the weight of the different MCM-48 tested as adsorbents.

2.4. Characterization of the material

X-ray powder diffraction data of the elaborated materials were collected on a diffractometer (XPRT) using $CuK\alpha$ radiation in the 2θ range of $1.7-10^\circ$ with a scan speed of $0.02^\circ/min$. N_2 adsorption-desorption isotherms were recorded using "Micromeritics ASAP2010" at 77 K. Prior to this analysis, calcined samples were degassed at $250^\circ C$ for 22h. The surface area was determined by using BET method. The pore volume, and pore diameter were calculated by using the BJH model.

FTIR-spectra were obtained on a PerkinElmer apparatus, "model Spectrum two L1600301", in 4000-

400 cm^{-1} region, using UATR technique. To determine the sample particle size and morphology, the scanning electron micrographs (SEM) of different samples were performed on a “Joel 6360LV” intensively operated.

The pH of point of zero charge (pH_{PZC}) of different MCM-48 was determined using the method described by (Cerovic et al., 2007). 20 mL aliquots of a solution of KNO_3 (0.01M) are adjusted to pH between 2-12 using NaOH (0.1N) and HNO_3 (0.1N) solutions. 0.1 g of different the adsorbents were placed in the different solutions and stirred for 24 hours. The final pHs of the different filtered solutions are measured; the pH_{PZC} corresponds to the value where the final pH and the initial pH have equal values.

3. Results and discussion

3.1. Characterization

3.1.1. X-ray diffraction

Fig. 1 shows the XRD patterns of the synthesized MCM-48 using C_{16}TABr , C_{16}TACl and C_{12}TACl as templates.

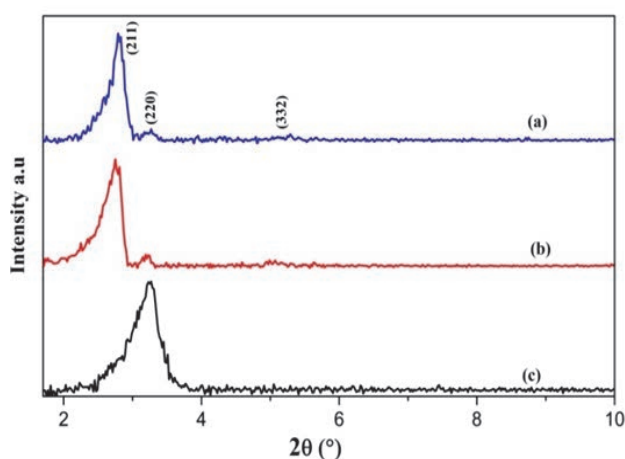


Fig. 1. XRD patterns of MCM-48 materials with different templates: (a) C_{16}TABr , (b) C_{16}TACl , (c) C_{12}TACl

The observed diffraction peaks at 2.7° , 3.2° and 5.2° corresponds respectively to (211), (220) and (332) plans according to the ASTM card N° 00-050-0511. These peaks are characteristics of Ia3d cubic structure of MCM-48 mesoporous materials (Schmidt et al., 1995). Approximately, similar diffraction patterns are observed in the case of MCM-48 prepared using C_{16}TABr and C_{16}TACl templates where (211) is the main peak in agreements with results (Melendez-Ortiz et al., 2014). However, in the case of MCM-48 prepared using C_{12}TACl template, the main peak is (220). This structure modification is probably responsible of the MCM-48 properties improvement (see results below). Duduman et al. (2018) found that the crystallinity degree increased by calcination.

3.1.2. Surface characterization

BET analysis technique encompasses external area and pore area evaluations to determine the total specific surface area yielding important information in understanding the adsorption mechanism. On the other hand, the Barrett–Joyner–Halenda (BJH) analysis allows determining pore area and specific pore volume using adsorption and desorption techniques. Both techniques are complete for the characterization of an adsorbent.

Nitrogen adsorption/desorption isotherms of elaborated materials with different templates are shown in Fig. 2. According to the IUPAC nomenclature, these isotherms can be classified as type IV, typical of mesoporous materials (Brunauer et al., 1940). Moreover, the observation of an abrupt rise at 0.2 relative pressure for the MCM-48 formed with C_{16}TABr and C_{16}TACl as template (Figs. 2 (a), (b)) due to capillary condensation is representative of mesoporous materials (Miriam et al., 2006).

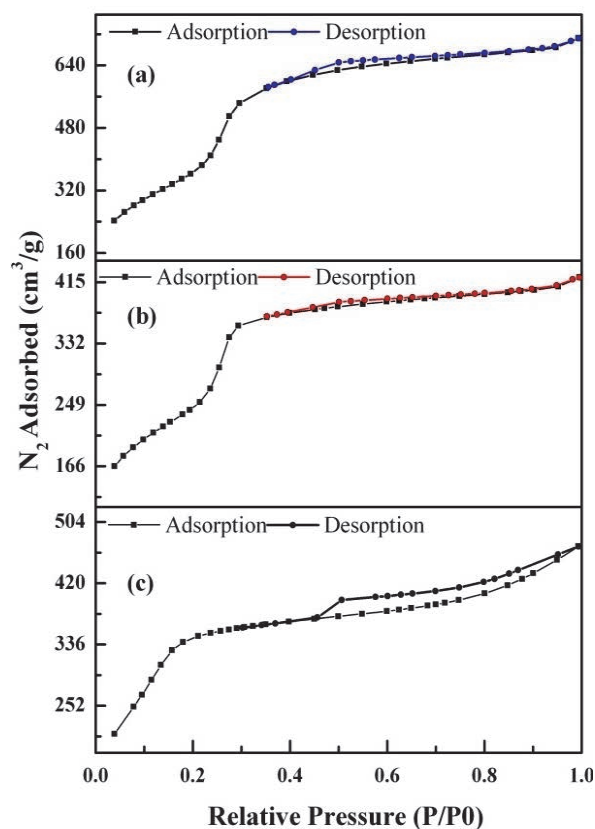


Fig. 2. Nitrogen adsorption-desorption isotherms of MCM-48 materials with different templates: (a) C_{16}TABr , (b) C_{16}TACl , (c) C_{12}TACl

All isotherms present H4 type hysteresis, assigned to slit shaped pores (Kumar et al., 2003). The isotherms are also characterized by a small hysteresis loop in the range of p/p_0 of 0.4-0.8, which is ascribed to the inter-particle volume. Table 1 presents the textural characteristics for the MCM-48 materials.

Table 1. Textural characteristics for the MCM-48

Samples	S_{BET} (m^2/g)	Pore volume (cm^3/g)	Pore diameter (nm)
MCM-48(a)	1334	1.05	2.57-4.58
MCM-48(b)	900	0.38	2.33-5.99
MCM-48(c)	1418	0.54	2.77-5.91

Table 2. Comparison of the physical characteristics of MCM-48 (a) with similar materials

Materials		MCM-48 Chen et al. (2013)	MCM-48 Longloilert et al. (2011)	MCM48 (a) Present work
Synthesis conditions	Temperature($^{\circ}C$)	110	140	25
	Time (h)	72	16	16
Characteristics	S_{BET} (m^2/g)	961	1288	1334
	Pore diameter (nm)	4.16	2.86	2.57-4.58
	Pore volume (cm^3/g)	1.00	0.92	1.05

This table shows high specific surface area values which are obtained for all MCM-48 materials (higher than $900m^2/g$). Their pore diameters are calculated by the BJH model that vary from 2.3 nm to 6.0 nm. Consequently, MCM-48 materials possess intrinsic characteristics that can lead to interesting adsorptive capacities.

The comparison of MCM-48 (a) with similar synthesized materials is summarized in Table 2. It is found that the characteristics are comparable, although the synthesis conditions are different.

3.1.3. FTIR results

As shown in Fig. 3, similar FTIR spectra were obtained for the different materials. The adsorption broad band in $3000-3700\text{ cm}^{-1}$ region, with maximum at around 3376 cm^{-1} , is due to the O-H stretch of silanol group's (Tavakoli et al., 2013).

The band in region $1050-1080\text{ cm}^{-1}$ was attributed to the asymmetric stretching of Si-O-Si groups. The peak at around 960 cm^{-1} was assigned to the Si-O stretching vibration of Si-OH groups. The symmetric stretching of Si-O-Si groups was observed at $\sim 800\text{ cm}^{-1}$ and the peak at 460 cm^{-1} is due to the bending mode of Si-O-Si, which are the characteristic adsorption peaks of SiO_2 . This indicates that the structure of the materials has not been damaged during the calcination step (Zuo et al., 2011).

3.1.4. SEM analysis

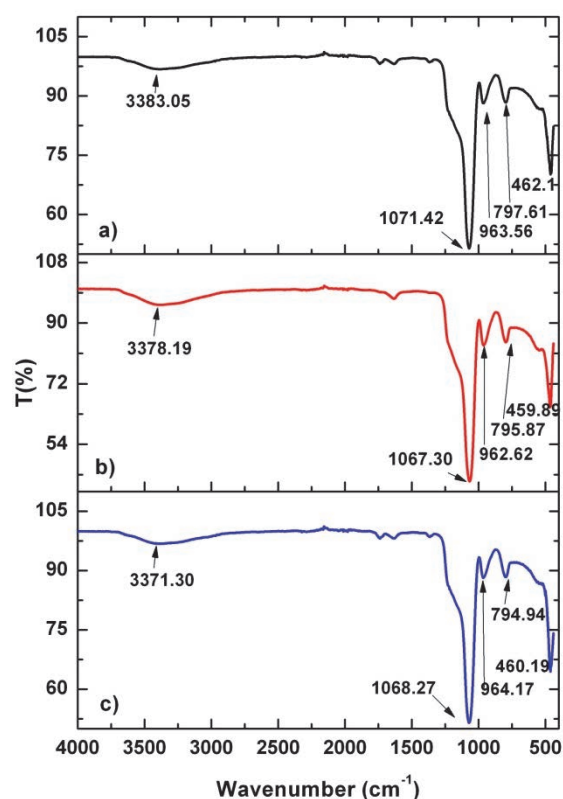
Fig. 4 shows that the different templates used did not induce any notable morphological changes. All samples showed a spherical morphology with sizes from 200 to 600 nm for $C_{16}TABr$ 100 to 500 nm for $C_{16}TACl$ and 200 to 700 nm for $C_{12}TACl$.

3.2. Sorption studies

3.2.1. Effect of contact time

The effect of contact time on the adsorption of U(VI) ions onto different MCM-48 was investigated for contact time varying from 0 to 10 hours. The experiments were performed at room temperature with an initial concentration of U(VI) of 50 mg/L. The experimental results are presented in Fig. 5. The uptake

was found to be fast and increases with an increase in contact time.

**Fig. 3.** FTIR spectra of the MCM-48 materials with different templates:(a) $C_{16}TABr$, (b) $C_{16}TACl$, (c) $C_{12}TACl$

At the equilibrium point, the desorbed amount of U(VI) was in equilibrium with the amount being adsorbed (overall adsorption $< 20\text{ mg/g}$). The same shape of the graphs and the same equilibrium time, suggest that the adsorption process is the same for the tested materials. The elaboration of the materials gives compounds of the same structure but with different characteristics due to the introduction of the different elements. Therefore, a contact time of 5 hours was selected as the optimum parameter for the following experiments.

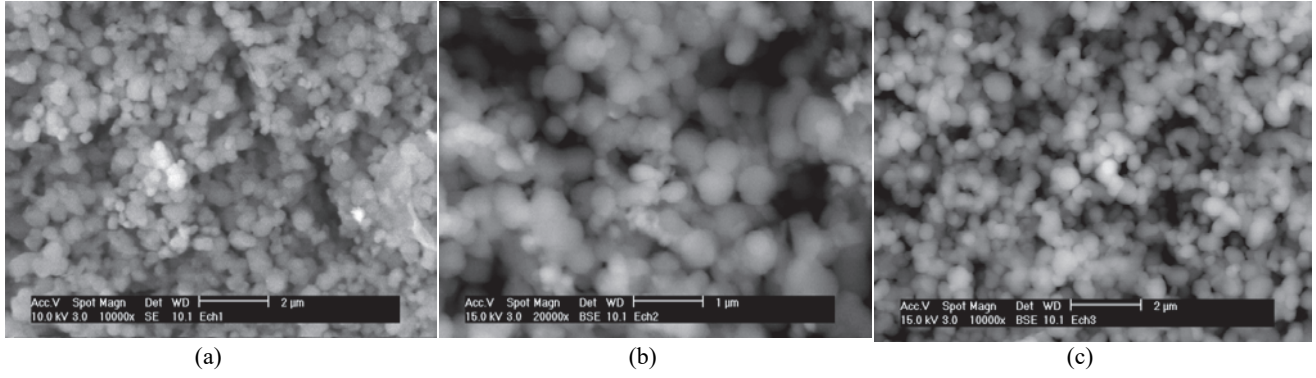


Fig. 4. Scanning electron micrographs of the MCM-48 materials with different templates: (a) C₁₆TABr, (b) C₁₆TACl, (c) C₁₂TACl

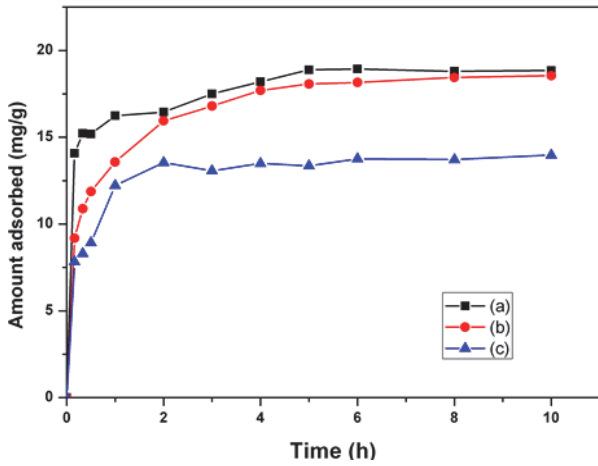


Fig. 5. Effect of contact time on the adsorption of uranium onto the MCM-48 materials with different templates: (a) C₁₆TABr, (b) C₁₆TACl, (c) C₁₂TACl ([U] = 50 mg/L, pH=4, m=21mg, V= 15 mL, T=25°C)

3.2.2. Adsorption mechanism and kinetics

Adsorption is a complex multistep process and kinetic studies will provide valuable insights of the sorption mechanisms which involve mass transfer, diffusion and surface reaction phenomenon. However, the kinetics helps for the prediction of adsorption rate, and the mechanism of the phenomenon which give important information for the modelling the process.

The data of the kinetics of adsorption were analysed using two different kinetic models: the pseudo-first-order and pseudo-second-order models.

- Pseudo-first-order model (Lagergreen, 1889) which has been widely used to predict adsorption kinetics and is given by the following equation (Eq. 3):

$$q = q_e(1 - e^{-K_1 t}) \quad (3)$$

Eq. (3) may be rearranged for linearized data plotting as shown by Eq. (4):

$$\ln(q_e - q_t) = \ln q_e - k_1 t, \text{ or}$$

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (4)$$

- Pseudo-second-order kinetic model is expressed by Eq. (5) (Plazinski et al., 2013):

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \quad (5)$$

which can be rewritten as Eq. (6):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (6)$$

where q_t (mg/g) is the amount of uranium adsorbed on different MCM-48 materials at time t , and q_e (mg/g) is the equilibrium amount adsorbed. The constant k_1 (h⁻¹) and k_2 (g/mg/h) are respectively the adsorption rate constant of pseudo-first and second order kinetic.

The determined parameters from the exploitation of the experimental data are presented in Table 3.

The results show that pseudo-second-order model fits the experimental data quite well, the R² values reach the unity and the experimental and theoretical uptakes are in good agreement. This indicates the applicability of the second-order kinetic model to describe the adsorption of uranium (VI) ions by MCM-48 materials. The rate constant for MCM-48 (a) is about 0.28 g/mg/h, as well as the amount adsorbed (≈ 20 mg/g). This was predictable in view of the intrinsic characteristics of the material.

Table 3. Kinetic model parameters

Samples	Pseudo –first order			Pseudo second order		
	K ₁ (1/h)	q _e (m/g)	R ²	k ₂ (g/mg/h)	q _e (mg/g)	R ²
MCM-48 (a)	0.28	4.16	0.887	0.28	19.18	0.998
MCM-48 (b)	0.32	9.15	0.894	0.05	19.55	0.996
MCM-48 (c)	0.41	3.74	0.731	0.35	14.16	0.999

3.2.3. Effect of pH

The effect of initial pH of the solution on the maximum uptake capacity of uranium on the different MCM-48 materials was studied using a pH range from 2.0 to 8.0 at 25°C for 5 hours (Fig. 6). It can be seen that the adsorption is strongly dependent on pH value. The amount of uranium adsorbed increases from 0 to 35 mg/g, as pH increases from 2 to 6. The pH of the solution influences the charge of the adsorbent with respect of the point zero charge (Li et al., 2014). This behaviour can be attributed to the synergic effect of functional groups in different MCM-48 materials.

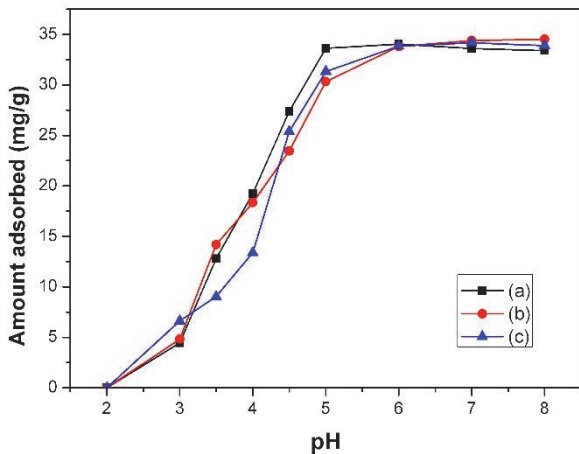


Fig. 6. Effect of pH on the adsorption of uranium onto the MCM-48 materials with different templates: (a) C₁₆TABr, (b) C₁₆TACL, (c) C₁₂TACL ([U] = 50 mg/L, t=5 h, m=21 mg, V= 15 mL, T=25°C)

Otherwise, the pH of point zero charge (pH_{PZC}) for different MCM-48 materials which are found to be approximately 4.6, 4.2 and 3.5 respectively. It is expected that the amount of negative binding sites at different MCM-48 materials can rapidly be increased after pH_{PZC}. On the other hand, the species of uranium are influenced by the pH of the solution; different mononuclear and polynuclear U (VI) hydrolysis products that could be present in aqueous uranium solution as a function of pH (Chen et al., 2013). At pH between 2.0 and 3.0, only UO₂²⁺ is present, from pH 3.0 to 5.0, the polynuclear products [UO₂(OH)]⁺, [(UO₂)₂(OH)₂]²⁺ and [(UO₂)₃(OH)₅]⁺ are present with UO₂²⁺. At pH between 6.0 and 7.0, the hydrolysis is more intense and other polynuclear product [(UO₂)₄(OH)₇]⁺ and [(UO₂)₃(OH)₅]⁺ are formed.

As a result, all these species can be adsorbed by the functional groups of different MCM-48 materials. Moreover, at pH higher than 7.0, carbonate uranyl ions UO₂(CO₃)₂²⁻ and UO₂(CO₃)₃⁴⁻ are formed (Bayou and al., 2017). The next experiments in this study were carried at pH 6.

3.2.4. Adsorption isotherms

Adsorption isotherm provides essential physiochemical data for assessing the applicability of the adsorption process as a complete unit operation. The

adsorption of uranium on MCM-48 was carried out under different initial concentrations from 10 to 300 mg/L at pH 6 for 5 hours.

Fig. 7 shows the amount of uranium adsorbed as function of initial uranium concentration.

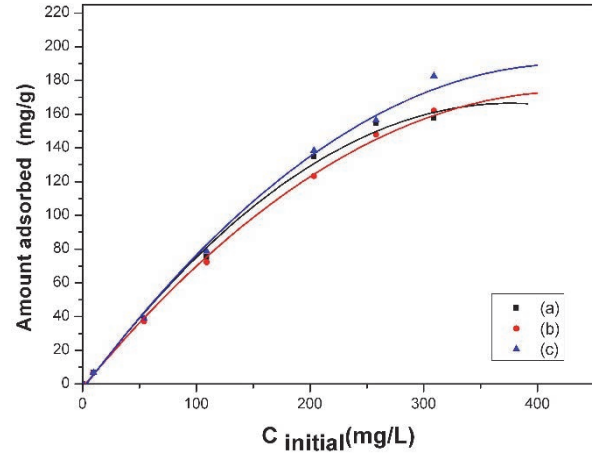


Fig. 7. Effect of initial uranium concentration on the adsorption of U(IV) onto the MCM-48 with different templates: (a) C₁₆TABr, (b) C₁₆TACL, (c) C₁₂TACL (pH = 6, t=5 h, m=21 mg, V= 15 mL, T=25°C)

Langmuir and Freundlich adsorption isotherms models were used to correlate the experimental data. According to the Langmuir model, the saturated monolayer isotherm (Sari et al., 2007) can be Eq. (7):

$$q_e = \frac{q_{max} k_L C_e}{1 + k_L C_e} \quad (7)$$

where q_e (mg/g) is the amount of U(VI) adsorbed per unit mass of adsorbent; C_e (mg/L) is the equilibrium concentration; q_{max} (mg/g) and k_L (L/mg) are Langmuir constants related to adsorption capacity and equilibrium constant related to the affinity of the binding sites and energy of adsorption respectively.

The Freundlich isotherm model assumes that adsorption occurs on a heterogeneous surface through a multilayer adsorption mechanism and that the adsorbed amount is represented by Eq. (8) (Oguz, 2007):

$$q_e = k_f C_e^{1/n} \quad (8)$$

where: k_f ((mg⁽¹⁻ⁿ⁾Lⁿ)/g) is defined as the adsorption capacity of the adsorbent. The $1/n$ (dimensionless factor) measures the adsorption intensity or surface heterogeneity.

All constants were calculated according to the slope and the intercept of the related lines. The determined parameters from the exploitation of the experimental data are tabulated in Table 4.

The high values of the correlation coefficients ($R^2 > 0.98$) of Langmuir plot isotherm shows an excellent fit to the experimental data. As noticed, the adsorbed

amounts are consistent ($q_{max} > 160$ mg/g) Moreover, the material MCM48 (a) present an increased affinity ($K_L = 0.93$ L/g) for the adsorbate.

In comparison to other kinds of adsorbents used under similar experimental conditions (Table 5), the MCM-48 (a) material presents a higher adsorption capacity.

3.2.5. Adsorption thermodynamics

The influence of temperature variation on the adsorption of uranium ions onto MCM-48 was investigated from 25 to 55°C (Fig. 8). From this figure, we can notice that the amount of uranium adsorbed decreases as temperature was increases. This phenomenon, in agreement with the Arrhenius law, suggests that the surface reaction is exothermic and that each temperature increase disadvantages its progress.

Thermodynamic parameters such as standard enthalpy ΔH° (KJ/mol) and standard entropy ΔS° (KJ/mol·K), were determined by using Eq. (9) (Kumar and Kacha, 2011):

$$\ln K_d = \left(\Delta S^\circ / R \right) - \left(\Delta H^\circ / RT \right) \quad (9)$$

where K_d is the distribution coefficient.

The values of enthalpy (ΔH°) and entropy (ΔS°) were obtained by the calculated linear fitting curve of $\ln K_d$ as function $1/T$. The standard free energy change was calculated as Eq. (10):

$$\Delta G^0 = -RT \ln K_d \quad (10)$$

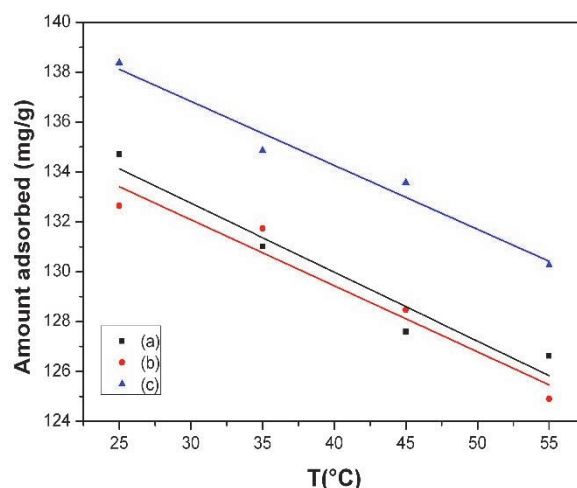


Fig. 8. Effect of temperature on the adsorption of uranium onto MCM-48 with different templates: (a) C₁₆TABr, (b) C₁₆TACl, (c) C₁₂TACl ([U] = 200 mg/L, pH = 6.0, t=5 h, m=21 mg, V= 15 mL)

The values of the thermodynamic parameters for the adsorption of uranium ions on the different MCM-48 are summarized in Table 6.

The observed positive values for the enthalpy suggest that the adsorption of uranium ions is endothermic. Moreover, the negative values of ΔS° revealed that the randomness decreased at the different MCM-48 solution interface during the adsorption process. The negative values of ΔG^0 observed for all MCM-48 materials indicate that the adsorption process of uranium is spontaneous and the degree of spontaneity decreases with increasing temperature.

Table 4. Parameters of the isotherm models for uranium adsorption on the MCM-48 materials

Samples	Langmuir model			Freundlich model		
	q_{max} (mg/g)	k_L (L/g)	R^2	k_F ((mg ¹⁻ⁿ L ⁿ)/g)	n	R^2
MCM-48 (a)	161.29	0.93	0.999	64.31	4.34	0.954
MCM-48 (b)	169.49	0.21	0.978	40.55	3.03	0.974
MCM-48 (c)	185.19	0.49	0.991	65.31	3.71	0.946

Table 5. Comparison of the uranium adsorption capacity of MCM-48 (a) with other adsorbents

Adsorbents	Experimental conditions	q_{max} (mg/g)	Reference
Graphene oxide nanosheets	pH=5, T=20°C	97.5	Zhao et al.(2012)
Carbone nanotubes	pH=6, T= 25°C	137.80	Chen et al.(2018)
Humic acid immobilized Zirconium pillared clay	pH=6, T=30°C	132.68	Anirudhan et al.(2010)
Natural clinoptilolite	pH =6, T=25°C	2.88	Camacho et al. (2010)
MCM-48 (a)	pH=6, T=25°C	161.29	This work

Table 6. Thermodynamic parameters

Samples	ΔH° (KJ/mol)	ΔS° (J/mol·K)	ΔG° (KJ/mol)			
			298K	308K	318K	328K
MCM-48 (a)	28.87	-56.40	-6.12	-5.43	-4.82	-4.47
MCM-48 (b)	21.43	-52.47	-5.68	-5.32	-5.00	-4.01
MCM-48 (c)	24.20	-57.99	-7.93	-5.67	-5.52	-5.58

4. Conclusions

From the various results obtained in this study, we conclude:

- The mesoporous materials MCM-48 elaborated at room temperature with different templates (C₁₆TABr, C₁₆TACl and C₁₂TACl) as the directing agent of the structure and TEOS as silica source, perfectly fulfilled the role for which they were synthesized. As adsorbent materials, it has a high uranium adsorption capacity greater than 160 mg/g; with an appreciable affinity for the uranium ions of the order of 1 g/L for MCM-48 (a).

- The kinetic studies indicate that the adsorption of uranium ions on MCM-48 can be described by a pseudo-second-order model.

- The adsorption of uranium onto MCM-48 materials was a strongly pH dependent.

- The adsorption equilibrium was fitted by Langmuir equation.

- The value of thermodynamic parameters confirmed the feasibility and spontaneity, as well the endothermic nature of the adsorption process of the uranium ions onto MCM-48 mesoporous materials.

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