



Synthesis of Ethylene diamine-modified Ordered Mesoporous Carbon as a New Nanoporous Adsorbent for Removal of Cu(II) and Pb(II) Ions from Aqueous Media

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The mesoporous carbon (CMK-3) functionalized with ethylenediamine (EDA) has been prepared (CMK-3-EDA) and used as a new mesoporous adsorbent for removal of Cu(II) and Pb(II) cations from aqueous solutions. Nitrogen adsorption-desorption measurements (BET) show that surface area, pore size and pore volume of CMK-3 have significantly changed after amine modification. The BET surface area and pore diameter of functionalized product were $344.74 \text{ m}^2 \cdot \text{g}^{-1}$ and 28.61 \AA , respectively. The adsorption conditions including contact time, pH value and adsorbent dosage of the sample solution have been studied in batch system and then determined by means of flame atomic absorption spectrometry. The adsorption capacity was $188.2 \text{ mg} \cdot \text{g}^{-1}$ and $196.64 \text{ mg} \cdot \text{g}^{-1}$ for Cu(II) and Pb(II) ions, respectively. The obtained high adsorption capacity of CMK-3 functionalized with EDA is due to the amine functional groups formed on the surface of CMK-3 which can react with Pb(II) and Cu(II) ions. Results show that the new synthesized porous material is a highly effective for sorption of Pb(II) and Cu(II) ions in comparison to other adsorbents.

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1. INTRODUCTION

Environmental pollution due to rapid technological development is a matter of serious concern [1]. The presence of high amount of heavy metals such as cadmium, nickel, lead, mercury, chromium, and copper in the environment may cause serious problems for the human health [2]. Lead present in air, soil, water, and foods causes neurological damage, heart and renal disease as well as brain and blood disorders [3]. Copper is a hazardous metal due to its non-biodegradability and its accumulation in the environment [4]. Different methods including chemical precipitation, adsorption, ion-exchange, membrane filtration and electrochemical processes have been applied for removing heavy metals from water media. Out of these mentioned methods, adsorption has been found to be one of the best techniques for this purpose [5]. Adsorption of heavy metals on mesoporous materials is a relatively recent

development. In the early 1990's, new ordered mesoporous materials were synthesized by researchers at Mobil Oil Corporation [6]. The hexagonally ordered mesoporous silica materials have many attractive characteristics such as well-ordered periodic pore structure, high thermal and chemical stabilities, controllable pore diameter, large surface area and their ability to incorporate specific binding materials [3, 7]. In 1998, Zhao et al. developed SBA-15 mesostructured silica. SBA-15 usually has wider pores than MCM-41 (SBA-15 pores range from 4.6 to 30 nm) and higher pore volumes [8]. For the first time, mesoporous carbon was synthesized in 1999 by Ryoo using MCM-48 as a hard template [9]. Mesoporous carbon, due to its high specific surface area, relatively large diameter and pore volume has been selected for removing the heavy metal ions from the aqueous solution [10]. Porous materials have also been applied in water purification [11-13], gas separation, catalyst support and electrode materials for electrochemical double layer capacitors and fuel cells [14].

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In the present work, SBA-15 mesoporous silica has been synthesized by a hydrothermal method and then used as template-silica along with sucrose as the carbon source for preparation of the CMK-3. Consequently, CMK-3 was carboxylated by ammonium persulfate ((NH₄)₂S₂O₈) and H₂SO₄, then functionalized with EDA. The resulting material has been used as a new adsorbent for removal of the Pb(II) and Cu(II) ions.

2. EXPERIMENTAL

2. 1. Reagents and Materials Tetraethylorthosilicate (TEOS), sulfuric acid (98%), phosphoric acid (85%), sucrose, sodium hydroxide, ethanol, ammonium persulfate ((NH₄)₂S₂O₈) ≥98%, thionyl chloride(SOCl₂) 97%, EDA, Copper(II) nitrate trihydrate, Lead(II) nitrate and hydrofluoric acid 38-40% (HF) were from E. Merck (Germany). P123 [poly(ethylene glycol)-block-poly(propylene glycol)-block-poly(ethylene glycol)] was from Aldrich.

2. 2. Synthesis of SBA-15 Mesoporous silica was synthesized by hydrothermal method reported earlier by [15]. The detailed procedure was as follows: first, 4g P123 was dissolved in 150 ml H₃PO₄ at temperature 40°C. The resulting solution was poured in a Teflon autoclave and then 9.6 ml of TEOS was added in it drop wise while stirring the solution. Stirring of the resulting solution was continued for 24h at 40°C. After this, the solution was aged at 100°C for 24 h without stirring. The produced solid product was obtained by filtration, then washed and dried at 100°C for 20 h. In order to remove the P123 template, the dried product was calcined under flow of air at 250°C for 3 h and at 550°C for 4 h.

2. 3. Synthesis of CMK-3 CMK-3 was prepared according to the process described earlier [16] using SBA-15 as template silica and sucrose as the carbon source. The detailed procedure was as follows: 1.25 g of sucrose and 0.14 g of H₂SO₄ in 5.0g of H₂O was dissolved and the obtained solution was added to 1g of SBA-15. The resultant mixture was dried in an oven at 100°C for 6h and continued by increasing the oven temperature up to 160 °C for 6h. In order to complete the polymerization and carbonization processes, 0.75 g sucrose, 0.08 g H₂SO₄ and 5.0 g H₂O was used and the resulted mixture was dried again at 100 °C for 6h followed by 160 °C for 6h. The resultant product was carbonized in an argon gas flow at 900 for 6 h with a heating rate of 2.5 °C.min⁻¹. Then removal of template silica was achieved twice using 1M NaOH solution (50% ethanol–50% H₂O) at 100°C. To ensure the complete removal of the silica, 5% wt hydrofluoric acid solution was used and then the mixture was filtered and

washed several times using ethanol and consequently dried at 120 °C for 4 h.

2. 4. Chemical Oxidation of CMK-3 CMK-3 was carboxylated according to the literature [9]. In This procedure, 300 mg CMK-3 was immersed into 18 ml solution containing 1.75 molar (NH₄)₂S₂O₈ and 2 molar H₂SO₄ and then refluxed at 40C° for 24 h. Finally, the oxidized CMK-3 was filtered and washed using distilled water until the pH reaches close to 7 and then dried at 108°C.

2. 5. Synthesis of CMK-3-EDA The CMK-3-COOH was dispersed in 15 ml thionyl chloride and then the mixture was heated at 70°C for 24 h to obtain CMK-3-COCl. The resultant mixture was filtered and dried under vacuum condition at ambient temperature for 10h. In the next stage, CMK-3-COCl was redispersed into 50 ml of EDA at 50°C for 48 h. After cooling up to ambient temperature, the mixture was filtered and then washed by ethanol to remove the excess EDA. Finally, CMK-3-EDA was obtained after drying the product under vacuum condition (Figure 1).

2. 6. Characterization FT-IR spectra of the adsorbent samples were recorded at ambient temperature on a DIGILAB FTS 7000 spectrometer equipped with an attenuated total reflection (ATR) cell. N₂ adsorption–desorption isotherms were measured at 77 °K by means of a Micromeritics ASAP 2010 instrument. SEM (PHILIPS XL30) was used to analyze product morphology. The concentrations of metal ions of the solution were measured by atomic absorption spectrometry (AAS, Philips, PU9400).

2. 7. Adsorption studies Analytical grade nitrate salts of Cu(II) and Pb(II) were used to prepare the stock solutions. In this regard, batch mode adsorption was selected because of its simplicity in contrast to other modes.

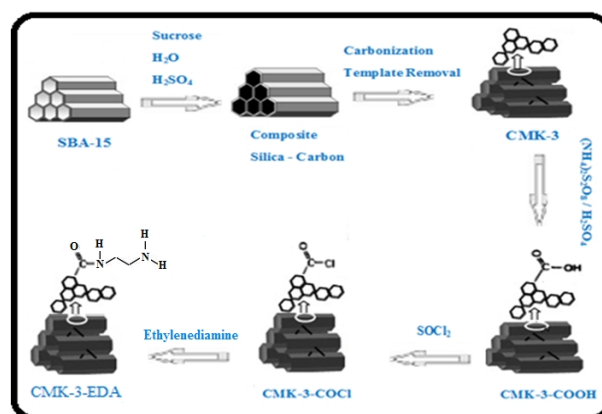


Figure 1. Schematic illustration of synthesis of CMK-3-EDA

Batch adsorption experiment was performed using 100 mg.L^{-1} Cu(II) and Pb(II) ions solution while stirring the mixture at 150 rpm at ambient temperature. After separating the adsorbent by filtration, the metal ions concentrations in the solution were determined by flame atomic absorption spectrometry. The adsorption capacity of the adsorbent at equilibrium conditions was also calculated by the following equation:

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

where, C_0 and C_e are the initial and equilibrium heavy metal ions concentrations in the solution in mg.L^{-1} , respectively, V is the volume of the solution in liters and W is the weight of the adsorbent in grams. The removal efficiency was calculated using the following equation:

$$R = \frac{(C_0 - C_e)V}{C_0} \times 100 \quad (2)$$

where, C_0 and C_e are the initial and equilibrium heavy metal concentrations in the solution in mg.L^{-1} , respectively.

3. RESULTS AND DISCUSSION

3. 1. Characterization of the Adsorbent

The FT-IR spectra of CMK-3 and CMK-3-EDA are recorded in Figure 2. FT-IR spectrum of the CMK-3 sample shows few broad absorption bands which are related to stretching vibration of the C–O–C and C=C aromatic group at 1097 cm^{-1} and 1597 cm^{-1} respectively (Figure 2a). Figure 1b shows the IR spectrum of the CMK-3-EDA, where the new bands at 1650 , 1125 and 1580 cm^{-1} are assigned to the C=O, C–N and N–H bonds, respectively. Figure 2b depicts that the CMK-3 was successfully modified by the EDA. The N_2 adsorption/desorption isotherms and pore size distributions of CMK-3 and CMK-3-EDA at 77 °K are shown in Figure 3. The BET surface area and pore volume data, presented in Table 1, were obtained from these isotherms. According to the IUPAC classification, both isotherms are of type IV. This implies that the ordered mesostructure of CMK-3 has not been destroyed after the reaction between CMK-3 and EDA. The reaction between CMK-3 and EDA shows a distinct decrease in BET surface area (from 851 to $344.7 \text{ m}^2 \cdot \text{g}^{-1}$) and pore volume (from 0.858 to $0.274 \text{ cm}^3 \cdot \text{g}^{-1}$), which indicates that the EDA group has been grafted on CMK-3.

SEM instrument was employed to characterize the morphology of the CMK-3-EDA. SEM images of CMK-3 and CMK-3-EDA samples are shown in Figure 4a and b. It is clear from Figure 4 that, both CMK-3 and CMK-3-EDA have a rod-like shape. This suggests that the shape of the structure is not changed after modification of the CMK-3 surface by EDA.

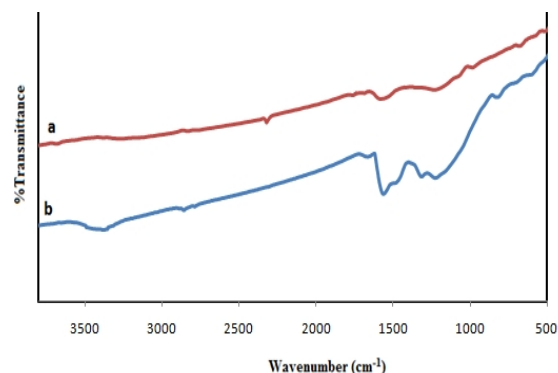


Figure 2. FT-IR spectra of CMK-3(a) and CMK-3-EDA (b).

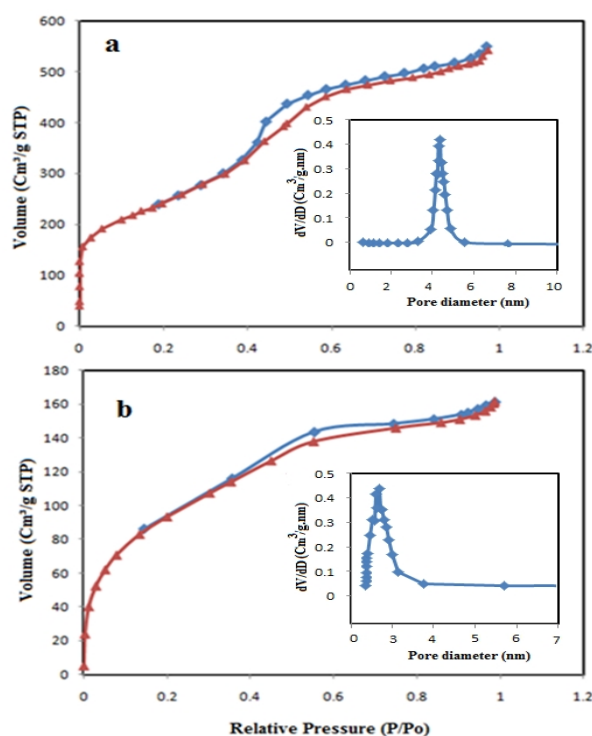
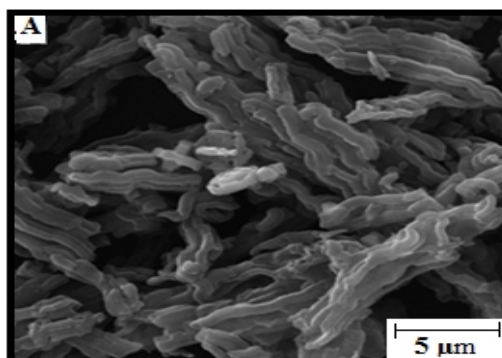


Figure 3. The N_2 adsorption-desorption isotherms and pore size distributions of CMK-3 (a) and CMK-3-EDA (b) powders at 77 K .



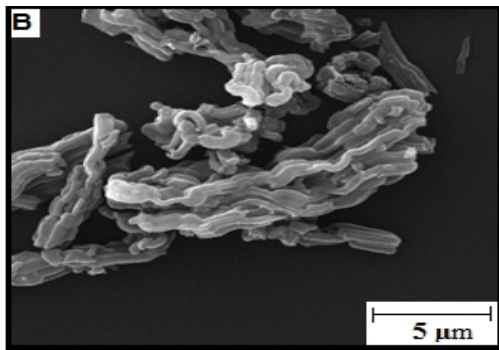


Figure 4. SEM images of CMK-3 (A) and CMK-3-EDA (B).

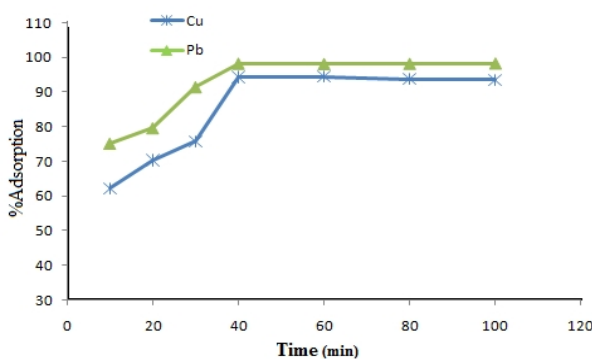


Figure 5. The effect of contact time on adsorption of the Pb(II) and Cu(II) ions was studied under the following conditions: adsorbent dosage 0.5 g.L^{-1} , initial concentration of metal ions 100 mg.L^{-1} , speed 150 rpm , pH (5.0 for Pb(II) and 6.0 for Cu(II)) and at ambient temperature.

TABLE 1. Textural properties of CMK-3 and CMK-3-EDA

Sample	$S_{\text{BET}}(\text{m}^2 \text{g}^{-1})$	$d(\text{nm})$	$V(\text{cm}^3 \text{g}^{-1})$
CMK-3	851	4.30	0.858
CMK-3-EDA	344.7	2.86	0.274

3. 2. Adsorption Studies

3. 2. 1. Effect of Contact Time The equilibrium time between metal ion and adsorbent is of significant importance in adsorption process. The effect of contact time on adsorption of the metal ions was studied under the following conditions: adsorbent dosage of 0.5 g.L^{-1} , initial concentration of metal ions 100 mg.L^{-1} , mixing speed of 150 rpm at ambient temperature. The results given in Figure 5 show that the adsorption percent of the metal ions was increases by increasing the mixing time until it reaches to equilibration time of 40 min (Figure 5). Thus, for all adsorption studies, the equilibration time was kept 60 min.

3. 2. 2. Effect of Adsorbent Dosage Adsorbent dosage is an important parameter in the determination of adsorption capacity and removal percent. The effect of

the CMK-3-EDA dosage on the removal of Pb(II) and Cu(II) ions at 100 mg.L^{-1} solution, at mixing speed of 150 rpm , ambient temperature and contact time of 60 min is depicted in Figure 6. As can be seen, increasing the adsorbent amount up to 0.5 g.L^{-1} leads to an increase in the removal percent of Pb(II) and Cu(II). This increase in removal percent is due to the availability of more active sites of the adsorbent. The decrease in adsorption capacity with increase in adsorbent dose is due to the fact that some of the adsorption sites remain unsaturated during the adsorption process.

3. 2. 3. Effect of pH Value The pH is one of the important parameters affecting the adsorption process. The solution pH may affect the surface charge of the adsorbents through protonation of the functional groups or hydrolysis of the species present in the solution.

The optimum pH for removal of the metal ions was measured at ambient temperature, agitation time of 60 min, mixing speed of 150 rpm , different pH values in the range of 3-6. Figure 7 shows that the maximum removal of Pb(II) occurs at a pH of about 5.0, while that of Cu(II) at pH of about 6.0. Also, Cu(II) and Pb(II) adsorption capacities increased with an increase in initial pH and reached to their maximum values at the initial pH of about 5.0 for Pb(II) and 6.0 for Cu(II).

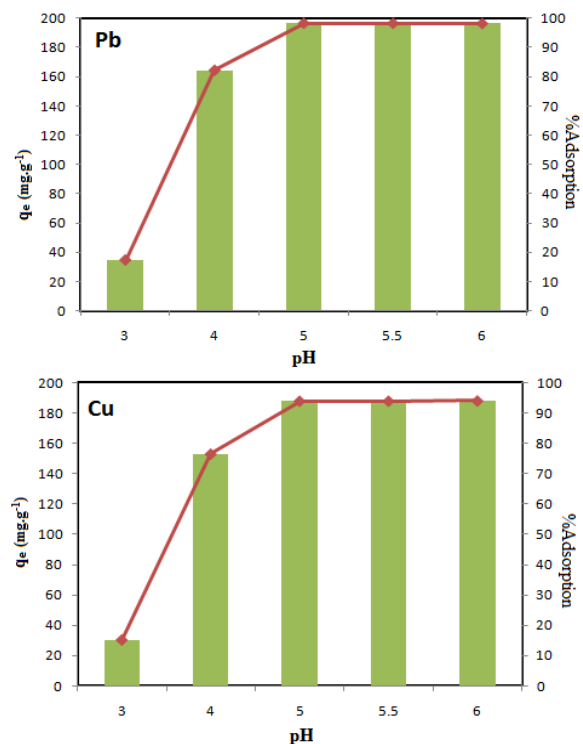


Figure 7. The effect of pH on adsorption of the Pb(II) and Cu(II) ions was studied under the following conditions: adsorbent dosage 0.5 g.L^{-1} , initial concentration of metal ions 100 mg.L^{-1} , agitation time 60, speed 150 rpm and at ambient temperature.

The increase in the adsorption capacity could be well explained by protonation properties of the adsorbent. At the low pH and high hydrogen ion concentration, the surface charge of the adsorbent is changed by protonation of the functional groups.

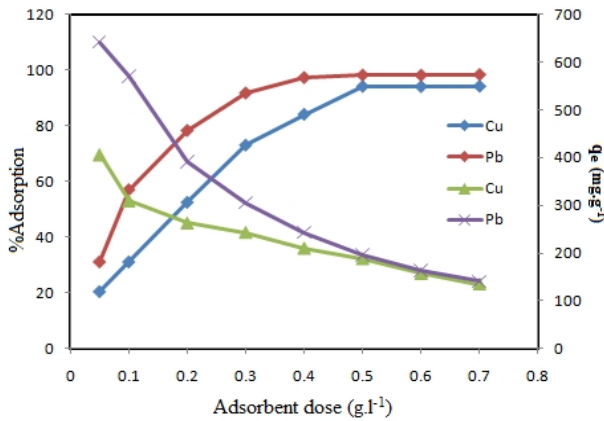


Figure 6. The effect of adsorbent dosage on adsorption of the Pb(II) and Cu(II) ions was studied under the following conditions: initial concentration of metal ions 100 mg.L⁻¹, agitation time 60 min, speed 150 rpm, pH (5.0 for Pb(II) and 6.0 for Cu(II)) and at ambient temperature.

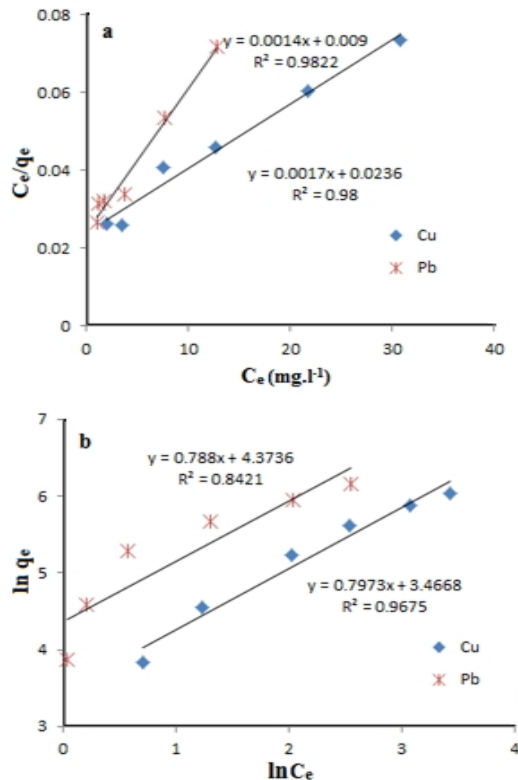


Figure 8. Langmuir (a), Freundlich (b) isotherms for adsorption Pb(II) and Cu(II) ions onto mesoporous adsorbent.

3. 2. 4. Adsorption Isotherms

The isotherm models of Langmuir and Freundlich were used to describe the equilibrium adsorption isotherms. The Langmuir isotherm assumes monolayer coverage of adsorbate over a homogeneous adsorbent surface. The Freundlich isotherm is considered for describing both multilayer sorption and sorption on heterogeneous surfaces. The Langmuir isotherm is given by Equation (3):

$$\frac{C_e}{q_e} = \frac{1}{q_m b} + \left(\frac{1}{q_m}\right) C_e \tag{3}$$

where, q_e is the amount of metal ions adsorbed at equilibrium (mg. g⁻¹), q_m (mg.g⁻¹) is the maximum monolayer adsorption capacity, C_e is the equilibrium concentration of metal ion in mg.L⁻¹, and b is the constant that refers to the bonding energy of sorption in L.mg⁻¹ [17]. The Freundlich isotherm is given by Equation (4):

$$\ln q_e = \ln K_f + \left(\frac{1}{n}\right) \ln C_e \tag{4}$$

where, q_e is the amount of metal ions adsorbed at equilibrium (mg.g⁻¹), C_e is the equilibrium concentration of metal ion in mg.L⁻¹, K_f and n are the Freundlich constants related to the adsorption capacity and adsorption intensity, respectively [17]. The values of Freundlich and Langmuir constants for the adsorption of copper and lead metal ions onto CMK-3-EDA are presented in Table 2. As recorded in Figure 8, the Langmuir model was able to provide a better fit to the experimental data than the Freundlich mode.

3. 2. 5. Adsorption Kinetics

Kinetics of the adsorption process provides essential information on the reaction pathways. In order to investigate the mechanism of the adsorption of Cu(II) and Pb(II) ions onto adsorbent, different kinetic models such as the pseudo-first-order, pseudo-second-order and Elovich has been studied. The pseudo-first-order equation can be used to determine the rate constant for the adsorption process as follows:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{5}$$

where, q_e and q_t indicate the amounts of Cu(II) and Pb(II) adsorbed at equilibrium (mg.g⁻¹) and at time t (min), respectively, and K_1 is the rate constant (L.min⁻¹) [3]. The pseudo-second-order equation is as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{6}$$

where, K_2 (g.mg⁻¹.min.₁) is the rate constant of the second-order equation, q_t (mg.g⁻¹) and q_e (mg.g⁻¹) for Cu(II) and Pb(II) adsorbed at time t (min) and at

equilibrium, respectively [3]. A simplified linearized form of Elovich kinetic equation is presented as follows:

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln(t) \quad (7)$$

where, α ($\text{mg.g}^{-1}.\text{min}^{-1}$) is the initial adsorption rate and β is related to the extent of surface coverage and the activation energy involved in chemisorption (g.mg^{-1}) [18].

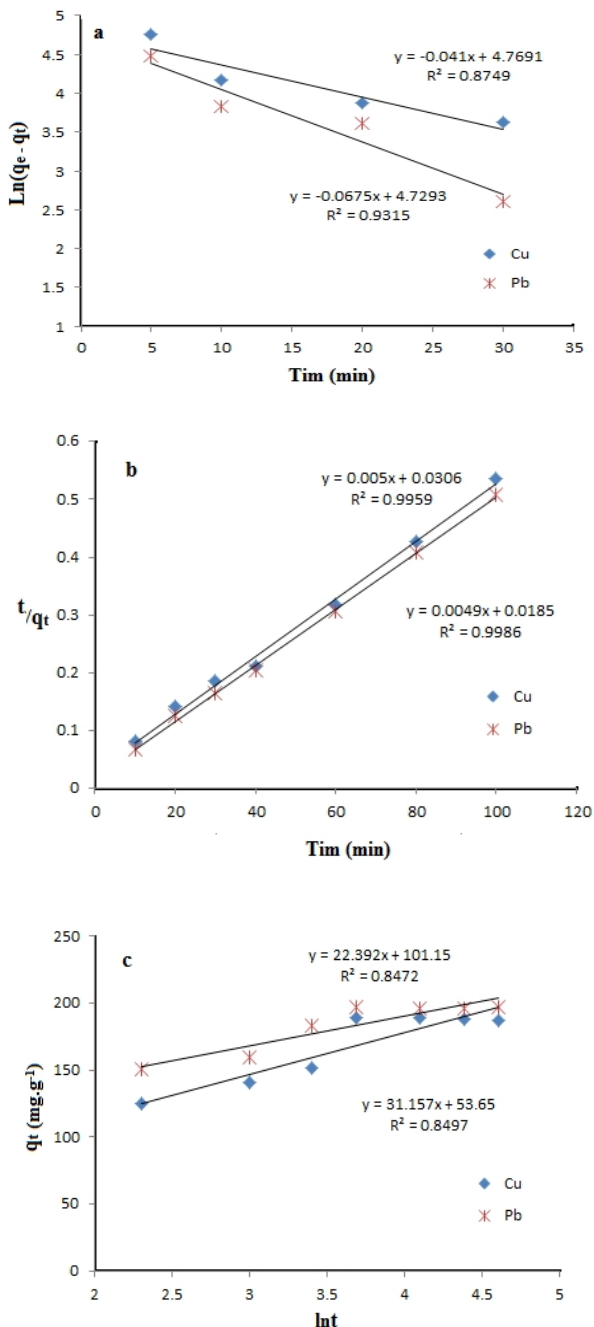


Figure 9. Pseudo-first-order (a), Pseudo-second-order (b) and elovich (c) kinetics models of Pb(II) and Cu(II) ions adsorption onto mesoporous adsorbent.

TABLE 2. Langmuir and Freundlich constants for adsorption of Cu(II) and Pb(II) on CMK-3-EDA.

Analyte	Freundlich			Langmuir		
	K_F (mg/g)	n (L/mg)	R^2	q_m (mg/g)	b (L/mg)	R^2
Lead	32.136	1.254	0.84	714.28	0.072	0.982
Copper	79.74	1.272	0.967	588.24	0.155	0.98

TABLE 3. Pseudo-first order, pseudo-second order and elovich constants for adsorption of Cu(II) and Pb(II) on CMK-3-EDA.

Different Kinetic Models	Coefficients	Cu (100 mg.L^{-1})	Pb (100 mg.L^{-1})
Pseudo-first order	$q_{e,\text{exp}}$ (mg.g^{-1})	188.7	196.7
	$q_{e,\text{cal}}$ (mg.g^{-1})	107.66	113.183
	K_1 (min^{-1})	0.041	0.0675
	R^2	0.875	0.935
pseudo-second order	$q_{e,\text{exp}}$ (mg.g^{-1})	188.7	196.4
	$q_{e,\text{cal}}$ (mg.g^{-1})	200	204
	K_2 ($\text{g.mg}^{-1}.\text{min}^{-1}$)	0.00816	0.00131
	R^2	0.996	0.999
Elovich	α ($\text{mg.g}^{-1}.\text{min}^{-1}$)	173.779	205.75
	β (g.mg^{-1})	0.0321	0.0445
	R^2	0.847	0.849

The results presented in Figure 9 show that the pseudo-second-order is able to provide better fit ($R^2 > 0.99$ for Cu(II) and Pb(II)) to the experimental data than the pseudo-first-order ($R^2 \leq 0.93$ for Cu(II) and Pb(II)) and the elovich ($R^2 \leq 0.85$ for Cu(II) and Pb(II)). Also, the calculated sorption capacity values obtained from pseudo-second order model are consistent with the experimental values of sorption capacity. The data presented in Table 3 were obtained from kinetic model equations.

3. 3. Comparison with other Adsorbents The q_e for the metal ions reported for different adsorbents is given in Table 4. In general, the CMK-3-EDA has higher adsorption capacity than the reported adsorbents. The main reason for this is attributed to the ordered mesoporous structure and the amine functional groups formed on the surface CMK-3.

TABLE 4. Comparison of adsorption capacity of Pb(II) and Cu(II) ions on various adsorbents.

Adsorbent	Metal	S _{BET} (m ² .g ⁻¹)	D _p (nm)	pH	Initial Concentration	Equilibrium time (min)	Q _e (mg.g ⁻¹)	Ref
EDTA-SBA-15	Pb	284	7.16	5.0	100 mg L ⁻¹	300	91.6	[7]
Fe ₃ O ₄ @APS@AA-co-CA.	Pb	-	-	5.5	100 mg L ⁻¹	90	78.8	[19]
CMK-3-COOH	Pb	2207	2.78	5	100 mg L ⁻¹	240	94	[20]
Fe ₃ O ₄ @APS@AA-co-CA.	Cu	-	-	5.5	100 mg L ⁻¹	90	62.4	[19]
Carbons derived from rice husk	Cu	425	-	6	200 mg L ⁻¹	180	22.62	[21]
CMK-3	Cu	851	4.30	6	100 mg L ⁻¹	60	34.21	This work
CMK-3	Pb	851	4.30	5	100 mg L ⁻¹	60	35.63	This work
CMK-3-EDA	Pb	344.7	2.86	5	100 mg L ⁻¹	60	196.64	This work
CMK-3-EDA	Cu	344.7	2.86	6	100 mg L ⁻¹	60	188.2	This work

4. CONCLUSION

In the present study, SBA-15 mesoporous silica has been synthesized by a hydrothermal method and then used as template silica along with sucrose as the carbon source for preparation of the CMK-3. Consequently, CMK-3 was carboxylated by ammonium persulfate ((NH₄)₂S₂O₈) and H₂SO₄, then functionalized with EDA. The resulting sample has been applied as a new adsorbent for removal of Pb(II) and Cu(II) ions. The results suggest that the adsorption is influenced by the solution pH, dosage of adsorbents and contact time. The Langmuir model is the best model to describe Cu(II) and Pb(II) ions adsorption behavior onto CMK-3- EDA. The pseudo-second-order was able to provide better fit (R² > 0.99 for Cu(II) and Pb(II)) to the experimental data than the pseudo-first-order (R² ≤ 0.93 for Cu(II) and Pb(II)) and the elovich (R² ≤ 0.85 for Cu(II) and Pb(II)). Under optimum experimental conditions, the maximum adsorption capacity were obtained as 188.2 mg. g⁻¹ and 196.64 mg. g⁻¹ for Cu(II) and Pb(II), respectively. The obtained high adsorption capacity of CMK-3-EDA is due to the amine functional groups formed on the surface of CMK-3 which can react with Cu(II) and Pb(II) in aqueous systems. Therefore, CMK-3- EDA can be used as a novel nanoporous sorbent for the effective removal of Cu(II) and Pb(II) ions from aqueous media.

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Synthesis of Ethylene diamine-modified Ordered Mesoporous Carbon as a New Nanoporous Adsorbent for Removal of Cu(II) and Pb(II) Ions from Aqueous Media

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مژوپروس کربن (CMK-3) عاملدار شده با اتیلن دی آمین سنتز گردید و به عنوان یک جاذب جدید برای حذف سرب و مس از محلول های ابی بکار برده شد. دستگاه جذب و واجذب نیتروژن (BET) نشان می دهد که مساحت سطح و سائز حفرات CMK-3 بعد از عاملدار شدن تغییر قابل توجهی کرده است. مساحت سطح و سائز حفرات محصول عاملدار شده به ترتیب $344.74 \text{ m}^2 \cdot \text{g}^{-1}$ و 28.61 \AA بود. شرایط موثر بر جذب شامل زمان تعادل، مقدار pH و مقدار جاذب، محلول نمونه در سیستم پیج بررسی گردید و سپس بوسیله اسپکتروسکوپی جذب اتمی شناسایی شد. تحت شرایط تجربی، ظرفیت جذب برای سرب و مس به ترتیب $196.64 \text{ mg} \cdot \text{g}^{-1}$ و $1188.2 \text{ mg} \cdot \text{g}^{-1}$ بود. ظرفیت جذب بالا در CMK-3 عاملدار شده با اتیلن دی آمین ناشی از گروه های عاملی امینی تشکیل شده بر سطح CMK-3 می باشد که می تواند با یون های مس و سرب برهمکنش داشته باشند. نتایج نشان می دهد که ماده متخلخل جدید سنتز شده ماده ای با کارایی بالا برای جذب سرب و مس در مقایسه با دیگر جاذب ها می باشد.

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