Preparation of Poly-Aniline-Magnetic Porous Carbon Composite for Using as Uranium Adsorbent

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Abstract: Rice husk as one of the agricultural by-products has been used as a starting material for the production of porous carbon (PC), the obtained porous carbon has been modified by poly aniline and magnetized to form poly aniline magnetic porous carbon composite (PA-MPC). The prepared PA-MPC has been characterized and examined for uranium adsorption from its pregnant solutions. The effect of the controlling parameters (pH; Contact time, temperature, initial uranium concentration, and adsorbent dosage) have been optimized. The obtained results indicate that the adsorption is pH dependant. The equilibrium data were well described by langmuir isotherm with theoretical capacity 94.3 mg/l at room temperature. These results demonstrate the potential use of this adsorbent for uranium ions removal from its pregnant solutions.

Keywords: Agriculture Waste, Rice Husk, Poly-Aniline, Uranium, Adsorption

1. Introduction

Rice is considered as one of the most important agriculture product all over the world. It plays an important role in food municipal consumption. Through rice refining process, rice husk is obtained as a by- product, which is characterized by low humidity and simple operation [1]. Rice husk ash characterizes by the presence 87-97% silica [2] as well as being highly porous and light in weight, also its surface area is very high.

Several studies have been performed on rice husk, where many researchers used rice husk in different applications relaying on its chemical and physical properties. From such uses are as a fuel in power plant [3 and 4], formation of activated carbon [5], source of silica and silicon compounds [6 and 7], insulating fire brick using RH [8], also in the manufacturing of building materials [9] and it has been used fuel in electricity generating [10].

The burning of husk led to the formation of rice husk charcoal, which has multilateral uses. Carbon and silica are the main constituents of rice husk and rice charcoal, so both rice husk and rice charcoal can be used as starting materials for the preparation of different adsorbent. The preparation of activated porous carbon from rice husk passes through two steps, carbonization then activation, the later can be done

either physically and/or chemically [11].

The aim of this work is to employ rice husk in the preparation of porous carbon, the later has been used in the preparation of magnetic poly-aniline porous carbon composite. The prepared composite has been tested as adsorbent for uranium ions removal. In this study, IR technique, TGA technique and BET theory have been applied to determine the physical characterization of prepared adsorbent. Batch adsorption technique has been done to examine the prepared composite for uranium adsorption. Equilibrium isotherms as well as kinetic and thermodynamic studies for U (VI) adsorption have been done.

2. Materials and Methods

2.1. Reagents

The chemicals used for different experiments are of analar grade. A stock solution from uranium (VI) (1000ppm) has been prepared by dissolving 2.11 gm of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ in de-ionized water besides adding 1 ml of concentrated HNO_3 .

2.2. Porous Carbon Preparation

The porous carbon has been prepared from rice husk as follow: the rice husk has been washed, dried and carbonized at 450°C in vacuum with nitrogen flow rate of 200 cm^3/min .

The carbonized product was heated with adequate weight from potassium hydroxide at 400°C for 60 min in order to dehydrate the mixture and increase the surface area. The mixture was activated by increasing the temperature in the range from 650 to 850°C for about one hour. Finally, the activated product was ground, washed with water and dried at 120°C to form the porous carbon (PC).

2.3. Preparation of Magnetic Porous Carbon

The magnetic porous carbon has been prepared using the modified method of Massart [12], where about 100 ml from ferric solution (0.2M) has been mixed with 100 ml from freshly prepared 0.1 M ferrous solution under stirring with ten gram from the produced porous carbon. After that, about 100 ml from ammonia solution (30%) has been poured to the $\text{Fe}^{+3}/\text{Fe}^{+2}/\text{PC}$ mixture and stirred vigorously. A black precipitate was formed which was left to crystallize for 30 min under stirring. The crystallized product was then washed with deoxygenated water under magnetic decantation until pH of suspension became below 7.5. The precipitate was dried at room temperature to give a black powder and marked as M- PC.

2.4. Polyaniline / Magnetic Porous Carbon Preparation

One gram from KIO_3 had been added to 100 mL from sulphuric acid (1 M) and stirred for about 30 min. to form homogenized solution. Then the mixture and 3 mL from fresh distilled aniline monomer were mixed with about two gram from porous carbon. The reaction was performed for about 5 hrs at room temperature with stirring in order to maintain the complete polymerization of aniline. After that, the resulted polymer was filtered on filter paper, washed several times with 0.2 M hydrochloric acid, then with acetone to separate the oligomers and impurities [13]. The final product was dried in oven at 60°C temperature for about 24 h and marked ad PA-MPC.

2.5. Materials Characterization

The prepared porous carbon and poly-aniline porous carbon composite has been characterized using the following techniques:

- (1) Fourier Transform Infrared Spectrometer (FTIR): (FTIR) model Thermo Scientific Nicolet IS10, Germany has been used for the determination of the structural characteristics of the prepared sorbent
- (2) The N_2 adsorption–desorption isotherms have been used to predict the pore size distribution. The specific surface area (SBET) has been recognized from the linear segment of the N_2 adsorption isotherms by applying Brunauer–Emmett–Teller (BET) theory.
- (3) Thermal stability and the functional group content

2.6. Batch Adsorption Experiments

To figure out the contribution of the controlling factors on the adsorption performance, several experiments have done using fixed amount from PA-MPC (0.1 gm), placed on a

capped flask which was set on a water bath shaker. Each of the controlling factors has been studied alone by varying its range and keeping all the other factors fixed. The ranges of each factor are as follow: pH from 1 to 9, contact time from 15 to 240 min, initial concentration from 50 to 1400 mg/L, temperature from 25 to 80°C, and solid dosage from 0.05 to 0.4g. The pH was maintained using 0.1 M from HCl and /or 0.1 M from NaOH. The effect of competing ions was investigated using different concentrations from these ions ranging from 50 to 500 mg/l and each has been studied solely.

The adsorbed uranium amounts obtained from different investigation are determined using the following function

$$q_e = \frac{(C_o - C_e)V_o}{M} \quad (1)$$

Where q_e is the adsorption amount in mg/g, M is the weight of the adsorbent (g), V is the volume of solution (L) and C_o and C_e are the initial and equilibrium concentrations of metal ions in solution.

2.7. Adsorption Isotherm

To describe the adsorption behavior of U (IV) ions onto PA-MPC, classical isotherm models have been studied. These models are:

Langmuir Adsorption Isotherm [14]: which depicts the formation of a monolayer adsorbate on the outer surface of the adsorbent [15] using the following equation:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{Q_m b} \quad (2)$$

Where Q_m is the saturated monolayer adsorption (mg/g), b is the Langmuir constant related to the affinity of binding sites and is a measure of the adsorption energy (mL/mg).

Freundlich Adsorption Isotherm [16] which figures the adsorption characteristics for the heterogeneous surface [17] by applying linearized equation:

$$\log q_e = \log K_f + \frac{1}{n \log C_e} \quad (3)$$

Where K_f is the adsorption capacity (mg/g) and $1/n$ is the Freundlich constant indicating adsorption intensity.

2.8. Kinetics Study

To investigate the mechanism of adsorption, pseudo first order and pseudo second- order models have been estimated. These models can be expressed in linear forms respectively [18 and 19]

Pseudo first order:

$$\ln(q_e - q_t) = \ln q_e - K_1 t \quad (4)$$

Pseudo second order:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_t} \quad (5)$$

Where q_t and q_e are the amounts of thorium ions adsorbed (mg/g) at time t (min) and at equilibrium time, respectively; k_1 (min^{-1}) and k_2 (g/mg min) are the pseudo-first-order and the pseudo second- order sorption rate constant, respectively

3. Results and Discussion

3.1. Materials Characterization

3.1.1. FTIR Spectroscopy

The figured data of FT-IR for PA-MPC presents in figure

Figure 1. FT-IR spectra of PA-MPC before and after uranium ions adsorption.

3.1.2. Raman Spectra

The second tool used for composite characterization is Raman spectroscopy which is considered one of the most useful tools for carbonaceous materials characterization. From figure (2), the typical G-band in Raman spectrum for PC consists of a strong line situated at 1574 cm^{-1} , the band of PC has a shoulder extending to higher wave numbers for disorder carbons additional bands appears at 1355 cm^{-1} (D1-band) [20], The second-order Raman band

is assigned to overtone at 2643 cm^{-1} broadened with increasing disorder. After the polymerization of PC with PANI, the spectrum of this polymer dominates in all samples. This observation confirms that polymerization occurs between PC and PANI. The peaks in the spectra are typical of protonated PANI and are located at 1593 , 1504 , 1330 , and 1171 cm^{-1} [21]. Their positions remain practically unchanged after polymerization with observed increase of the intensity of the peaks.

Figure 2. Raman spectra of prepared adsorbent.

3.1.3. Thermal Stability Studies

Thermo gravimetric analysis (TGA) of PC and PANI show decomposition in the temperature range from 190 to 590°C and residual weight after this temperature is negligible (figure 3), while in the case of PC/PANI composite shows

thermal stability up to 640°C and become completely decomposed above 750°C, the TGA profile also confirmed that a significant amount of the functional groups have been chemically grafted on the carbon surface, and the resulting material has excellent thermal stability [22 and 23].

Figure 3. Thermo gravimetric analysis of prepared adsorbent.

3.1.4. Surface Area and Porosity

Figure (4) shows the N₂ adsorption isotherms and the corresponding pore size distribution curves of the porous carbon and the PC/poly-aniline composite sorbants, respectively. The BET surface area, total pore volume and

pore size of these materials are presented in table 1. It is noteworthy that when poly-aniline was introduced into porous carbon substrates, the BET surface area and total pore volume of PC/poly-aniline significantly decreased to approximately half the level of porous carbon.

Figure 4. N₂ adsorption-de-sorption isotherms for the porous carbon and porous carbon/ poly-aniline composite.

Figure (5) shows that the value of the dV/dD greatly decreased in the pore size range of 1–20 nm, but the peak shifted slightly to a lower value as poly-aniline was incorporated into Porous carbon. These results suggest two

possibilities: (1) the surface of the nano-pores of porous carbon is modified with poly-aniline or (2) some parts of the nano-pores of porous carbon are filled with poly-aniline.

Figure 5. The corresponding pore size distribution for the porous carbon and porous carbon/ poly-aniline composite.

Table 1. The BET surface area, total pore volume and pore size of porous carbon and porous carbon/ poly-aniline composite.

Sample	$S_{BET}(\text{m}^2/\text{g})$	$V_t(\text{ml/g})$	$V_{\text{micro}}(\text{ml/g})$	$V_{<1\text{nm}}(\text{ml/g})$
Porous carbon	1639	1.26	1.21	2.86
	1632	1.23	1.18	2.89
PA Composite	831	0.495	0.99	2.21
	828	0.491	0.94	2.23

3.2. Effect of Controlling Factors

3.2.1. Effect of pH

The solution pH is considered as the most dominant factor that has a clear effect on the adsorption phenomena in most

cases, because pH not only affects the solubility and speciation of metal ions in solution but also on the net charge of the adsorbent. The uranium species found in the solution depends on the pH as well as the composition of the solution [24]. The plotted results in figure (6) assure that the solution pH is a controlling factor in the uranium adsorption onto the surface of PA-MPC. In the high acidic media the uranium adsorption is minimum ($\text{pH} \leq 1$), with the increase in the pH from 2 to 5, the percent of adsorption increases and the maximum adsorption has been attained at pH 5. With the increase in the pH of the solution the adsorption percent decrease (figure 6).

Figure 6. Effect of pH on uranium ions adsorption onto PA-MPC surface.

The adsorption onto PA-MPC occurs mainly through two ways: physical (which is back to the properties of the adsorbent surface) and/ or chemically through complexation with the functional group (mainly oxygen-containing groups) on the carbon surface which is affected by the solution pH. Complexation played an important role where at lower pH, the surface functional groups linked to the H^+ , making these inaccessible for uranium ions. With increasing the pH value, the de-protonation of the functional groups provided the chance to make a complex with uranium ions resulting in higher adsorption capacity. Similar results had been observed for uranium adsorption on hematite, and amorphous iron hydroxide [25 and 26].

3.2.2. Effect of Initial Uranium Concentration

The effect of initial uranium concentration on the adsorbed

quantity under three different temperature has been shown in figure (7). It can be seen from the figure that the percent of adsorbed uranium decrease with the increase in the uranium concentration at different temperature degrees. This may be back to the following, at lower initial concentrations; sufficient adsorption sites are free for metal ions adsorption, which gives an indication that the fractional adsorption is metal ions initial concentration independent. At higher concentrations, the uranium ions are relatively higher compared to availability of adsorption sites. Hence, the adsorption percent depends on the initial metal ions concentration and decreases with increase in initial metal ions concentration. Also the obtained results show that the temperature has a positive effect on the uranium adsorption percent.

Figure 7. Effect of initial uranium concentration on uranium ions adsorption efficiency onto PA-MPC.

Adsorption Isotherms

The obtained results of uranium adsorption isotherms tabulated in table (2) show that under the experimental conditions listed previously, the obtained data fits well with langmuir isotherm rather than freundlich isotherms, although the correlation coefficient (R^2) for freundlich isotherms is

above 0.99, but the experimental data here is the controlling factor. The maximum adsorption value obtained in accordance to langmuir is 94.3 mg/g at room temperature. The obtained value of n in freundlich isotherm (table 2), is less than 1 indicating normal adsorption [27].

Table 2. Parameters for plotting Langmuir, Freundlich adsorption Isotherms of uranium ions onto PA-MPC.

Temperature	Langmuir pararmeters				Freunlich pararmeters			
	$q_{max}(mgg^{-1})$	$B (Lmg^{-1})$	R^2	equation	$K_f (mg g^{-1})$	n	R^2	equation
298	94.3	0.063	0.99	$Y=0.0106x+0.1683$	26.06	0.2	0.97	$y = 0.2175x + 1.416$
313	103.09	0.057	0.98	$Y=0.0097x+0.1669$	26.01	0.21	0.98	$y = 0.2315x + 1.4188$
343	117.22	0.04	0.98	$Y=0.0085x+0.1832$	26.02	0.255	0.98	$y = 0.2552x + 1.4151$

3.2.3. Effect of Adsorbent Dose

In order to investigate the effect of adsorbent dosage on uranium adsorption, several experiments have been performed with constant uranium concentration (1500 mg/L) and different PA-MPC dosage ranging from 0.025 to 0.125 g/L. The obtained experimental results of adsorption plotted

in Fig. (8) show that the adsorption capacity increased with increasing adsorbent amount and then became constant indicating that 0.05 g for every 20 ml was the optimum adsorbent dosage and will be used in the following sorption experiments.

Figure 8. Effect of adsorbent dosage on uranium adsorption efficiency onto PA-MPC.

3.2.4. Effect of Contact Time

Figure (9) shows the influence of contact time on adsorption efficiency of PA-MPC for uranium ions. The experimental data indicate that, the adsorption capacity of PA-MPC increased by increasing contact time. This is probably due to the higher contact between the surface of

PA-MPC and uranium ions [28]. The obtained data also reveals that the initial uranium concentration has a significant role on the adsorption capacity, where by increasing the concentration of uranium ions the adsorption capacity increases. At all concentrations the system reached equilibrium state after 60 minute.

Figure 9. Effect of contact time on uranium ions adsorption efficiency onto PA-MPC.

Adsorption Kinetics

The obtained results of uranium adsorption kinetics listed in table 3, show that the correlation coefficient (R^2) of the pseudo-second-order are 0.98, 0.98 and 0.95 and the theoretical capacity are 49, 104.1 and 128.2 mg g^{-1} which is close to the experimental value for the investigated

concentrations 100, 400 and 600 mg g^{-1} respectively. While the theoretical capacity calculated from pseudo first order is far away from the real experimental capacities. These results indicate that the adsorption process of uranium ions on PA-MPC can be expressed by pseudo-second-order kinetic model.

Table 3. Kinetic model parameters applied to uranium ions sorbed onto PA-MPC.

concentration mg/L	First order				Second order			
	q_e (mgg ⁻¹)	K_1 (min ⁻¹)	R^2	equation	q_e (mgg ⁻¹)	K_2 (g mg ⁻¹ .min ⁻¹)	R^2	equation
100	93.	0.0175	0.7	$y = -0.0762x + 4.5404$	49.4	7.06×10^{-4}	0.98	$y = 0.0204x + 0.5363$
400	199.9	0.083	0.65	$y = -0.0427x + 4.1557$	104.1	5.9×10^{-4}	0.98	$y = 0.0096x + 0.177$
600	129.58	0.081	0.75	$y = -0.0811x + 5.6073$	128.2	2.06×10^{-4}	0.95	$y = 0.0078x + 0.2949$

3.2.5. Influence of Coexisting Cations

The adsorption behavior of uranium ions in the presence of different cations like Ca^{2+} , Na^+ , Fe^{3+} , Mn^{2+} and Mg^{2+} has been tested using fixed concentrations of these cations (100 mg/L). Adsorption experiment has been performed using these cations gathered together and the extraction efficiency has been determined. The obtained results show that these cations have no influence on uranium adsorption efficiency, where the adsorption efficiency is almost the same $\approx 92\%$ except for iron which is slightly increased to reach 94 %. This may be back to the following: Fe^{3+} has also the capacity of adsorption. From this point of view, adsorbed Fe^{3+} on PA-MPC has role in the adsorption process. These results is in agree Kütahyalı and Eral [29], where they found that Fe^{3+} has a positive effect on the uranium adsorption by using chemical modified activated carbon.

3.2.6. Influence of Coexisting Anions

The adsorption behavior of uranium ions in the presence of different anions like Cl^- , CO_3^{2-} , HCO_3^- , SO_4^{2-} , OH^- and NO_3^- has been examined. The investigated anions concentrations have been fixed at 100 ppm and the pH of the solution was kept constant at 5. Sodium ion was used as the cationic component for the investigated anions. The obtained results show that PA-MPC show good affinity for uranium extraction even in the presence of high concentration of anionic complexing agents and common electrolyte species the uranium extraction efficiency was 20, 92, 90, 55, 95 and 72 % for the investigated anions CO_3^{2-} , HCO_3^- , SO_4^{2-} , OH^- and NO_3^- respectively.

The presence of some anions can enhance the adsorption capacity by pairing with the uranium species, hence, reducing the repulsion between adjacent uranium molecules adsorbed on the surface. This enables the carbon to adsorb more of the positive uranium ions since the repulsive forces between the surface and uranium ions and repulsive force among positive uranium ions are reduced. It has been noticed that, although carbonates ions are the most known anions which interfere with the sorption of U (VI) due to the formation of stable tricarbonat complex, $\text{UO}_2(\text{CO}_3)_3^{4-}$, but it did not interfere with the uranium adsorption at pH 5. This can be explained as follow: for pH higher than 7.5 and pH lower than 3.5, the uranium complexes with phosphates and carbonates predominate. In the region between 5.5 and 7.5, the predominant species were the polynuclear hydroxo-complexes while for $3.5 < \text{pH} < 5.5$, the uranyl ion was the major uranium species formed [30].

De-sorption and reusability

One of the parameters used in the judgment of the prepared adsorbent is its regeneration or reusability ability in order to improve its cost effectiveness for reuses in multiple

cycles. As desorbing agent, HCl and H_2SO_4 solutions were used for desorbing previously adsorbed U (VI) ions onto PA-MPC. Various concentrations from HCl and H_2SO_4 ranging between 0.001 and 0.2 M were tested for adsorbed U (VI) ions elution from PA-MPC. It was found that that HCl was better than H_2SO_4 as desorbing agent. The obtained U (VI) de-sorption percent was 70.1, 89.2, 98.2, 98.5 and 98.9% with 0.01, 0.05, 0.10, 0.15 and 0.2 molL⁻¹ HCl, respectively. Therefore, the best optimum concentration was set as 0.1 mol·L⁻¹ in terms of economical process.

The regeneration-reuse property is important for the application of adsorbents in real work, because an appropriate adsorbent should present not only excellent selective adsorption capability but also regeneration–reuse property. In this paper, the reusability of PA-MPC composites was investigated by washing with 0.1 mol/L HCl and rinsed with water thoroughly

The regenerated PA-MPC composites showed similar removal percentage of U (VI) as compared with pristine PA-MPC composites. It indicates that PA-MPC composites can be easily regenerated by treating U (VI)-loaded PA-MPC composites with conventional acidic solution (0.1 mol/L HCl). Moreover, these high adsorption capacities of regenerated PA-MPC composites were kept even after (at least) six cycles of regeneration–reuse applications.

4. Conclusion

Poly-aniline magnetic porous carbon composite has been prepared, characterized and examined for uranium adsorption from its pregnant solution. The controlling factors have been determined and the obtained results reveal the following: the adsorption of uranium ions onto PA-MPC is pH dependent with maximum adsorption at pH 5 and reaching the equilibrium after 60 minutes. The experimental maximum adsorption capacities of PA-MPC is found to be 94, 103 and 117 at 298, 313 and 343 K respectively, which indicates that the temperature has a positive effect on uranium adsorption. HCl can be used as desorbing agent for uranium ions from the surface of PA-MPC.

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