



# Uranium Removal from Aqueous Solution by Zeolite from Fly Ash-Iron Oxide Magnetic Nanocomposite

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**Abstract** – The adsorption characteristic of zeolite was combined in a composite with the magnetic properties of iron oxide to produce magnetic adsorbent material. Magnetite nanoparticles in suspension were synthesized by precipitating  $Fe^{2+}$  ions in a basic solution. The zeolite was synthesized by alkaline hydrothermal treatment of coal fly ash. The zeolite-iron oxide magnetic nanocomposite was prepared by mixing zeolite from coal fly ash with magnetite nanoparticles in suspension. The magnetic nanocomposite was used for the removal of U(VI) from aqueous solutions by a batch technique. The effects of contact time and initial concentration on the removal process were evaluated. The kinetic experimental data were analyzed using three kinetic equations including pseudo-first order equation, pseudo-second order equation and intraparticle diffusion model. The experimental data fit the second-order kinetic model. The Freundlich and the Langmuir models have been applied and the results indicated that the Langmuir provided the best correlation of experimental data. Copyright © 2012 Praise Worthy Prize S.r.l. - All rights reserved.

Keywords: Adsorption, Coal fly ash, Magnetic Nanocomposite, Uranium, Zeolite

## I. Introduction

Uranium is present in the environment as a result of emissions from the nuclear industry, slowly released from natural deposits, discharge from mill tailings, combustion of coal and other fossil fuels, and use of uranium-containing phosphate fertilizers. The uranium concentration in Brazilian phosphate fertilizers ranges from 5.17 to 54.3 mg kg-1 [1]. In oxidizing environments it occurs in the valence state + VI as the uranyl ion,  $\rm UO_2^{2^+}$ . Uranium is both radioactive and toxic with the liver and kidneys being the main targets of uranium's toxic effects and with reservoirs of uranium concentrating in the kidneys and bones.

Uranium ends up in the human body through eating or drinking it or by inhaling dust particles containing uranium. It can cause cancer and its primary toxic effect when consumed in water is that of heavy metals [2]-[3].

Hence, research into the separation of U(VI) ions from wastewater is important. Many processes have been proposed for U(VI) ion removal from industrial wastewaters and radioactive wastes, with chemical precipitation, ion-exchange, membrane processes, solvent extraction and adsorption being the most commonly used methods employed [4]-[8]. A number of adsorbent materials are being investigated for the removal of U(VI) ions from aqueous solutions including magnetite [9], natural [10] and synthetic zeolites [11]. Zeolites are crystalline aluminosilicates with framework structure and have excellent cation exchange properties.

They can be natural minerals that are mined in many parts of the world or synthetically produced.

Coal Fly ash is a major solid waste from coal-firing power stations. In Brazil, more than 4 million tons/year of fly ash was generated with a tendency to increase every year.

Although many applications or utilizations and researches on the effective utilization of coal fly ash have been made, the most coal fly ash is used in construction-related applications [12], mainly cement and concrete manufacturing.

Under these circumstances, the effective utilization of coal ashes is strongly desired from environmental, ecological and economical point of view. Some zeolitic materials have also been successfully synthesized from coal ashes by relatively inexpensive and quick hydrothermal conversion processes and have been widely used as low-cost adsorbents [13]-[14].

In our group we have been developing zeolites synthesized from coal fly ashes to remove metals ions and dyes from aqueous effluents [14]–[18].

Magnetic composites based on zeolite/iron oxide have been prepared and used for the removal of contaminants from aqueous effluents [19]-[21]. Magnetically modified zeolites offer the advantage of easy separation and recovery of powdered zeolites after environmental remediation by means of an applied magnetic field.

The objective of this study was to assess the feasibility of using zeolite from fly ash-supported magnetite nanoparticles for removing U(VI) from aqueous solutions. For this purpose, the magnetic nanocomposite was prepared by mixing zeolite from coal fly ash with magnetite nanoparticles in suspension.

## II. Materials and Methods

#### II.1. Materials

All chemicals used in this study were of analytical grade. The sample of coal fly ashes was obtained from Presidente Médici coal-fired power plant located in Candiota, state of Rio Grande do Sul in the south of Brazil. A standard solution of uranyl nitrate was prepared by dissolution of  $U_3O_8$  nuclear pure obtained from Environment and Chemistry Centre at Nuclear and Energy Research Institute (IPEN), Brazil. Solutions of uranium (VI) were prepared by dissolving an appropriate amounts of standard stock solution of  $UO_2$  ( $NO_3$ )<sub>2</sub>.6H<sub>2</sub>O ( $130.07 \pm 0.01 \text{ g L}^{-1}$ ) in with deionized water (Milli-Q System) at the required initial concentrations. The initial pH values of the working solutions were adjusted by addition of  $0.5 \text{ mol L}^{-1}$  NaOH solution.

#### II.2. Preparation of the Zeolite from Fly Ash

Coal fly ash was used as starting material for zeolite synthesis by means of hydrothermal treatment. In synthesis experiment, 20 g of fly ash was heated to 100  $^{\circ}$ C in an oven for 24 h with 160 mL of 3.5 mol L<sup>-1</sup> NaOH solution. The zeolitic material was repeatedly washed with deionized water until pH 11 and dried at 50  $^{\circ}$ C for 12 h [22]. The zeolitic product obtained was labeled as ZFA.

## II.3. Preparation of the Zeolite from Fly Ash-Iron Oxide Magnetic Composites

In the first step, magnetite particles were prepared by adding of 2 mol L<sup>-1</sup> NaOH solution drop by drop in a 100 mL solution of ferrous sulfate (1.8 g) with agitation (Ultra-Turrax Q252-K28, Quimis) until the pH reached 11. The slurry was heated on a water bath. After that, the magnetite was washed with distilled water and dried at room temperature. One part of magnetite was dried at room temperature to obtain magnetite powder (MAG).

Other part of magnetite particles was redispersed in aqueous solution and zeolite synthesized from fly ash (ZFA) was added slowly with agitation. The zeolite/magnetite ratio was 3:1 (w/w). The obtained zeolite from fly ash-iron oxide magnetic composite was washed with distilled water, dried at room temperature and milled [21]. The magnetic nanocomposite obtained was labeled as ZFAM.

## II.4. Adsorption Experiments

The adsorption was performed using the batch procedure. The experiments were carried out by shaking 10 mg of adsorbent (ZFAM) with 1 mL of uranium solutions with initial concentrations of 100, 225 and 500 mg  $L^{-1}$  at room temperature (25  $\pm$  2°C) in a shaker operated at 200 rpm for time interval between 5-180 min in glass bottles. Samples were withdrawn at appropriate time intervals and placed on top of a magnet for 2 min to

settle the particles and an aliquot of 0.9 mL of supernatant was withdrawn with a pipette and placed in a new flask. This flask was then placed on the magnet for 1 h. The final concentration of uranium in the supernatant was measured at 650 nm using a spectrophotometer UV-Vis (model B582, Micronal) by the arsenazo III method. For adsorption isotherms, samples with concentrations ranging from 100 to 500 mg  $L^{-1}$  were agitated till the equilibrium was achieved. The experiments were triplicate to determine the precision of the results. The adsorption capacity (mg g<sup>-1</sup>) of adsorbent was calculated using the Eq. (1):

$$q_t = \frac{\left(C_0 - C_t\right)V}{M} \tag{1}$$

where  $q_t$  is the adsorbed amount of adsorbate per gram of adsorbent at any time t,  $C_0$  and  $C_t$  the concentrations of the adsorbate in the initial solution and at any time t, respectively (mg  $L^{-1}$ ); V the volume of the adsorbate solution added (L) and M the amount of the adsorbent used (g). Through the following equation (Eq. (2)), it was possible to determine the extraction efficiency in the different experiments performed:

$$R(\%) = \left(\frac{C_0 - C_t}{C_0}\right) \times 100 \tag{2}$$

where R is the efficiency of extraction or retention percentage,  $C_0$  (mg  $L^{-1}$ ) is the initial concentration of each adsorbate and  $C_t$ (mg  $L^{-1}$ ) represents the concentration of the adsorbate at time t.

## II.5. Kinetic Models

A study of adsorption kinetics is desirable as it provides information about the mechanism, which is important for the efficiency of the process. It is important to be able to predict the rate at which contamination is removed from aqueous solutions in order to design an adsorption treatment plant. Several kinetic models are available to examine the controlling mechanism of the adsorption process and to test the experimental data. The rate constant of the adsorbate removal from the solution by zeolite-iron oxide magnetic composites was determined using Lagergren pseudo-first-order (Eq. (3)) and pseudo-second-order (Eq. (4)) with equations as follows:

$$log\left(q_{e}-q_{t}\right) = log - \frac{k_{1}t}{2.303} \tag{3}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \tag{4}$$

where  $q_e$  is the amount of ion adsorbed at equilibrium (mg g<sup>-1</sup>), qt is the amount of ion adsorbed at time t (mg g<sup>-1</sup>),  $k_I$  is the rate constant of the pseudo-first-order

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International Review of Chemical Engineering, Vol. 4, N. 3 Special Section on Open Door Initiative (ODI) - 2<sup>nd</sup> edition sorption (min<sup>-1</sup>), and  $k_2$  is the rate constant of the pseudo-second-order kinetics (g mg<sup>-1</sup> min<sup>-1</sup>) [23]-[24].

The initial adsorption rate, h (mg g<sup>-1</sup> min<sup>-1</sup>), as  $t\rightarrow 0$  can be defined as (equation (5)):

$$h = k_2 q_e^2 \tag{5}$$

where  $k_2$  is the rate constant of the pseudo-second order kinetics (g mg<sup>-1</sup> min<sup>-1</sup>)and q<sub>e</sub> is the maximum adsorption capacity (mg g<sup>-1</sup>).

The possibility of intraparticle diffusion resistance affecting adsorption was explored using the appropriate intraparticle diffusion model as [25]:

$$q_t = k_{id}t^{0.5} + C (6)$$

where  $k_{id}$  is the intraparticle diffusion rate constant (mg g<sup>-1</sup> min<sup>-0.5</sup>). According to Eq. (6), a plot of qt versus  $t^{0.5}$  should be a straight line with a slope  $k_{id}$  and intercept C when adsorption mechanism follows the intraparticular diffusion process. Values of the C give an idea about the thickness of boundary layer, i.e., the larger the intercept is the greater the boundary layer effect.

## II.6. Adsorption Isotherms

The equilibrium adsorption isotherm is of importance in the design of adsorption systems. Several adsorption isotherm equations are available and two important isotherms are selected in this study, the Langmuir and Freundlich isotherms [26]–[27].

The equilibrium data obtained in the present study were analyzed using the linear forms of the expressions of Langmuir (Eq (7)) and Freundlich (Eq (8)) isotherm models:

$$\frac{C_e}{q_e} = \frac{1}{Q_0 b} + \frac{C_e}{Q_0} \tag{7}$$

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \tag{8}$$

where  $C_e$  is the equilibrium concentration (mg L<sup>-1</sup>),  $q_e$  the amount adsorbed at equilibrium (mg g<sup>-1</sup>),  $Q_0$  is the maximum amount of adsorbate per unit weight of adsorbent to form a complete monolayer on the surface (mg g<sup>-1</sup>); b is the Langmuir isotherm constant (L mg<sup>-1</sup>), related to the affinity of the adsorption sites;  $K_F$  [(mg g<sup>-1</sup>) (L mg<sup>-1</sup>)<sup>1/n</sup> and n are the Freundlich constants related to adsorption capacity and adsorption intensity of adsorbents, respectively.

## II.7. Characterization Techniques

Chemical composition of materials was determined by X-ray fluorescence (XRF) in a Rigaku RIX-3000 equipment. X-ray diffraction (XRD) with an automated

Rigaku MiniFlex II diffractometer using  $CuK\alpha$  radiation ( $\lambda$ =1.5405Å) was used to identify the main crystalline compounds. The crystalline phases present in the samples were identified with the help of ICDD (International Centre for Diffraction Data).

## III. Results and Discussion

#### III.1. Characterization of Materials

The chemical composition of ZFA and ZFAM determined by X-ray fluorescence (XRF) is shown in Table I. The materials were characterized mainly as silica, alumina, iron oxide and sodium oxide. A significant amount of Na element incorporated in the ZFA is due to the hydrothermal treatment of coal fly ash with NaOH solution. The magnetic composite was prepared by the precipitation of iron oxide or hydroxide onto a zeolite surface. It can be observed that ZFAM has a higher content of Fe oxide, and the impregnation increases the content of iron to 87%.

TABLE I
CHEMICAL COMPOSITION OF ZFA AND ZFAM

CHEMICAL COMI OSTTION OF ZI A AND ZI AM				
Compound	ZFA	ZFAM		
	(% weight)	(% weight)		
$SiO_2$	$43 \pm 1$	23 ± 1		
$Al_2O_3$	$32 \pm 1$	$17.2 \pm 0.8$		
$Na_2O$	$10.1 \pm 0.5$	$2.7 \pm 0.1$		
$Fe_2O_3$	$7.1 \pm 0.4$	$53 \pm 2$		
CaO	$3.5 \pm 0.2$	$1.9 \pm 0.1$		
MgO	$1.5 \pm 0.1$	$0.8 \pm 0.1$		
$TiO_2$	$1.3 \pm 0.1$	$0.7 \pm 0.1$		
$K_2O$	$0.7 \pm 0.1$	$0.43 \pm 0.04$		
$SO_3$	$0.39 \pm 0.04$	$0.21 \pm 0.03$		
MnO	$0.05 \pm 0.01$	$0.07 \pm 0.01$		
$Cr_2O_3$	$0.03 \pm 0.01$	$0.02 \pm 0.01$		
ZnO	$0.02 \pm 0.01$	$0.02 \pm 0.01$		
SrO	$0.02 \pm 0.01$	< 0.01		
NiO	< 0.01	$0.02 \pm 0.01$		
CuO	< 0.01	< 0.01		
$ZrO_2$	< 0.01	< 0.01		
$Y_2O_3$	< 0.01	< 0.01		
$Rb_2O$	< 0.01	< 0.01		

Fig. 1 shows the X-ray diffraction (XRD) pattern of ZFAM sample. XRD analyses indicated that Na-P1 and hydroxysodalite were the dominant zeolites type. The characteristic XRD peaks of mullite and quartz still remain after 24 h of hydrothermal reaction of coal fly ash, which demonstrated the low reactivity of these phases in alkaline solutions. The presence of peaks in 20 values of 18.2, 30.0, 43.1, 56.95, and 62.9 can illustrate the incorporation of magnetite in the zeolite structure.

## III.2. Adsorption Kinetic Study

The rate of U(VI) adsorption on ZFAM was determined as a function of the initial U(VI) ions concentrations. The uptake of U(VI) ions for three initial U(VI) concentrations at different contact time is listed in Fig. 2. As seen from Fig. 2, lowest concentration showed the highest % removed U(VI) and, the efficiency of ion removal was increased as the agitation time increased.

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International Review of Chemical Engineering, Vol. 4, N. 3 Special Section on Open Door Initiative (ODI) - 2<sup>nd</sup> edition The removal decreased from 100 to 45% by increasing ZFAM concentrations from 100 to 500 mg L<sup>-1</sup>. It can be attributed that the active sites on adsorbent for ion removal decreases when ion concentration increases.

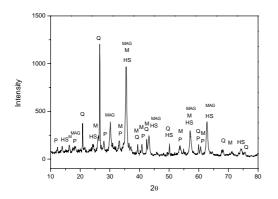


Fig. 1. X-ray diffractogram for ZFAM. M = mulitte; Q = quartz; MAG= magnetite; H = zeolite hydroxysodalite; P = zeolite Na-P1

The results showed that kinetics of adsorption of U(VI) ions consisted of two phases: an initial rapid phase where adsorption was fast and contributed significant to equilibrium uptake and a slower second phase whose contribution to the total U(VI) ions adsorption was relatively small. It is clear from Fig. 2 that the adsorption efficiency increases rapidly during the first 50 min and it was necessary 180 min to attain the equilibrium. This equilibrium time were used in all subsequent adsorption tests.

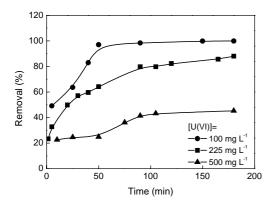


Fig. 2. Effect of contact time and initial concentration on U(VI) adsorption onto composite adsorbent (pH= 3.0; T= 25 °C)

In order to investigate the adsorption kinetics of U(VI) onto ZFAM, the pseudo-first order kinetic model, pseudo-second order kinetic model and intraparticle diffusion model were applied. The results of the experimental data fitting with these models for the adsorption of uranium (VI) on zeolite ZFAM are shown Table II. The coefficients of correlation,  $R_2$  for the pseudo-second-order adsorption model have a high value ( $\geq 0.97$ ). In addition, equilibrium adsorption capacities from the experiment ( $q_e$ exp) were better closed to those determined from the pseudo-second order model ( $q_e$ calc) than those from the pseudo-first order model. These facts suggest that the pseudo-second order adsorption

mechanism is predominant. The values C provide information about the thickness of the boundary layer because the resistance to the external mass transfer increases as the intercept increases. Correlation coefficient ( $R_i$ ) values given in Table II ranged between 0.381 and 0.973, confirming that the rate-limiting step is not actually the intraparticle diffusion. The nonlinearity of the plot of U(VI) with initial concentration of 100 mg  $L^{-1}$  demonstrated that intraparticle diffusion did not play a significant role in the uptake of the ion by adsorbent in this concentration.

TABLE II KINETIC PARAMETERS OF U(VI) IONS ADSORPTION ONTO ZFAM

[U(VI)] (mg/L)  RINETIC PARAMETERS OF U(VI) IONS ADSORPTION ONTO ZFAM  [V(VI)] (mg/L)  Pseudo-first order					
	K <sub>1</sub> (1/min)	q <sub>e</sub> calc (mg/g)	q <sub>e</sub> exp (mg/g)	$R_1$	
100	3.87 x 10 <sup>-3</sup>	0.448	10.4	0.244	
225	26.9 x 10 <sup>-3</sup>	15.0	19.3	0.992	
500	9.42 x 10 <sup>-3</sup>	11.5	23.3	0.814	
Pseudo-second order					
	$\mathbf{K}_2$	h	qecalc	q <sub>e</sub> exp	D
	(g/mg min)	(mg/g min)	(mg/g)	(mg/g)	$R_2$
100	68.7 x 10 <sup>-3</sup>	7.09	10.2	10.4	0.999
225	$3.44 \times 10^{-3}$	1.46	20.6	19.3	0.995
500	2.13 x 10 <sup>-3</sup>	1.17	23.4	23.3	0.966
Intraparticle Diffusion					
	С	$K_{i}$	$R_{i}$		
	(mg/g)	$(mg/g min^{0.5})$	<b>K</b> i		
100	9.78	0.0261	0.381		
225	4.72	1.27	0.973		
500	9.60	0.910	0.841		

## III.3. Adsorption Isotherms

To optimize the design of an adsorption system for the removal of adsorbate, it is important to establish the most appropriate correlation for the equilibrium data. In this paper, the Langmuir and Freundlich isotherm models were adopted. For analysis of data using linear regression, the fitness of the model is often evaluated based on the value of the correlation coefficients (R).

Table III shows the values of the parameters of the two isotherms and the related correlation coefficients. A comparison is also made between two isotherms plotted in Fig. 3, which shows the experimental data points and the two theoretical isotherms plotted on the same graph.

As seen from Table III, the Langmuir model yields a somewhat better fit (R=0.999) than the Freundlich model (R=0.898).

TABLE III
COMPARISON OF EQUILIBRIUM ISOTHERM MODELS

COMMITTEE OF EQUIPMENT TO THE TENT TO THE				
Langmuir				
$Q_0 \text{ (mg g}^1\text{)}$ b (L mg $^{-1}$ )	22.4			
b (L mg <sup>-1</sup> )	1.88			
R	0.999			
Freundlich				
$K_{\rm f}$ [(mg g <sup>-1</sup> )(L mg <sup>-1</sup> ) <sup>1/n</sup> ]	6.13			
n	3.41			
R	0.898			

The fact that the Langmuir isotherm fits the experimental data very well maybe due to homogenous

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distribution of active sites on the ZFAM surface, since the Langmuir equation assumes that the surface is homogeneous.

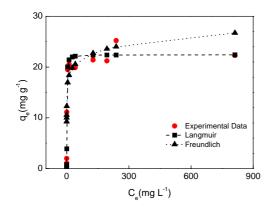


Fig. 3. Equilibrium isotherms for the removal of U(VI) by ZFAM (pH= 3.0; T= 25  $^{\circ}$ C)

## IV. Conclusion

In this study, zeolite from fly ash/iron oxide magnetic nanocomposite (ZFZM) was synthesized and used for the removal of U(VI) from aqueous solutions. The adsorption isotherms and kinetics of U(VI) onto ZFAM was investigated.

The equilibrium of adsorption was suitably described by the Langmuir models with a monolayer adsorption capacity of 22.4 mg g<sup>-1</sup>. The process of adsorption was relatively rapid and was best described by the pseudo-second-order kinetic model. The magnetic properties of ZFAM adsorbent allowed their separation from water by applying a magnetic field, indicating that composite could be used as a promising and effective adsorbent for the removal of uranium(VI) from water. Converting fly ash into zeolites not only alleviates the disposal problem but also converts a waste material into a marketable commodity.

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