

Influence of Humic Acids on Radium Adsorption by Coir Pith in Aqueous Solution (Pengaruh Asid Humik terhadap Penjerapan Radium oleh Sabut Kelapa di dalam Larutan Aqueus)

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ABSTRACT

This study examines the influence of humic acids (HA) on adsorption of radium (Ra) ions onto coir pith (CP) in aqueous solution. The adsorption behaviours of Ra ions onto CP under the influence of HA in aqueous solution were investigated in the series of batch mode adsorption experiments. The effects of various experimental conditions such as pH, contact time, adsorbent dosage and initial concentration of Ra ions have been studied. The results revealed that the presence of HA in aqueous solution enhanced the adsorption of Ra ions onto CP. The adsorption results showed that the percentage of Ra adsorbed was increased with an increase in the pH or alkalinity of aqueous solutions. Time dependence of the batch studies showed that a contact time of one day was sufficient to reach equilibrium. The result also showed that there was no significant difference on the effect of adsorbent dose on adsorption of radium onto CP. It was shown that the equilibrium data could be fitted by Freundlich equation.

Keywords: Adsorption; coir pith; humic acid; radium

ABSTRAK

Kajian ini dijalankan bagi mengkaji pengaruh asid humik (HA) terhadap penjerapan ion radium (Ra) ke atas sabut kelapa (SK) di dalam larutan akueus. Kelakuan penjerapan ion Ra oleh SK dengan pengaruh HA di dalam larutan akueus dijalankan dalam siri eksperimen penjerapan mod kelompok. Kesan pelbagai keadaan eksperimen seperti pH, masa, dos bahan penjerap dan kepekatan ion Ra telah dikaji. Hasil kajian menunjukkan kehadiran HA di dalam larutan akueus menambahkan penjerapan ion Ra ke atas SK. Peratusan Ra terjerap juga meningkat dengan peningkatan pH atau kealkalian larutan akueus. Pergantungan masa dalam kajian kelompok menunjukkan masa penjerapan satu hari memadai bagi mencapai keadaan keseimbangan di samping tiada perbezaan signifikan bagi kesan dos bahan penjerap terhadap penjerapan Ra ke atas SK. Data keseimbangan menunjukkan ia dapat dipadankan dengan persamaan Freundlich.

Kata kunci: Asid humik; penjerapan; radium; sabut kelapa

INTRODUCTION

The interaction of radionuclides with natural organic materials is relatively poorly understood, although these materials may play an important role in controlling the behaviour and mobility of radionuclides in the environment (Choppin 1988). Natural organic materials can be divided into humic acids (HAs), fulvic acids (FAs) and humin. Generally, HAs are natural organic compounds of soil, which result from decomposition of organic matter. HAs are thought to be complex aromatic macromolecules with amino acids, amino sugars, peptides, aliphatic compounds involved in linkages between the aromatic groups (Koezorowska et al. 2002). The main functional groups present in HAs are carboxylic acids, alcohols, phenols, carbonyls, phosphates, sulphates, amides and sulphides (Baek & Yang 2004). Thus, complex structure of HA has provided this compound to form ionic, donor-acceptor interactions (including hydrogen bonding and charges

transfer complexes) and hydrophobic bonding, respectively (Perminova et al. 2000).

HA have attracted great attention because of their complexation ability with metal ions (Omar & Bowen 1982; Tan et al. 2007; Fukurawa & Takahashi 2008) and radionuclides (Wang et al. 2006; Barbot et al. 2007). Due to their high complexing capacity, HAs influence the speciation of metals ions e.g actinides and therefore, the migration and/or immobilisation of these pollutants in the environment (Pompe et al. 2000). Stevenson (1982) reported that humic acids, along with other colloidal materials were strongly affecting a wide range of environmentally important reactions and process. Ibrahim et al. (2008) studied the interaction of thorium with HA and they found that thorium could interact with humic acids in a wide pH range. The effect of HA on sorption-desorption of radiocesium on various sorbents has been studied by Shaban and Mikulaj (1996). Their study showed

that the presence of humic acids resulted in an enhanced desorption of cesium from zeolite. Wang et al. (2006) have investigated the influence of pH on sorption/complexation of Eu (III) at HA-coated alumina surfaces. Their result showed that the presence of HA affects the sorption of Eu (III) on alumina significantly, which gave the positive effect at low pH and the negative effect at high pH values. Thus, the presence of HAs significantly influences the adsorption of radionuclides onto various adsorbents.

Adsorption process is one of the common methods to study interaction behaviour between HAs and radionuclides. Most of the previous papers (Chen et al. 2007; Moulin & Moulin 1995; Shaban & Mikulaj 1996; Reiller et al. 2003; Wang et al. 2006; Tan et al. 2007) were focused on the complexation and sorption behaviour of HAs with a mono, triv-, hexa- and pentavalent element such as cesium, americium, uranium, thorium, and europium. Therefore, in this paper we focus our study on the influence of HAs on the adsorption of divalent element i.e. radium onto coconut coir pith (CP). The series of adsorption experiments in batch method were conducted to evaluate the adsorption of radium under the influence of HAs in various variables such as pH, contact time, adsorbent dose and the concentration of radium ions.

MATERIALS AND METHODS

CHEMICALS AND INSTRUMENTS

The chemicals used were of analytical grade. ^{226}Ra stock solution was obtained from Isotope Products Laboratories (Eckert & Ziegler Company). A stock solution of 245.48 Bq of ^{226}Ra (1.4×10^{-6} mM) was prepared by diluting 1.7 g of ^{226}Ra stock solution (361.3 kBq) (2.2×10^{-3} mM) with 250 ml of distilled water in a 250-ml volumetric flask. A series of ^{226}Ra working solutions with the concentrations of 2 to 16 Bqml⁻¹ was freshly prepared by appropriate dilution of the stock solution prior to their usage. pH was measured using a pH meter (HANNA instrument Model pH 211) and a flask shaker machine (Stuart model SF1, UK) was used for batch adsorption studies. A Canberra n-type high purity germanium (HpGe) gamma spectrometer (30% relative efficiency, resolution of 1.9 keV at 1.33 MeV) was employed for the measurement of ^{226}Ra concentration in solution. The system was calibrated in a similar manner as described by Omar et al. (2004) using a multinuclide standard solution source. The ^{226}Ra activity was determined through its 186.2 keV gamma energy peak.

ISOLATION OF HUMIC ACIDS

HAs samples used in this study were isolated from peat soils, which were collected from Kanchong Darat, Banting, Selangor Darul Ehsan. The peat soils were dried at room temperature, ground and passed through a 2.0 mm sieve after the removal of plant roots. HAs were extracted from the peat soils using the method recommended by International Humic Substances Society (IHSS) with minor

modification (Ibrahim et al. 2008). Then, HAs stock solution with 0.2 mg/ml was prepared by dissolving 0.1g HAs in 0.01M NaOH in a 500 ml volumetric flask.

COIR PITH PREPARATION

Coir pith (CP) was prepared from coconut husk obtained from Bagan Datoh, Perak Darul Ridzuan. It was ground and sieved by USA Standard Sieve No. 10, 14, 18 and 35 (corresponding to 2000, 1410, 1000, 500 μm , respectively) with a sieve shaker (Fritsch model Analysette 3 Spartan, Germany). The CP was suspended in 500 ml of 5% NaOH with constant stirring for about 24 h followed by a thorough washing with distilled water. It was then filtered and oven-dried at 105°C.

ELEMENTAL AND FUNCTIONAL GROUP COMPOSITION

Some properties of HA and CP were investigated. Elemental analysis was performed using a CHNS Analyser (model CHNS-932, USA). The surface morphology was examined by a scanning electron microscope (SEM) (model FEI 400). The analysis on the functional groups of HA and CP was performed using Fourier transform infrared spectrophotometer (FTIR) (model Spectrum 2000/L183, USA) in the range 400 - 4000 cm^{-1} . Total acidic groups, carboxyls and phenol-hydroxyls contents of HAs were measured in triplicate by barium hydroxide and calcium acetate method as described in detail by Stevenson (1982). The point of zero charge (pH_{pzc}) of CP was determined by using potentiometric mass titration (PMT) method as described by Fiol (2008). The equilibrium pH values were plotted as a function of acid volume added to obtain the potentiometric curves. pH_{pzc} was identified as the intersection point of the potentiometric curve with the blank curve.

ADSORPTION EXPERIMENTS

The adsorption of Ra^{2+} on CP in the presence of HAs was studied by batch technique. All adsorption experiments were conducted in triplicate and the results were reported as average. Batch experiments were conducted to determine the effect of pH, contact time, adsorbent dose and initial concentration of Ra ions. The effect of pH on the adsorption of radium was studied by adding 40 ml of a fixed concentration of ^{226}Ra working solution to 10 ml of fixed concentration of HAs solutions in 50 ml centrifuge tubes containing 0.2 g of CP, adjusting the pH to 3, 5, 7, 9 and 11 with HCl and/or NaOH solutions and the final volume was made up to 50 ml. The centrifuge tubes were sealed with screw caps. The mixtures were then shaken using a flask shaker machine at 300 oscmin^{-1} for 24 hours at room temperature. At the end of the adsorption period, the mixtures were filtered and the activity concentration of ^{226}Ra in the filtrate was determined by a gamma-ray spectrometer. The adsorbed ^{226}Ra was obtained from the difference between the initial and the final ^{226}Ra activity concentrations.

The percentage of radium adsorption was determined by following equation:

$$\text{Radium adsorption (\%)} = \left(\frac{C_o - C_f}{C_o} \right) \times 100 \quad (1)$$

where C_o and C_f are the initial and final activity concentration of aqueous phase (Bqml⁻¹), respectively.

The distribution coefficient, K_d for this batch adsorption experiment was determined by following equation:

$$K_d \text{ (ml/g)} = \frac{(C_o - C_f) V \text{ (ml)}}{C_i \text{ m (g)}} \quad (2)$$

where V is the volume of solution (ml) and m is the weight of the adsorbent (g).

In the second part of this study, the effect of contact time in the intervals of between 1 to 7 days were studied on a batch of Ra²⁺ solutions with fixed concentration and pH. At the end of predetermined time intervals, the adsorbent was removed by filtration, and the concentration of radium ions in the filtrate was determined by gamma-ray spectrometer. Isotherm studies were conducted with 0.2 g adsorbent dose and varying initial concentration of Ra ions (2-16 Bqml⁻¹). The adsorption capacity (q_e) was determined by the following equation:

$$q_e = \frac{(C_o - C_f) V}{m} \quad (3)$$

where C_o and C_f are the initial and final activity concentration of aqueous phase (Bqml⁻¹), respectively, V the volume of solution and m is the mass of adsorbent (g).

The effect of adsorbent dosage on Ra ions was conducted by the same batch adsorption procedure described above by fixing the pH and contact time but varying the adsorbent doses between 0.2 g - 1.0 g.

RESULTS AND DISCUSSION

CHARACTERIZATION OF HAS AND COIR PITH

The characteristics of HA and CP are shown in Table 1. Elemental analysis shows that HA and CP has high carbon content. It was also found that C, O and N in HAs extracted from peat soils in this study were lower than reported by Husni et al. (1996). However, our results were still within the range for the tropical peat as reported by FAO (1988). As shown in Table 1, the majority of total acidic groups for HAs are carboxyls and these groups could release H⁺ in aqueous solutions (Li et al. (2008). Besides, the amount of carboxylic groups the most important group is functional group for protolysis and complexation behavior of humic acid (Pompe et al. 2000). SEM images in Figure 1 and 2 shows the morphological of HA and coir pith (magnification of 200x). It could be seen that the HA materials varied in size and were irregular in shape. The SEM image for CP shows that the CP surface consists of close thin-walled ribbon shape cells and porous pith tissue. Figure 3 shows the SEM image of CP after the Ra adsorption (magnification of 200x) under the presence of HAs in aqueous solution. From the image, it could be seen that there were slightly changed of the CP surfaces after the adsorption process. The structure of porous pith tissue was disrupted and tended to coagulate after Ra adsorption.

The FTIR spectra of HA, CP and Coir Pith-Humic Acids-Radium (CPHARa) are shown in Figure 4. The FTIR spectrum of HA revealed the main absorption bands are at 3362.8 cm⁻¹ (H-bonded OH groups), 2920.3 cm⁻¹ (aliphatic C-H stretching), 1705.6 cm⁻¹ (C=O stretching of COOH and ketonic C=O), 1610 cm⁻¹ (aromatic C=C and H bonded C=O) and 1250 cm⁻¹ (C-O stretching and O-H deformation of COOH groups). There are small bands at 1507.0 cm⁻¹ (aromatic C=C), 1407.8 cm⁻¹ (C-H deformation of CH₂), 1365.2 cm⁻¹ (O-H deformation,

TABLE 1. Characteristics of humic acid and coir pith

	Parameters	Value
Humic acid	Elemental composition (%)	30.1
	C	4.6
	N	1.0
	H	3.6
	S	5.87
	Total acidity (meqg-1)	3.53
	COOH	2.34
	OH	
Coir Pith	Elemental composition (%)	
	C	55.2
	N	5.8
	H	1.5
	S	0.36
	Point of zero charge (pH _{pzc})	6.3
	pH _{pzc}	60.7
	Activity concentration (Bqkg ⁻¹)	<9.72
	226Ra	
	228Ra	

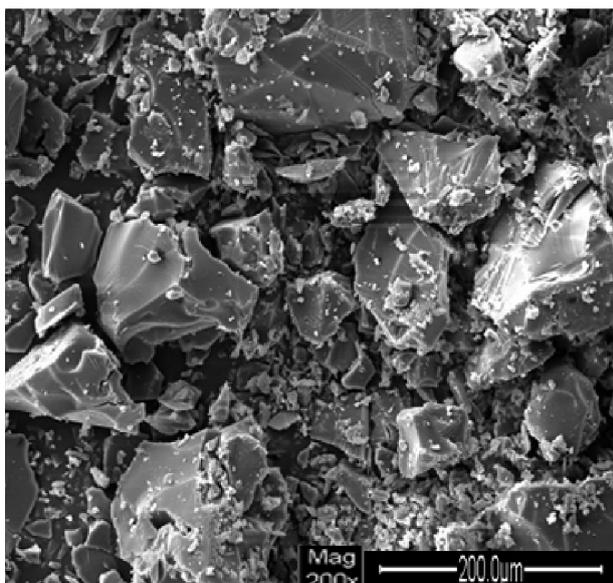


FIGURE 1. Scanning electron micrograph of shape and particle size of humic acid

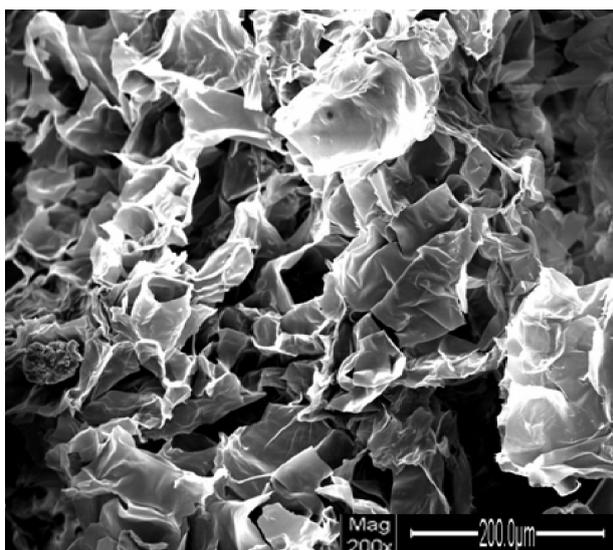


FIGURE 3. Scanning electron micrograph of coir pith after Ra adsorption under the presence of HAs at pH9

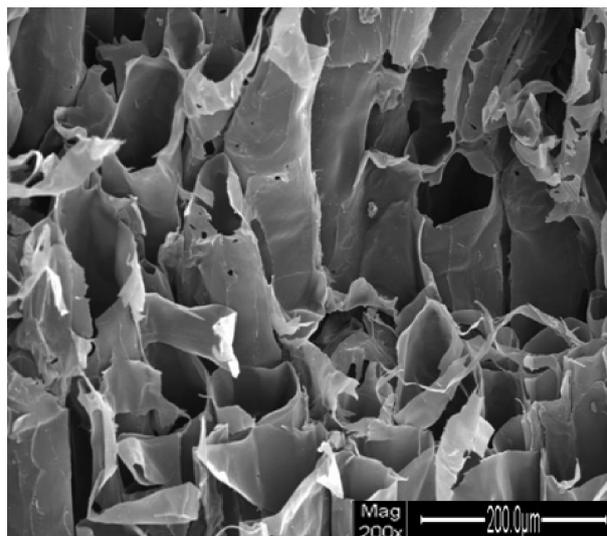


FIGURE 2. Scanning electron micrograph of raw coir pith

CH_3 bending, or C-O stretching). The FTIR spectrum of CP before adsorption reveals a broad peak around 3334.5 cm^{-1} , which can be attributed to the O-H groups from cellulose structure (Anirudhan & Unnithan 2007). The peak observed at 2914.85 cm^{-1} corresponds to C-H stretching. The spectra at $1800 - 1000 \text{ cm}^{-1}$ were fingerprint regions (Suksabye et al. 2007). These peaks are corresponding to the carbonyl stretching groups (1726.9 cm^{-1}), C=C stretching of aromatic ring vibration (1605.3 cm^{-1} , 1509.2 cm^{-1}), methoxy groups (O-CH_3 , 1429 cm^{-1}) from lignin structure of CP (Khan et al. 2004), O-H deformation of phenolic group (1365.2 cm^{-1}) and C-OH stretching (1262.9 cm^{-1} , 145.3 cm^{-1} , 1035 cm^{-1}). In the FTIR spectrum of CPHARa, it is clear that an OH stretching vibrational band shows a frequency shift to lower wave number (3341.5 to 3329.9 cm^{-1}) after the adsorption process. The shift to lower wave number indicated that the interaction of -OH groups was greater after Ra adsorption in the presence of HA in aqueous solutions. It was also observed that there were a small shift of wave number for an aromatic C=C and C-OH groups. The shift of wave numbers might indicated that there were a chemical interaction between the adsorbent, HA and Ra ions.

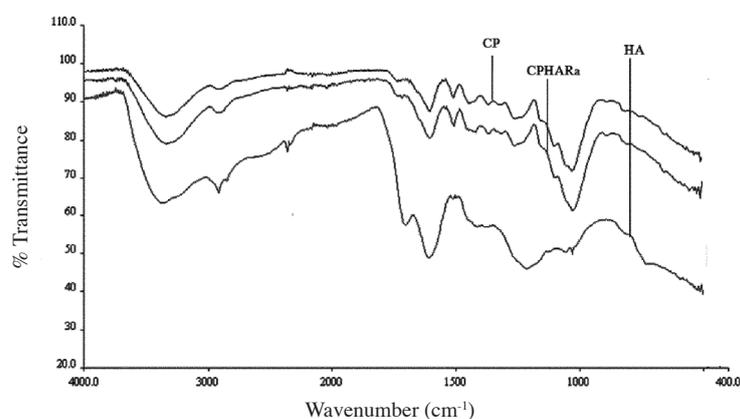


FIGURE 4. FTIR spectra of HA, CP (before adsorption) and CPHARa (after adsorption at pH 9)

EFFECT OF pH

The pH of a solution plays an important role in adsorption of divalent ions onto adsorbents. As it can be seen from Figure 5, in the presence of HAs, Ra adsorption increased with increasing pH of the solution and reaching a maximum (about 96%) at pH range 7.0 - 11.0. At low pH values, the active sites of CP surfaces are less available for Ra ions due to protonation. Hence, the electrostatic repulsion occurred between Ra^{2+} ions and positively charged CP surface and resulted in the decreased percentage of Ra adsorption. However, the negative charged of HAs (carboxylic group and phenolic group) in the aqueous solution could be easily adsorbed by positively charged CP surfaces. Particularly, HAs have a macromolecular structure (Chen et al. 2007) and only a small fraction of surface-adsorbed carboxylic and phenolic groups of HAs directly interact with CP surface sites. The remaining group is free to interact with Ra ions. Thus, the adsorbed HA on CP surface might provide additional adsorption sites for Ra ions and therefore might enhance the adsorption of Ra ions at low pH values. It was observed that at pH 5 about 75% of Ra adsorption was achieved.

At high pH values, an increase of Ra ions adsorption might reflect the presence of fewer H^+ ions in aqueous solutions that could compete with Ra^{2+} ions for available adsorption sites. The effect of pH on the adsorption of Ra ions onto CP under the presence of HAs can be interpreted with the help of the structure and the surface charge of adsorbent. Since the pH_{pzc} of the CP was found to be 6.3 (Figure 6), the surface was expected to be negatively charged at $pH > 6.3$. This is normally beneficial for the adsorption of positively charged cationic species like radium. Thus, at high pH values, the degree of protonation of CP surface reduced to almost zero at 6.3 (pH_{pzc}) resulting in a gradual increase in adsorption. Generally, HAs are a combination of varying functionality from non-polar poly-methylene chain to highly polar carboxylic acid fraction (Ghosh et al. 2009). Since carboxylic groups start to dissociate into $HA-COO^-$ at pH 4 - 6 (Paajanen et al. 1997), it is believed that the non-polar sites of HAs might be adsorbed to negatively charged CP surface and therefore, provide additional surface for Ra adsorption. On the whole, the adsorption of Ra onto CP under the presence of HAs was more favourable at neutral to alkaline

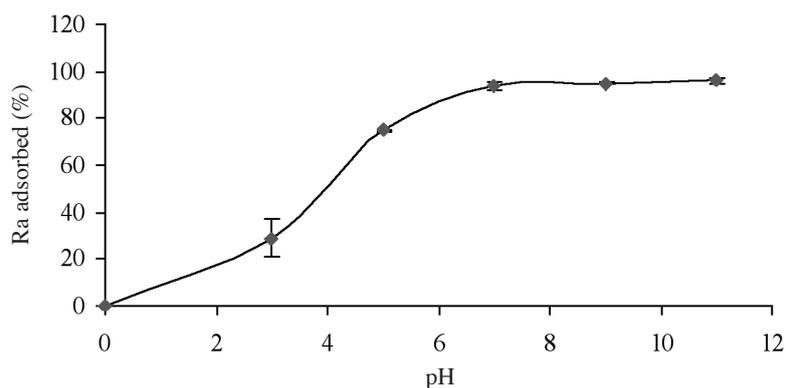


FIGURE 5. Effect of pH on the adsorption of Ra ions onto CP in the presence HA in aqueous solution

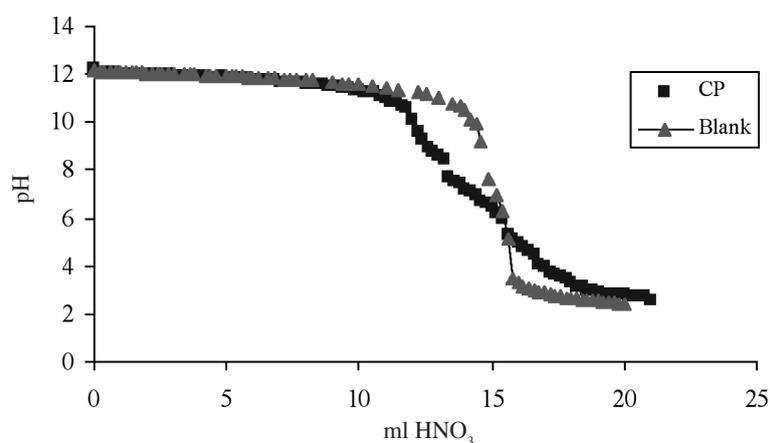


FIGURE 6. Experimental potentiometric mass titration curves for the determination of pH_{pzc} of coconut coir pith

condition. In this study, the maximum adsorption value for CP was obtained at $\text{pH} > \text{pH}_{\text{pzc}}$. Therefore, it is clear that electrostatic attraction (coulombic) between negatively charged CP surface and Ra ions could take place and thus influences the adsorption process of Ra onto CP surfaces under the presence of HAs.

The distribution coefficient (K_d) is very important in estimating the adsorption potential of dissolved contaminants by adsorbent materials. Figure 7 shows the effect of pH on K_d for Ra adsorption onto CP under the influence of HA. The result showed that K_d was strongly depending on the pH of aqueous solution ($R^2=0.9527$). The maximum K_d value occurs at pH 11 (i.e 5544.44 mlg^{-1}). Thus, this K_d value indicates that maximum concentration of Ra ions that could be adsorbed onto CP surfaces under the influence of HA.

EFFECT OF CONTACT TIME

Figure 8 shows the percentage of Ra adsorbed as a function of time at pH 9. It was observed that the percentage of Ra adsorption increased with time and slowly reaching saturation after one day. Preliminary studies indicated that contact times of less than 24 hours were not enough to attain the equilibrium time. Thus, in this study the effect of

contact times of longer than one day on the adsorption of Ra ions was investigated. As a result, the time of one day was considered sufficient for a significant removal of Ra ions and therefore was chosen for all further experiments.

EFFECT OF ADSORBENT DOSAGE

Figure 9 shows the effect of adsorbent dosage from 0.2 g to 1.0 g at pH 9. It was found that there was no improvement in percentage of adsorption by increasing further adsorbent dosage of more than 0.2 g. It could be suggested that the presence of HA in aqueous solution might enhance the Ra ions adsorption onto CP and allowed the adsorption process to reach equilibrium faster. The data of CP dosage subjected to analysis of variance and means separation using LSD/Duncan test. The results were not significantly different at $P \leq 0.05$ indicating that Ra ions adsorption by CP from 0.2 g to 1.0 g was not different. This indicated that under the presence of HA in aqueous solution the optimum dosage of Ra ions adsorption was 0.2 g at 96 % of Ra ions adsorption.

ADSORPTION ISOTHERM

Adsorption isotherm was used to describe the equilibrium established between adsorbed Ra ions on CP (q_e) and Ra

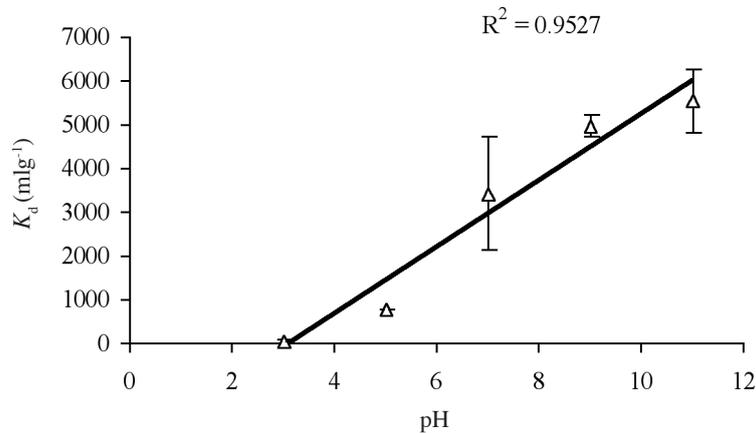


FIGURE 7. Effect of pH on the distribution coefficient (K_d) on the adsorption of Ra ions onto CP under the presence of HAs

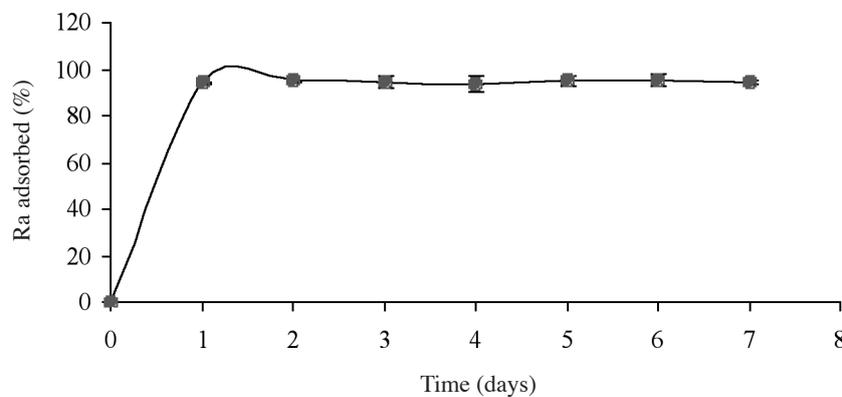


FIGURE 8. Effect of contact time on the adsorption of Ra ions onto CP under the influence of HAs

ions remaining in the solution (C_e). The Langmuir and Freundlich equations are commonly used for describing adsorption equilibrium for water and waste water treatment applications (Parab et al. 2005). In this study, the data obtained from adsorption experiments was fitted to Freundlich isotherm (Figure 10). The empirical equation based on adsorption on heterogeneous is as follows:

$$q_e = K_f C_e^{1/n} \quad (4)$$

where C_e is the equilibrium concentration (Bqml^{-1}), q_e is the amount adsorbed (Bqg^{-1}) and K_f and $1/n$ are constants indicative of adsorption capacity and intensity, respectively. Equation (4) can be linearised in logarithmic form as follows:

$$\log q_e = \log K_f + 1/n \log C_e \quad (5)$$

From the slopes and intercepts of the straight lines parameters $1/n$ and K_f were calculated and the values obtained were 1.061 (Bqml^{-1}) and 3586 (Bqg^{-1}), respectively. The Freundlich isotherm equation provides the best fit for the data for the whole range of Ra concentrations. According to Treybal (1980), it has been shown that n values between 1 and 10 represent beneficial adsorption.

CONCLUSION

The adsorption of Ra ions onto CP under the influence of HAs was strongly dependent on pH and contact time but a dosage of adsorbent did not have a significant effect on the adsorption process. The adsorption of Ra ions was increased with increasing pH, indicating that Ra ions are adsorbed onto CP surfaces mainly through the ion-exchange mechanism. The FTIR and SEM characterization of the adsorbents has shown a clear difference in the native and Ra ions loaded adsorbents under the presence of HA in aqueous solution. The K_d value depends upon the pH of solution. The results obtained in this study showed good fit to Freundlich isotherm. The presence of HAs in aqueous solution was shown to have a significant effect on adsorption process by enhancing the Ra adsorption at low and high pH values and thus, allowing the adsorption process to reach equilibrium faster.

ACKNOWLEDGEMENTS

The authors thank the Ministry of Science, Technology & Innovation, (MOST) for the funding under the Sciencefund program (Project No. 03-03-01-SF0027) and Nuclear Malaysia for providing laboratory facilities for this work.

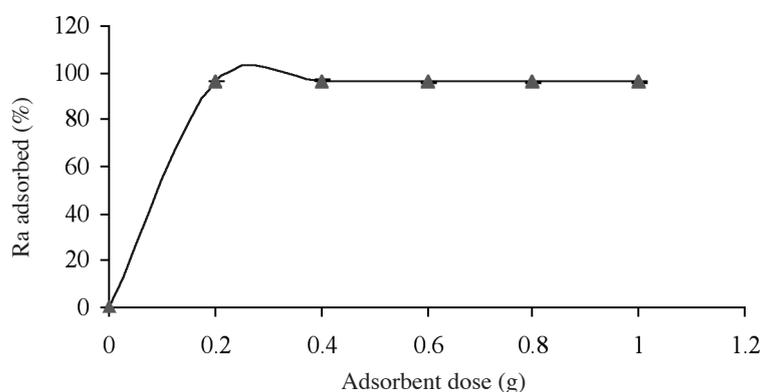


FIGURE 9. Effect of adsorbent dose on Ra ions adsorption under the influence of HAs

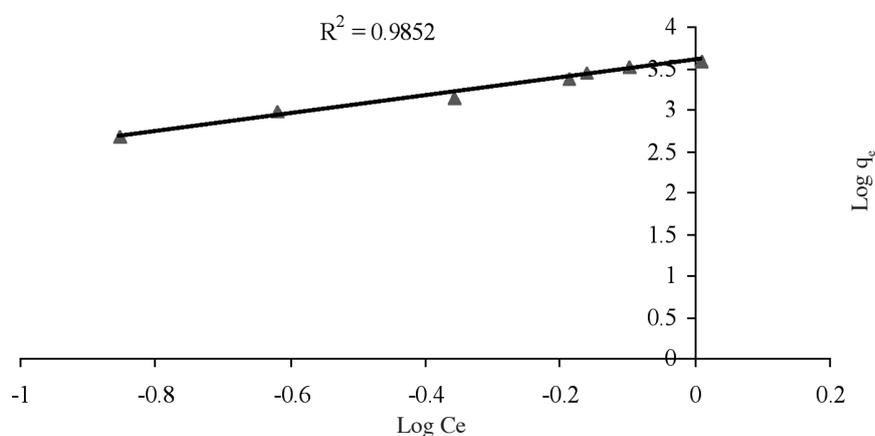


FIGURE 10. Freundlich isotherm for adsorption of Ra onto CP in the aqueous solution under the presence of HAs

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Received: 14 March 2009

Accepted: 19 June 2009