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EFFECT OF Al^{3+}, Ca^{2+}, Cu^{2+} AND Fe^{2+} ON REMOVAL OF RADIUM-226 FROM RADIUM-CONTAMINATED SOIL USING HUMIC ACID

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ABSTRACT

Effect of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} on the removal of radium-226 from radium-contaminated soil using humic acid extracted from a Malaysian peat soil was investigated using batch washing method. The concentration of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} ranged from 0 to 100 ppm. The radioactivity concentration of radium-226 was determined by gamma spectrometer. The removal of radium-226 was enhanced in the presence of Al^{3+} with concentration between 20 – 60 ppm. Meanwhile, higher concentration of 80 – 100 ppm did not lead to further increase in the removal of radium-226. The removal of radium-226 was decreased in the presence of Ca^{2+} and Cu^{2+}. In the presence of Fe^{2+} with concentration between 20 – 60 ppm, decrease in the removal of radium-226 was also observed. Nevertheless, at concentration between 80 – 100 ppm, the presence of Fe^{2+} led to increase in the removal of radium-226.

Keywords: Al^{3+}; Ca^{2+}; Cu^{2+}; Fe^{2+}; Radium-226; Soil; Humic Acid

ABSTRAK

Kesan Al^{3+}, Ca^{2+}, Cu^{2+} dan Fe^{2+} ke atas penyingkiran radium-226 daripada tanih tercemar radium menggunakan asid humik yang diekstrak daripada tanih gambut Malaysia melalui kaedah pembasuhan berkelompok telah dikaji. Kepekatan Al^{3+}, Ca^{2+}, Cu^{2+} dan Fe^{2+} adalah dalam julat antara 0 sehingga 100 ppm. Kepekatan radioaktiviti radium-226 ditentukan menggunakan alat spektrometer gama. Penyingkiran radium-226 ditingkatkan dalam kehadiran Al^{3+} berkepekatan antara 20 – 60 ppm. Sementara itu, kepekatan yang lebih tinggi iaitu antara 80 – 100 ppm tidak lagi menyebabkan kenaikan dalam penyingkiran radium-226. Penyingkiran radium-226 menurun dalam kehadiran Ca^{2+} dan Cu^{2+}. Dalam kehadiran Fe^{2+} berkepekatan antara 20 – 60 ppm, penurunan dalam penyingkiran radium-226 diperhatikan. Walau bagaimanapun, pada kepekatan antara 80 – 100 ppm, kehadiran Fe^{2+} menyebabkan penurunan dalam penyingkiran radium-226.

Kata kunci: Al^{3+}; Ca^{2+}; Cu^{2+}; Fe^{2+}; Radium-226; Tanih; Asid humik

INTRODUCTION

Contamination of soil with Ra arises from past activities that involve radioluminescence dial painting (Murphy et al., 1980; Eisenbud and Gesell, 1997) as well as mining and processing of U (Snodgrass, 1990), monazite and phosphogypsum (Paul and Pillai, 1990). Such contamination can lead to detrimental effects cause by both Ra and radon if not manage properly. Besides that, contamination of soil also results in wastage of sites for utilization and development purposes. U.S. Department of Energy has reported the shortage of sites available for development due to contamination of land with radionuclides and heavy metals (Peters, 1999). Soil is a complex matrix consistently interacting with water and air (Schwarzenbach et al., 2003). Thus contamination of soil can lead to contamination of water and air (Fent, 2003) and eventually resulted in prolong environmental contamination (Conte et al., 2005). Therefore, decontamination of the contaminated soil is necessary to eliminate the detrimental effects of Ra and to return the treated soil to the environment for reutilization.

Decontamination and treatment of contaminated soil by soil washing method using various extractants such as acids, bases, chelating agents, surfactants, reducing agents, oxidizing agents, solvents, water and alcohol has been extensively explored (Peters, 1999). In comparison to other methods, soil washing method is more efficient and cost-effective (Kuhlman and Greenfield, 1999). Decontamination and treatment of Ra-contaminated soil using synthetic extractants namely EDTA and HF-HNO_3 (Greeman et al., 1999), sym-Di[4(5)-tert-butylbenzo]-16-crown-5-oxyacetic acid (DTBDB16C5-OAcH), NACl/HCl and CaCl/NaCl salt solution (Lan et al., 2000) and HNO_3 and NH_4NO_2 (Sklifasovskaya et al., 2009) has been carried out. Nevertheless, due to the toxicity of these synthetic extractants, application of non-toxic natural extractants is anticipated.

Humic acid (HA), a natural organic substance extracted from humic substances, has been found to interact with metal ions (Bowen et al., 1979; Omar and Bowen, 1982; Ghabbour et al., 2001) and radionuclides (Marquardt, 2000; Moulin, 2005) in particular Ra (Hansen and Huntington, 1969; Kaplan, 2001; Ibrahim et al., 2009, Laili et al., 2010; Phillip et al., 2011; Phillip et al., 2012) owing to the presence of functional groups especially carboxyl (COOH), phenolic-OH and carbonyl groups (Stevenson, 1982). Conte et al. (2005) has shown that HA is as effective as synthetic surfactants namely sodium dodecyl sulfate (SDS) and Triton X-100 in treating contaminated soil.

The removal of metal and radionuclide species from soil depends on the retention and mobility of these species in the soil matrix. The metal and radionuclide species present in the soil through numerous mechanisms such as dissolution in soil solution, occupation of exchange sites on inorganic soil constituents, adsorption on inorganic soil constituents, association with insoluble soil organic matter, precipitation as pure or mixed solids and complexation with solution-phase organic or inorganic species (Wang et al., 1997). Therefore, the removal efficiency for these metal and radionuclide species depends on the acidity, ionic strength, redox potential and complexes formation (Moutsatsou et al., 2006) that can be altered in the presence of other metal ions. The retention and mobility of the metal and radionuclide species may be influenced by the presence of other metal ions particularly those with similar charge as the metal and radionuclide species especially since most of the species are adsorbed by ion exchange mechanism (Wang et al., 1997).

The objective of this study is to investigate the effect of other metal ions such as Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} on the removal of ^{226}Ra from Ra-contaminated soil using HA. The concentration of the metal ions ranged from 0 – 100 ppm.

MATERIALS AND METHODS

HA used was extracted from peat soil obtained from Bachok, Kelantan, Malaysia. Meanwhile, Ra-contaminated soil was obtained from a former compass dial painting facility site in Malaysia. The Ra-contaminated soil sample had been excavated from the site and stored at the radioactive waste storage facility in Nuclear Malaysia since year 2000.

Ra-226 standard solutions were obtained from Isotope Products Laboratories (an Eckert & Ziegler Company). Other reagents and chemicals used were of analytical grade from R&M Chemicals or Sigma Aldrich.

Extraction of HA from peat soil

HA was extracted from peat soil according to the acid-base extraction method described by International Humic Substances Society (IHSS). HA stock solutions were prepared by dissolving the extracted solid HA in diluted sodium hydroxide (NaOH) solutions. HA working solutions were prepared by diluting the stock solutions also in diluted NaOH.

Preparation of Ra-contaminated soil sample

The Ra-contaminated soil sample was air-dried at room temperature in a fume hood prior to manual grinding. The ground soil sample was then sieved using a 2 mm sieve and finally stored in a sealed container for experimental purposes.

Determination of ^{226}Ra radioactivity concentration

^{226}Ra radioactivity concentration was analysed using a gamma spectrometer (Oxford Instruments Inc., 30% relative efficiency and 1.9 keV at 1.33 MeV resolution with a CANBERRA n-type hyper-germanium detector (HPGe)).

Effect of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+}

Al (III), Ca (II), Cu (II) and Fe (II) salt of certain weight (to achieve metal ion concentration of 20, 40, 60, 80 and 100 ppm) was agitated using an orbital shaker with 1 g of Ra-contaminated soil sample and 20 mL HA solution of concentration 100 ppm and pH 7 in 100 mL conical flask at 100 rpm for 24 h at room temperature. pH of the soil suspension was measured. The soil suspension was then centrifuged at 4000 rpm for 10 min and the supernatant was collected by decantation. The collected supernatant was immediately sealed in a plastic container and was left to equilibrate for at least 21 days before analysis with gamma spectrometer. Precision was established by preparing triplicates for each test.

Removal efficiency of Ra-226 was calculated based on the equation

$$\text{Removal efficiency, \%} = \frac{\text{Ao} - \text{A}}{\text{Ao}} \times 100\%$$

A_o = Initial Ra-226 activity concentration in Ra-contaminated soil sample, Bq/kg

A = Final Ra-226 activity concentration in Ra-contaminated soil sample after washing, Bq/kg

RESULTS AND DISCUSSION

Our findings had shown that the removal of ^{226}Ra using HA of concentration of 100 ppm and pH 7 by batch washing method was altered in the presence of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} depending on types and concentration of the interfering metal ions. Fig. 1 shows the effect of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} of concentration between 0 – 100 ppm on the removal of ^{226}Ra from the Ra-contaminated soil studied.

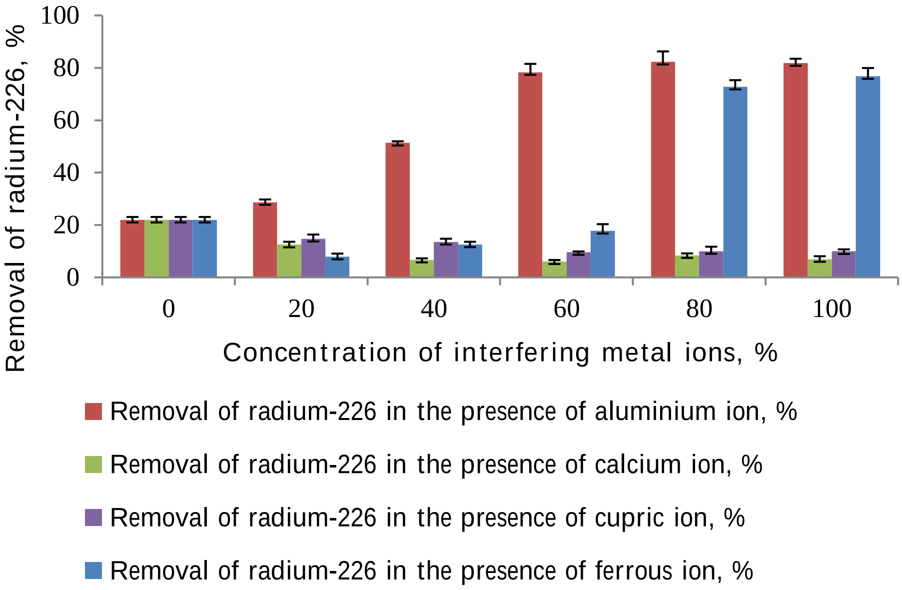

Fig. 1. Effect of Al^{3+}, Ca^{2+}, Cu^{2+} and Fe^{2+} of concentration between 0 – 100 ppm on the removal of ^{226}Ra from Ra-contaminated soil using HA (HA concentration = 100 ppm, HA pH = 7, 24 h, liquid/solid = 20 mL/g)

Initial metal content analysis had detected the presence of Al in the Ra-contaminated soil studied (Phillip et al., 2012). In the presence of additional Al^{3+}, the final pH of the soil-HA suspension was found to be around 5. The removal of ^{226}Ra increased with the concentration of additional Al^{3+} (20 – 80 ppm). The increase observed was approximately 7 – 60 %. It was anticipated that the increase in the removal of ^{226}Ra was due to the interaction between ^{226}Ra species and Al^{3+}. Skeen and Sumner (1967) had described the exchangeability of Al in soil with other alkaline earth metal cations. Therefore, in this study, it was assumed that Al could be exchangeable with Ra. Besides that, as an analog to Ba (Beek et al., 2009), Ra was capable in competing with Al to secure exchange sites on soil matrix (Carbonell et al., 1999). Thus, in the presence of additional Al^{3+} with increasing concentration, liberation of ^{226}Ra species from soil matrix also increased due to occupation of Al^{3+} on the soil matrix replacing ^{226}Ra. However, the removal of ^{226}Ra remained constant at Al^{3+} concentration higher than 80 ppm, At this concentration, all of the exchangeable ^{226}Ra species were liberated and any increase in Al^{3+} concentration would not lead to any change in the removal of ^{226}Ra. Our findings also revealed that in the presence of additional Al^{3+}, HA did not play an important role in the removal of ^{226}Ra. Furthermore, HA molecules preferred binding with Al than ^{226}Ra species as humic substances were strongly complexed with Al in pH range between 4 – 5 (Elkins and Nelson, 2002).

Ca^{2+} was not detected in the Ra-contaminated soil studied. In the presence of Ca^{2+}, the final pH of the soil-HA suspension remained at neutral pH. The addition of Ca^{2+} led to decrease in the removal of ^{226}Ra by HA indicating competition between Ca^{2+} and ^{226}Ra species to bind with HA. Clark and Lucas (1998) had mentioned that metal ions such as Ca could bind to the negatively charged carboxylic groups of HA molecules. In the presence of Ca^{2+}, the removal of ^{226}Ra was highest at Ca^{2+} of concentration 20 ppm. However, in the presence of Ca^{2+} of concentration between 40 – 100 ppm, the removal of ^{226}Ra decreased and remained almost constant. Ca and Ra were both alkaline earth metals showing similar chemical properties. Therefore, Ca could serve as a strong competitor to Ra. However, the constant removal of ^{226}Ra suggested that optimum competition was reached and increase in Ca^{2+} concentration would not lead to further removal of ^{226}Ra. It could also indicate that the binding of Ca^{2+} with HA molecules was site specific.

Cu^{2+} was also not detected in the Ra-contaminated soil studied. In the presence of Cu^{2+}, the final pH of the soil-HA suspension remained at pH around 7. In the presence of Cu^{2+}, decrease in the removal of ^{226}Ra was observed thus suggesting the competing effect of Cu^{2+} on the removal of ^{226}Ra by HA. Gondar et al. (2006) had described

Cu (II) binding by peat humic acids. Cu^{2+} was bound to HA molecules by adsorption onto HA colloid and complexation with functional groups (Pandey et al., 2000) in particular carboxyl groups (Gardea-Torresdey et al., 1996). Therefore, the competition between Cu^{2+} and ^{226}Ra species to bind with HA molecules had resulted in the decrease of the removal of ^{226}Ra. In the presence of Cu^{2+} of concentration between 20 – 40 ppm, the removal of ^{226}Ra obtained was approximately 14 – 15 %. In the presence of Cu^{2+} of higher concentration between 60 – 100 ppm, the removal of ^{226}Ra decreased slightly from the previous concentration of 20 – 40 ppm with the removal efficiency maintained at 10 %, thus suggesting the optimum competition and the site specific nature of Cu^{2+} binding with HA molecules.

The presence of Fe had earlier been detected in the Ra-contaminated soil sample. In the presence of additional Fe^{2+}, pH of the soil-HA suspension was shifted to around 5. As shown in Fig. 1, decrease in the removal of ^{226}Ra was observed in the presence of Fe^{2+} of concentration between 20 – 60 ppm probably due to competition between ^{226}Ra and Fe^{2+} to interact with HA. Generally, HA molecules formed both soluble and insoluble complexes with metal ions (Weber, 1983). Tipping (2002) had mentioned that natural organic matter (NOM) could form dissolved complexes with Fe^{2+} and Fe^{3+}. Mak and Lo (2011) had specifically described the complexation of HA with Fe^{2+} to form Fe-humate complexes. Meanwhile, Park and Yoon (2009) had reported the effect of Fe species on the removal of HA through the formation of HA-Fe chelate complexes. Therefore, in this study, it was anticipated that the added ferrous (Fe (II)) salt could lead to the formation of HA-Fe complexes that eventually could reduce the amount of ^{226}Ra being removed by HA. The decrease in the removal of ^{226}Ra could also be explained by Fe^{3+} that was also present in the soil-HA suspension due to oxidation of Fe^{2+} (Tsezos et al., 1986). Owing to its low solubility, fine iron oxyhydroxides (FeOOH) precipitates could be formed (Tsezos et al., 1986). These fine FeOOH precipitates could inhibit the adsorption of U (Tsezos, 1983) and Ra (Tsezos et al., 1986) onto microbial biomass as they could coat the microbial cell surfaces thus hindered the accessibility of U and Ra onto the microbial biomass. Therefore, in this study, similar effect could be anticipated for binding of HA molecules with ^{226}Ra species as FeOOH precipitates formed could coat the soil particles surface and eventually hampered the interaction between HA molecules and ^{226}Ra species. As a consequence, reduction in the removal of ^{226}Ra was observed. Nevertheless, the decrease in the removal of ^{226}Ra observed was only about 3 – 14 %. Park and Yoon (2009) also reported that in weak acidic condition (pH 4.5 – 6.5), HA removal was not significant when Fe (II) salt was added. In weak acidic condition, the interaction of HA with Fe (II) salt was mainly governed by charge neutralization and adsorption mechanisms as HA had more negatively charged sites (Park and Yoon, 2009) and Fe (II) salt existed as lower positively charged species such as $Fe(OH)^{+}$, $Fe(OH)_2$, $FeOH^{2+}$, $Fe(OH)^{2+}$ and $Fe_2(OH)^{4+}$ (Weber, 1983). The low removal of HA was mostly due to the low positive charge of hydrolyzed Fe species that was not sufficient to neutralize the negative charge of HA molecules to form HA-Fe complex (Park and Yoon, 2009). Even though the removal of ^{226}Ra decreased in the presence of Fe^{2+}, nevertheless the removal observed was found to increase with Fe^{2+} concentration between 20 – 60 ppm. In the presence of Fe^{2+} of concentration between 80 – 100 ppm, huge increase of approximately 51 – 55 % was observed in the removal of ^{226}Ra. It was assumed that the interaction between HA and ^{226}Ra species no longer served as an important mechanism that led to the removal of ^{226}Ra. The pH of the soil-HA suspension in the presence of Fe^{2+} of concentration between 80 – 100 ppm was slightly lower than Fe^{2+} of concentration between 20 – 60 ppm. Findings obtained by Park and Yoon (2009) indicated that interaction of HA with Fe (II) salt reduced with decreasing pH. Therefore, it was anticipated that the much higher removal of ^{226}Ra was not due to the HA-Fe complexes. The removal of ^{226}Ra could be due to the interaction of ^{226}Ra species with Fe species. Taskaev et al. (1977) had reported the incorporation of ^{226}Ra in iron organic compounds following the extraction of soil with $FeCl_3$ solutions.

CONCLUSIONS

The effect of Al^{3+} on the removal of ^{226}Ra from the Ra-contaminated soil studied depended on the concentration of Al^{3+}. Additional Al^{3+} with concentration 20 – 60 ppm encouraged the removal of ^{226}Ra. However, as concentration increased to 80 and 100 ppm, further increase in the removal of ^{226}Ra was not detected. Generally, Ca^{2+} and Cu^{2+} served as competitor to ^{226}Ra species as decrease in the removal of ^{226}Ra was observed in the presence of these metal ions. Nonetheless, it was shown that Ca^{2+} served as a slightly better competitor compared to Cu^{2+}. Meanwhile, Fe^{2+} with concentration between 20 – 60 ppm also competed with ^{226}Ra. Nevertheless, increase in the removal of ^{226}Ra was detected in the presence of Fe^{2+} of higher concentration of 80 and 100 ppm,

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