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MODIFIED LEACHING OF ^{238}U AND ^{232}TH FROM WATER LEACH PURIFICATION (WLP) RESIDUE

(Modifikasi Larut Lesap Bagi ^{238}U dan ^{232}TH dari Residu Pemurnian Larut Lesap Air (WLP))

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Abstract

In recent years, the importance of actinide element mobile fractions in risk assessment evaluation of contaminated sites has increased. The knowledge of the sorption kinetics of ^{238}U and ^{232}Th and the thermodynamics behind the sorption process on radioactive waste is important to understand their mobility. In this study, the leaching process of ^{238}U and ^{232}Th from water leach purification (WLP) residue of Lynas Advanced Materials Plant was investigated to obtain the optimum leaching conditions by combining synthetic precipitation leaching procedure and batch method for the simulation of acid rain and heavy flooding, correspondingly. The initial ^{238}U and ^{232}Th concentrations, and the concentration at varies pH and contact time of ^{238}U and ^{232}Th from the WLP residue were studied. Results show that the initial concentrations of ^{238}U and ^{232}Th in the WLP residue are 6.6 and 206.1 mg/kg, respectively. In general, the highest values of the concentration after the leaching process of ^{238}U and ^{232}Th are 0.363 and 8.288 mg/kg, correspondingly. These results show the maximum potential remobilisation of ^{238}U and ^{232}Th at pH 4 with the same contact time of 14 days. At a similar duration, the maximum percentages of leaching are 5.50% and 3.99% for ^{238}U and ^{232}Th, respectively. Moreover, at pH 7, the minimum leaching percentages of ^{238}U and ^{232}Th are 4.7% and 3.61%, correspondingly. Thus, remobilising ^{238}U and ^{232}Th shows that the rate of leaching is influenced by the pH of leachant used. The maximum concentrations of ^{238}U and ^{232}Th are obtained at low pH such as pH 4. At pH 7 and 8, the leached amounts of ^{238}U and ^{232}Th are minimum. Therefore, combining the SPLP and batch method is practical for estimating the leaching and remobilisation of ^{232}Th and ^{238}U from WLP residues. The combined method may be useful for monitoring and risk assessment in environmental studies.

Keywords: leaching, WLP residue, uranium, thorium

Abstrak

Dalam tahun-tahun kebelakangan ini, kepentingan pergerakan unsur aktinida menjadi perhatian semasa penilaian risiko tapak-tapak yang tercemar. Pengetahuan tentang penyerapan kinetik dan termodinamik di sebalik proses penyerapan sisa radioaktif adalah penting untuk memahami pergerakan ^{238}U dan ^{232}Th. Dalam kajian ini, proses kelarut lesapan ^{238}U dan ^{232}Th dari residu pemurniaan larut lesap air (WLP) dari Lynas Advanced Materials Plant telah dikaji untuk mendapatkan keadaan kelarut lesapan yang optimum menggunakan gabungan prosedur larut lesap pemendapan sintetik dan kaedah kelompok, masing-masing sebagai simulasi semasa keadaan hujan asid dan banjir. Kepekatan awal ^{238}U dan ^{232}Th, dan kepekatan ^{238}U dan ^{232}Th terhadap pH dan masa berbeza dari residu WLP turut dilihat. Keputusan menunjukkan kepekatan awal ^{238}U dan ^{232}Th dalam residu WLP masing-masing adalah 6.6 mg/kg dan 206.1 mg/kg. Secara amnya, nilai tertinggi kepekatan selepas proses larut lesap bagi ^{238}U dan ^{232}Th masing-masing adalah 0.363 mg/kg dan 8.288 mg/kg. Kedua-dua hasil menunjukkan bahawa potensi pergerakan maksimum adalah pada pH 4 bagi ^{238}U dan ^{232}Th dalam masa 14 hari. Dalam tempoh masa yang sama, peratusan kelarut lesapan maksimum dicatatkan sebanyak 5.50% dan 3.99% masing-masing bagi ^{238}U dan ^{232}Th. Selain itu, pada pH 7, peratusan larut lesap minimum bagi ^{238}U dan ^{232}Th masing-masing adalah 4.7% dan 3.61%. Oleh itu, pergerakan ^{238}U dan ^{232}Th menunjukkan ia dipengaruhi oleh pH pelarut yang digunakan. Kepekatan maksimum ^{238}U dan ^{232}Th diperoleh pada pH yang lebih rendah seperti pH 4. Pada nilai pH 7 dan 8 mencatatkan kepekatan larut lesap ^{238}U dan ^{232}Th yang minimum. Kesimpulannya, gabungan kaedah SPLP dan kaedah kelompok adalah praktikal untuk menganggar larut lesap dan pergerakan ^{238}U dan ^{232}Th dari residu WLP. Gabungan kaedah ini boleh digunakan sebagai tujuan pemantauan dan penilaian risiko terhadap kajian alam sekitar.

Kata kunci: larut lesap, residu WLP, uranium, torium

Introduction

Demands for rare earth elements (REEs) have been increasing in many new energy technologies and systems that contribute to reduced greenhouse gas (GHG) emission and fossil fuel depletion [1]. However, producing REEs is far from being environmentally sustainable because it requires significant material and energy consumption whilst generating considerable air/water emissions and solid waste. Although many REEs are critically important for sustainable mobility and energy supply, producing REEs incurs serious environmental damages [2].

The Mount Weld mine is operated by Lynas Corporation Ltd. which is involved in mining and concentrating REE ore. On average, the ore contains 0.075% ThO_2 and 0.003% U_3O_8; these contents correspond to the average activity concentrations of 2.7 Bq g^{-1} of ^{232}Th and 0.3 Bq g^{-1} of ^{238}U [3]. The mining and processing concentrations of REE ore at Mount Weld is followed by a shipment of the concentrate to an REE processing facility, that is, Lynas Advanced Materials Plant (LAMP) at Gebeng, Pahang State, Malaysia, where further processing is conducted to produce high-purity REE compounds [3]. LAMP processes lanthanide concentrates at an integrated processing site which utilises physical and chemical treatment processes. Water leach purification (WLP) residues resulted from the leaching and purification of water-soluble lanthanide components from the calcined, cracked concentrate. According to the United States Environmental Protection Agency, rare earth concentrate is classified as a Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) [4]. REEs have been considered beneficial and profitable in Malaysia, but occupational, public safety and health risks related to REEs may be addressed at their mining, transportation, processing, waste disposal and decommissioning stages [5]. Thorium is a waste product in REE mining; it is mainly eliminated during the leaching process of the desired metals. Furthermore, thorium which denotes a radioactive impact of REE mining in addition to uranium and their decay products has raised environmental concerns for the last several years [6].

In addition to WLP residues, LAMP produces flue gas desulphurisation (FGD) and neutralised underflow (NUF) residues. Among the residues, the WLP is considered the most toxic given its elevated levels of uranium, thorium, associated radioactivity and heavy metals [7]. The WLP residue process generates the most radioactive and contaminated tailings. The WLP residue contains most of uranium and thorium separated from the REE products during the refining process. The concentrate used as feedstock for the refining process contains uranium (30 ppm) and thorium (approximately 1700 ppm) generated between 3.5 and 8 Bq g^{-1} of a specific activity. Theoretically, all the radioactive elements are removed during the refining process and are expected to be transferred to waste streams and disposed of in the tailing pond.

Leaching is a process in which contaminants are transferred from a stabilised matrix to liquid media, such as water or other solutions. Chemical reagents, such as acids, bases and chelating agents, are used to create a leaching solution to investigate the mobilisation of heavy metals from the soils [8]. Previous researchers have used leaching test to investigate the leachability of environmental contaminants from oil sludge to assess potential environmental impact [9, 10]. Then, results from leaching have been extrapolated to assess the long-term release of toxic elements from the oil sludge [10, 11]. Similar studies that used modification and combination of leaching tests have also been conducted for ^{238}U and ^{232}Th in WLP samples and have been discussed elsewhere [12, 13]. A leaching test has similar concepts adopted for soil washing.

Soil washing is frequently used in soil remediation because this process (i) completely removes contaminants, thus ensuring the rapid clean-up of a contaminated site, (ii) satisfies specific criteria, (iii) reduces or eliminates long-term liability, (iv) may be the most cost-effective solution and (v) may produce recyclable materials or energies.

Chemical reagents must be added to the washing water because washing the soils with water alone will remove the low amount of cations in the leachates [14-17]. Generally, soil washing with acids relies on ion exchange and dissolution of soil components or discrete metal compounds to extract metals [8]. Strong acids attack and degrade the soil crystalline structure at extended contact times [18]. Many research studies have mentioned different types of acids used as leaching agents, thereby resulting in different metal removal efficiencies [19]. The effectiveness of soil washing is closely related to the ability of the extracting solution to dissolve the metal contaminants in soils. However, the strong bonds between the soil and metals complicate the cleaning process [18]. Moreover, efficiency is affected by extraction time with chemical reagents because the rate of metal extraction is a factor of time [8]. Although many studies on soil washing which uses different chemical reagents have been conducted, the number of reports that focused on leaching process kinetically and thermodynamically have remained insufficient in literature.

This study aims to simulate the potential remobilisation of ^{238}U and ^{232}Th that leach from the WLP residue over a range of pH with different contact time. Synthetic precipitation leaching procedure (SPLP) experiments have been performed to provide information on ^{238}U and ^{232}Th release, thus simulating the acid rain condition.

Materials and Methods

Sample preparation

The WLP residue samples were collected from LAMP, Kuantan, Malaysia. The sample was dried and sieved using a mechanical sieve to separate <2 mm particles from a coarse material. Then, the samples were transformed into pellets with a ratio of 1:6, that is, 1.0 g of WLP residue and 6.0 g of boric acid. The initial concentration of ^{238}U and ^{232}Th was determined using X-ray fluorescence spectrometer (XRF) Bruker/S8 Tiger/2009 at the Centre for Research and Instrumentation, Universiti Kebangsaan Malaysia (UKM), Bangi. The leaching test was applied to the sample to eliminate any possibility of redistributing actinides between different phases given air oxidation.

Leaching experiments

The SPLP test (Method 1312) is designed in accordance with the procedure of Solid Waste Manual 846 [20] to determine the mobility of organic and inorganic analytes present in liquids, soils and wastes. The SPLP test is used to simulate the effect of acid rain on land-disposed wastes. However, the leaching experiment is modified from the SPLP by adding pH 7 and 8, thereby simulating groundwater pH. It can also be used to determine the concentration of a contaminant that will leach from the soil. The method uses a diluted sulphuric and nitric acids (with a pH of 4.2) as the leaching solution. This method can simulate weathering for 100 years of exposure to acid rain [21] and has been studied with other leaching tools [10, 11]. This single-batch extraction method is used to assess the leaching potential of a waste exposed to rainfall. The procedure is performed using the same equipment used to perform the toxicity characteristic leaching procedure and requires using one of two leaching fluids depending on the location of the waste disposal site. A 20:1 liquid-to-solid (L/S) ratio and an extraction time of 18 ± 2 hours were used in this test. The leachate fluid consists of slightly acidified deionised water that is formulated to simulate natural precipitation. A mixture of 60:40 (H_2SO_4:HNO_3; by weight) is used to achieve the appropriate pH for the extraction fluid [20].

Combined with the previous procedure, 10 g of WLP residue was equilibrated with 200 mL of leaching solution at a 20:1 ratio with various pH values (4, 5, 7 and 8) as a function of time. Additional information on batch leaching test can be found elsewhere [9-13]. The pH of the solution was adjusted prior to the experiments, thus simulating the potential remobilisation of ^{238}U and ^{232}Th after heavy flooding of the catchment area. The meter was calibrated prior to each set of measurements, and checks were obtained on other buffers between readings to ensure that all buffers are read and recorded at ±0.05 pH units. The pH analysis did not require a sample treatment but the sample solution should be measured immediately. The agitation was held at 200 rpm using a stirrer at a fixed temperature (27 °C) for 18 hours. Then, the samples were left in the room with different contact time. This approach presents the correlation between leachates and contact time. All the samples were then observed on the 1st, 2nd, 7th and 14th day. Then, the solid/liquid slurry is filtered and analysed by using inductively coupled plasma–mass spectrometer (ICP-MS) ELAN 9000 PerkinElmer SCIEX at the Malaysia Nuclear Agency, Bangi. Data were recorded, and each extraction method was prepared in duplicate. Blanks of leaching solutions (pH 4, 5, 7 and 8) without WLP were run as controls.

Percent of contaminant mass in the SPLP leachate also known as percentage uptake was calculated as equation 1:

$$\frac{(C_L \times V_L)}{(C_T \times M_S)} \times 100, \tag{1}$$

where C_L is the leachate concentration (mg/L), V_L is the volume of leachate in litres (0.2 L), C_T is the total sample concentration (mg/kg) and M_S is the mass of the sample in kilograms (0.01 kg) [22].

Results and Discussion

The initial concentration of ^{238}U and ^{232}Th in WLP residue samples was analysed using the XRF. The analysis finding shows that the concentration of ^{238}U and ^{232}Th is 6.6 and 206.1 mg/kg, respectively [12, 13]. According to the Malaysia Nuclear Agency [23], the WLP residue with a production of approximately 32,000 tonnes per year contains 1655 ppm ^{232}Th and 22.5 ppm ^{238}U. The radioactive residue is stored in retention ponds which are built with impermeable soil and are lined with a high-density polyethylene cover to prevent any seepage. WLP residue was also classified as a radioactive residue in accordance with the preview of Act 304 and regulations [24]. The treated effluent was analysed for its thorium radionuclide contents, ^{226}Ra and ^{228}Ra, but the concentrations did not reach the detection limit of 0.5 Bq/L during a test run. Thorium is assumed to be completely precipitated as thorium pyrophosphate (ThP_2O_7) during the separation with MgO at pH 3.5 and discharged in the WLP. The WLP residues contain abundant compounds in addition to ThP_2O_7, such as iron, barium, calcium or silica species. The composition of WLP residue affects the elemental solubility in an aqueous phase. ThP_2O_7 is water-insoluble formed by converting the sulphate of thorium, whereas the rare earth sulphates remain stable in sulphate solution [24,25].

The leaching process depends on several physical and chemical factors and occurs whenever a liquid phase percolates or is in contact with a solid material, thereby dissolving inorganic or organic substances [26]. The most important physical factors are size, shape and porosity of the particles, homogeneity of mineral phases, temperature, time frame, flow rate or liquid amount of leachant and degree of saturation and wet/dry cycles. The most important chemical aspects are related to pH and redox conditions, control mechanisms (kinetic or equilibrium), availability for leaching, complexation and sorption processes [27]. The leaching processes may involve precipitation/dissolution, diffusion, sorption, surface coating or wash-off which may be controlled kinetically or by equilibrium conditions [28]. Numerous tests, such as identifying leachable constituents [29], may be used depending on the objectives to evaluate the leaching processes. The present study focuses on ^{238}U and ^{232}Th. The most important variables that must be established for a specific leaching test are the sample preparation, leachant composition (pH, redox conditions), method of contact, L/S ratio, contact time, temperature and leachant separation method [28].

The pH values of leachate used in modified SPLP method are illustrated in Figure 1. The pH is selected to simulate acid rain (pH = 4 ± 0.05 and pH = 5 ± 0.05) because air includes several acidic emissions such as hydrogen fluoride (HF) and sulphuric acid (H_2SO_4) [7]. The modified SPLP was adjusted to pH = 7 ± 0.05 and pH = 8 ± 0.05 to simulate groundwater condition. Soil and water contaminants may be released by natural processes, such as weathering or human mismanagement, when constructing defective impoundment areas which may break or leak. Environmental effects of acid gases are popular and can lead to localised ‘acid rain’ which can damage crops and acidify freshwater bodies such as lakes and streams [30]. LAMP which is situated on the east coast peninsula of Malaysia receives considerable rains during monsoon season. These climates would lead to heavy flash floods and might cause several dams to overflow, thereby dispersing the WLP residue by wind and rain. The leaching of ^{238}U, ^{232}Th and other metals may contribute to increased groundwater contamination.

Figure 1 demonstrates that the average ^{238}U concentration leached from the WLP residue is in the ranges of 0.271–0.363 mg/kg (pH 4), 0.256–0.349 mg/kg (pH 5), 0.283–0.31 mg/kg (pH 7) and 0.235–0.323 mg/kg (pH 8), whereas that of the ^{232}Th concentration is in the ranges of 2.078–8.228 mg/kg (pH 4), 2.054–8.08 mg/kg (pH 5), 2.139–7.44 mg/kg (pH 7) and 1.827–8.026 mg/kg (pH 8). At low pH, the concentration of ^{238}U and ^{232}Th is the maximum. The maximum concentration value is frequently reported as the availability for leaching. For both radionuclides, at pH 7 and 8, the leached amount is minimum. The mixture of two concentrated acid solutions does not have a buffering characteristic. Thus, the solution mixture does not resist changes in the concentrations of hydronium and hydroxide ions, as well as the pH. The resistance of a buffer solution to pH change is based on Le Chatelier's principle and the common ion effect [31].

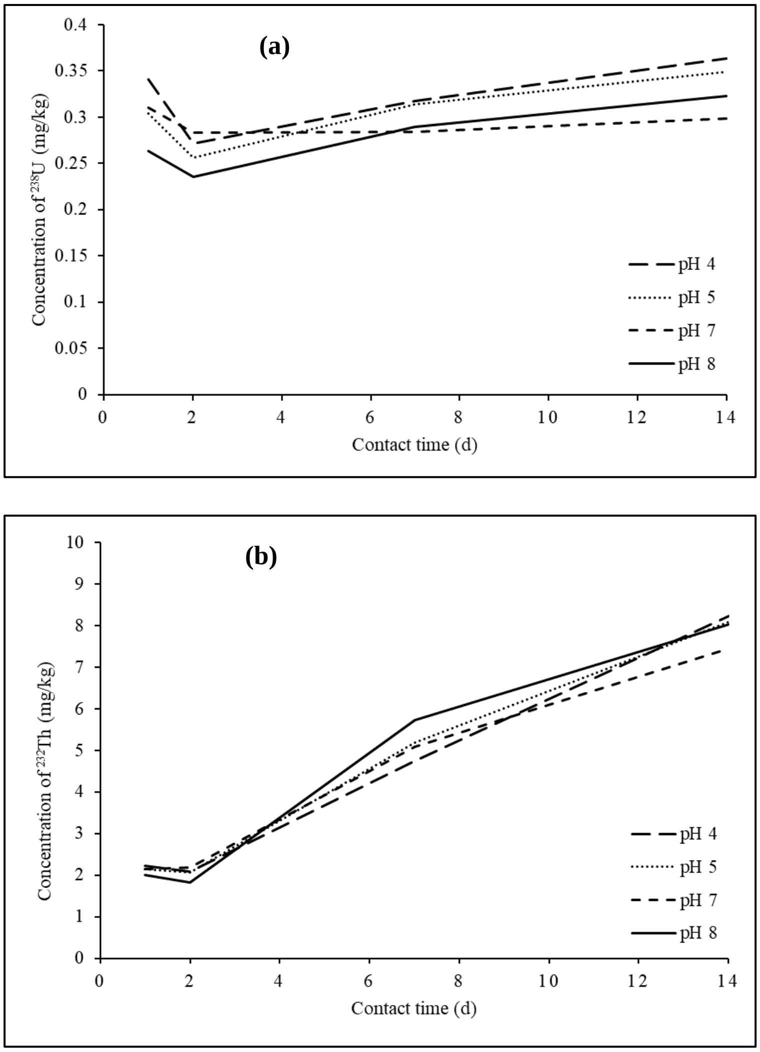

Figure 1. Effect of contact time and pH on the leaching concentrations of (a) ^{238}U and (b) ^{232}Th.

Figure 1 exhibits that the highest values of the concentration of ^{238}U and ^{232}Th are 0.363 and 8.228 mg/kg, correspondingly [12, 13]. The concentration of ^{232}Th is higher than ^{238}U probably due to the ^{232}Th chemical and physical characteristics, namely, strong sorption to solid matrices, thereby resulting in an extremely low bioavailability and mobility. Thorium (Th^{+4}) is highly insoluble in water. Under acidic conditions below pH 4, the solubility increases, and complexation with carbonates, sulphates or organic substances enhances its solubility [32]. Al-Areqi et al. [24] found that the maximum recovery and concentration of ^{232}Th are obtained by HNO_3 and H_2SO_4 in comparison with NaOH. Thorium is mostly available as an insoluble material during the physical beneficiation process, but it is transformed during the chemical processes into extra soluble species, such as $Th(NO_3)_4$, $Th(OH)_4$, Th-oxalate and other species [32]. In soils, humic substances are considered particularly important in adsorbing thorium. In most cases, sediment resuspension and mixing may control the transportation of particle-sorbed thorium in water; however, in several cases, the concentration of dissolved thorium in waters may increase due to formation of soluble complexes with carbonate, humic materials or other ligands in the water [33]. The contamination of groundwater by transporting thorium from soil to groundwater does not occur in most soils, except those with low sorption characteristics and the capability to form soluble complexes. Chelating agents produced by certain microorganisms (*Pseudomonas aeruginosa*) present in soils may enhance the dissolution of thorium in soils [32].

Contact intervals of 1–14 days were applied to evaluate leaching as a function of contact time. This approach is different from previous studies [10, 11] which have extended intervals based on a different standard leaching procedure. This period was selected because it was reported as a period of flooding on the east coast peninsula of Malaysia which receives abundant rain during the monsoon season. In Figure 1(a), the concentration of ^{238}U slightly increased with contact time, whereas Figure 1(b) displays that the concentration of ^{238}Th rapidly increased with contact time. Moreover, Figure 2 depicts the average percentage release for ^{238}U and ^{232}Th. The patterned release for ^{238}U and ^{232}Th was presented in a log scale line to represent the release behaviour on the basis of the calculated percentage release using Eq. (1). The maximum percentages of leaching are 5.50% and 3.99% for ^{238}U and ^{232}Th, respectively. These results show the maximum potential remobilisation of ^{238}U and ^{232}Th at low pH (pH=4) during the end of the duration test at 14 days. By contrast, the minimum leaching efficiencies at pH 7 were recorded for ^{238}U and ^{232}Th at 4.7% and 3.61%, correspondingly.

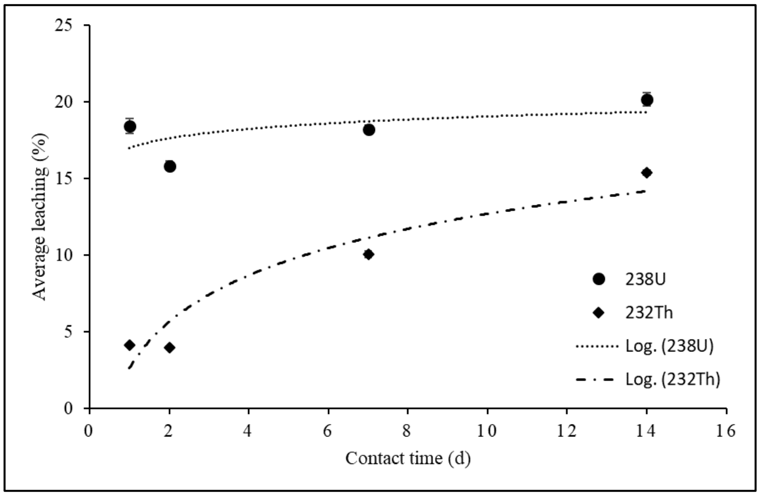

Figure 2. Percentage of average ^{238}U and ^{232}Th release as function of time

Conclusion

In summary, the effect of ^{238}U and ^{232}Th leaching release was moderately influenced by the pH of the solution used. The findings indicate that the maximum potential remobilisation percentages of ^{238}U and ^{232}Th at pH 4 at the final-day testing interval (14 days) are 5.50% and 3.99%, respectively. By contrast, at pH 7, the minimum leaching release was found for both waste products. Potential mobilisation of ^{238}U and ^{232}Th was observed from the WLP in the aqueous medium during the monsoon and flooding seasons.

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