American Journal of Environmental Engineering 2018, 8(1): 11-16
DOI: 10.5923/j.ajee.20180801.03

Radionuclide Adsorption Performance in Cement and Soil Medium for Safety Issue of Radioactive Waste Disposal Facility

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Abstract Radionuclide migration from a disposal facility is one of the most crucial factors for protecting man and environment. To ensure safety of a disposal facility, radionuclide migration or adsorption behavior is a key issue in operational and post operational phase. In this perspective, laboratory based column experiments was carried out to investigate the adsorption behavior of radionuclide on the cement and soil mediums. Present experiments simulate the migration behavior of radionuclide through the cement and soil barrier into the environment if the waste packs inside a disposal site encounter rain infiltration. In this study, some simulant cement chunk was produced with a water-cement ratio of 0.50 each, and soil samples were prepared with various grain sizes from 90 µm to 2 mm. In this study adsorption and hence migration behavior of solid spent resin was verified experimentally. In addition, adsorption of two radionuclides, namely ^{60}Co and ^{137}Cs was investigated as liquid waste by using the gamma spectrometry analysis. The highest activity distribution of ^{60}Co at 1173.2 keV and ^{137}Cs was found in concrete medium 53.49%, and 46.82 in the soil medium, respectively. Thus, locally prepared multiple barriers of cement and soil medium could reasonably be suitable for liquid waste immobilization.

Keywords Adsorption, Migration, Disposal, Spent resin, Cobalt-60, Cesium-137

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1. Introduction

Near-surface repositories are accepted publicly in many countries as a good option for low or intermediate level radioactive waste (L/ILW) disposal that generates at nuclear power plants [1]. The concept of secure isolation criteria of the waste in repository development has enhanced the confidence in the feasibility of safe disposal. The safety evaluation of these repository concepts on radionuclide migration behavior has been demonstrated through rigorous safety assessment methods, development of safety technics and independent review of these assessments [2]. Migration of radionuclides in a wet geological formation and engineered barrier is one of the most important factors to be considered for safety evaluation of a radioactive waste disposal facility [3, 4]. This is particularly important for Bangladesh due to high precipitation rate, as it potentially cause the release of radionuclides from shallow land disposal of L/ILW into clayey soil formation in the natural geological environment [5]. This is because, excessive precipitation promotes radionuclide release from shallow land disposal to the engineered barrier, and consequently the clayey soil of natural geological formation can raise potential safety concern. The operational safety and post-closure safety of a disposal facility needs to be ensured for the safety of the public and environment. Operational safety is provided by means of engineered features and operational controls. Post-closure safety is provided by means of engineered and geological barriers; it does not depend on monitoring or institutional controls after the facility has been closed. That is, a facility needs to design to be passively safe [6]. Thus, understanding of the performance of this type of disposal system and its safety features and processes needs to be investigated based on the experimental observation and scientific knowledge [7].

Nowadays, L/ILW is generating from various facilities across the country, including the TRIGA Mark-II Research Reactor (RR) of Bangladesh Atomic Energy Commission (BAEC). In RR facility, ion exchanging resin is commonly used in resign column to adsorb radio-elements from primary coolant circuit, and eventually resign makes themselves a

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Published online at http://journal.sapub.org/ajee

radioactive waste [8]. Generally, spent resin is considered as L/ILW, and hence it can be disposed near surface level. During the operation of RR, L/ILW is generally produced. These wastes should be properly treated and disposed in order to minimize the potential hazards of environments. In this regard a cost effective retarding materials was prepared by using locally available cement to introduce in the near surface disposal concept in order to minimize the disposal cost. In addition, clayey soil layer was used in the present study to investigate its retarding capacity as a backfilling medium. The performance of the retarding materials was verified by passing fresh water through spent resin, and then through the retarding materials to check the radionuclide adsorption. A similar technique was used to verify adsorption behavior of the retarding materials while liquid solution of ^{60}Co and ^{137}Cs were passed through column. In this study, Madhupur clay collected from Gajipur district of Bangladesh and locally made cement blocks were used in the column experiments as the barrier material. Madhupur clay is highly weathered, brownish red to brick red clay. It is sticky, inter-bedded with fine sand and silt and contains ferruginous concretions and ferruginous and calcareous Madhupur nodules. The barrier effectiveness of this clayey soil was verified in a laboratory based column experiments focusing on the rain fall condition. Cemented blocks were used as an engineered barrier to be useful in a near surface disposal facility. Failure of engineered barrier will lead the radionuclide release into the host soil medium. Keeping this in mind, present study of radionuclide adsorption and hence migration behavior was performed in the aforesaid two mediums. The prime objective of this paper is to characterize the local soil to be useful as a potential backfilling medium and to prepare a cost effective cemented engineered barrier for retardation of nuclides, if get released from a prospective near surface repository in case of rain infiltration. The scope of this research work incorporates the safety evaluation methodologies for shallow land disposal concept in clayey soil formation. In this regard solid spent resin, ^{137}Cs and ^{60}Co radioisotopes have been selected as a simulant radioactive waste to analyze their migration, and hence adsorption behavior in local soil and cement matrix for the safety performance of a potential disposal site in Bangladesh.

2. Materials and Methods

2.1. Sample Collection and Preparation

Ion exchanging AMBERLITE IRN150 radioactive spent resin sample was collected from the onsite interim of RR facility. Spent resin samples of total weight 300 g were collected in a plastic container. Then, resin sample was distributed into three plastic pots with 95.68 g each for each experimental run. During sample collection the dose rate of the resin sample was found to be 68 μSv/h, and surface activity concentration level was 46 Bq/cm^2. To prepare the standard solution of ^{60}Co and ^{137}Cs, some samples of 0.125 mg were collected from the sealed vial using micro-pipette that was stored in a pit source room of central radioactive waste processing and storage facility (CWPSF). The collected samples are then diluted by adding the normal tap water for experimental run. The activity concentration of all the collected samples was characterized by using the high purity germanium (HPGe) detector. Soil samples were collected from Madhupur tract of Joydebpur. This soil is known as deep red-brown terrace soil which contributes about 1.54% of total soil in Bangladesh. The grain sizes of each soil sample were measured by using the sieve technique. Then, soils of various grain sizes were mixed up to ensure the multiple size particle content in each soil sample. The properties of characterized soil samples are presented in Table 1.

Table 1. Characterized grain size of the soil samples used for each experimental run

Weight of soil sample, W (g)	Test sieve size	Weight of sieved soil Sample, X (g)	Grain size percentage (%) = (X/W)*100%
282.51	2 mm	184.37	65.26%
	1 mm	52.21	18.48%
	400 μm	27.35	9.68%
	90 μm	14.8	5.25%

Cement chunks were prepared from the ordinary Portland Cement (OPC) available in the local market. This type of cement is generally produced from grinded clinker and a certain amount of gypsum, fly ash, slag and limestone. These chunks were made in rectangular dimension (1.5 cm × 1.5 cm × 1cm) using this locally available OPC with an optimized water-cement ratio of 0.50. To prepare the concrete chunks from locally available cement and sand a cement-sand ratio of 1:1.5 was maintained. Representative properties of the cemented samples, and used OPC are presented in Table 2.

Table 2. Properties of the cemented samples, and used OPC material for each experimental run

Sample size (cm)			Compressive Strength (psi)	Type of Cement	Content of Cement	
Length	width	Thickness			Gypsum	Slag, Fly Ash & Limestone
1.5	1.5	1.0	1500 - 1550	Portland Composite Cement (PCC)	0 - 5%	21 - 35%

2.2. Activity Calculation of Samples

The initial activity concentration of each sample spent resign and liquid solution was measured prior to perform the respective experimental run. Then, in each experimental run with spent resin fresh water was passed through the soil and cement medium inside the experimental column. In the course of the experimental progression, activity gained in the column medium due to migrated radionuclide from the solution to the concrete and soil medium. After completion of each experimental run the moisten soil and concrete samples were collected, and activity was measured by the HPGe detector. The leached out (i.e., passed out) residual water was collected to measure its gained activity by the same method. For this purpose, a HPGe detector of 40% efficiency and 2 KeV resolution (at 1.33MeV) was used. The activity concentration is calculated based on the following equation (1),

$$A = \frac{cps}{E * I_{\gamma} * w} \quad (1)$$

Where,
$A =$ Activity concentration of the sample in Bq.kg^{-1};
CPS = Net count per second (CPS of each sample – CPS of background value);
$E =$ % counting efficiency;
$w =$ Weight of the sample in g;
$I_{\gamma} =$ Absolute intensity of the gamma ray;
The standard deviation of the detector's counts was estimated by the following equation (2)

$$2\sigma = 2 * [\frac{N_a}{{T_a}^2} + \frac{N_b}{{T_b}^2}]^{\frac{1}{2}} \quad (2)$$

Where,
$N_a =$ Counts measured in time T_a;
$N_b =$Background measured in time T_b.

2.3. Experimental Procedure

Laboratory based column experiments was performed to verify the adsorption behavior of radionuclides in the soil and cement medium. The experimental setup and flow diagram with various processing steps are shown in Fig. 1 (a) and (b). In this column experiments, gravity driven discharge of fresh tap water was passed through the radioactive spent resign layer, and successively through the concrete and soil layers. In the experimental runs with Cobalt-60 (^{60}Co) and Cesium-137 (^{137}Cs) sources, their radioactive solution was directly passed through the studied mediums. Throughout the experiment, water flow rate of around 0.028 mls^{-1} was maintained by using the stopping knob of a burette. In this study, Madhupur clays were used as soil layer to simulate the backfilling material of a potential repository. Cement chunks were used in the experimental column to simulate the nuclide adsorption behavior in the cemented engineered barrier system (EBS) of a repository. Locally manufactured EBS was used in experiments as an absorbent structure to verify its capacity to limit the migration of radionuclides caused by groundwater pathway. In this study, simplified laboratory based column experimental technique was used to evaluate the radionuclide immobilization and migration behavior in some potential host, and EBS medium, such as soil layer, and cemented medium. In the experiments, a series of column runs were performed for the radioactive spent resin, and liquid solution of ^{60}Co and ^{137}Cs sources. To prepare the experimental columns, resign samples were collected from the onsite interim of Research Reactor facility, where radioactive spent resin was safely stored for the future disposal. Standard liquid solution of ^{60}Co and ^{137}Cs were prepared from their concentrated solution that was collected for R&D purposes from the pit sources of (CWPSF). Consequently, column experiments were performed with the prepared liquid solution of ^{60}Co and ^{137}Cs to analyze their adsorption behavior in the studied medium. In the column experiments, fresh water was passed through the concrete and soil layers, and then leached out water was collected from the lower part of the column matrix. The concrete chunks were used in the experiments as a simulant of the EBS absorbent that were prepared with the water-cement ratio of 0.50. In the soil layer of the experimental column, Madhupur clay was used which is deep red-brown terrace soil and collected from Madhupur tract of Joydebpur in Gajipur district of Bangladesh. After completing each experimental run, the activity concentration in each part of the column was estimated. In this purpose, HPGe detector was used to analyze the radionuclide adsorption affinity of cement and soil medium. Consequently, the differences in adsorbed activity of each layer in the column were detected.

The experimental procedures are presented in a flow diagram as shown in Fig. 1(b). This diagram describes the various procedures involved in the experimental runs. These procedures were followed in every experimental run with radioactive spent resin, ^{60}Co and ^{137}Cs sources.

3. Result and Discussion

3.1. Assessment of Adsorption for Spent Resign

The initial activity concentration of the resin sample was measured prior to perform the respective experimental run. The observed activity concentration in spent resin is presented in Fig 2. From this graph it is evident that no radionuclide dissolution occurred during water flow through the radioactive resin to the soil and cement medium in the column. This observation indicates that spent resin can safely be disposed in a simplified cost effective manner in a near surface repository without any imposing any significant risk to the environment.

It is commonly observed that along with spent resin, Cesium-137 (^{137}Cs) and Cobalt-60 (^{60}Co) are artificially produced in nuclear reactors. Therefore, their adsorption behavior needs to be verified in the same column experiments to be useful for the safety assessment of a prospective repository.

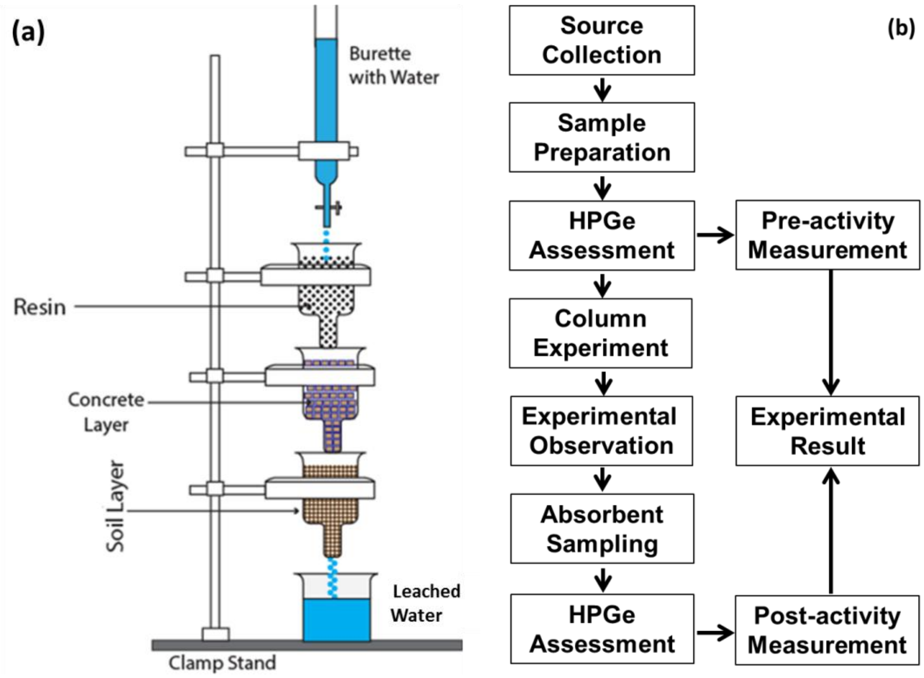

Figure 1. (a) Experimental setup for the column experiments, (b) Flow diagram of the overall experimental steps

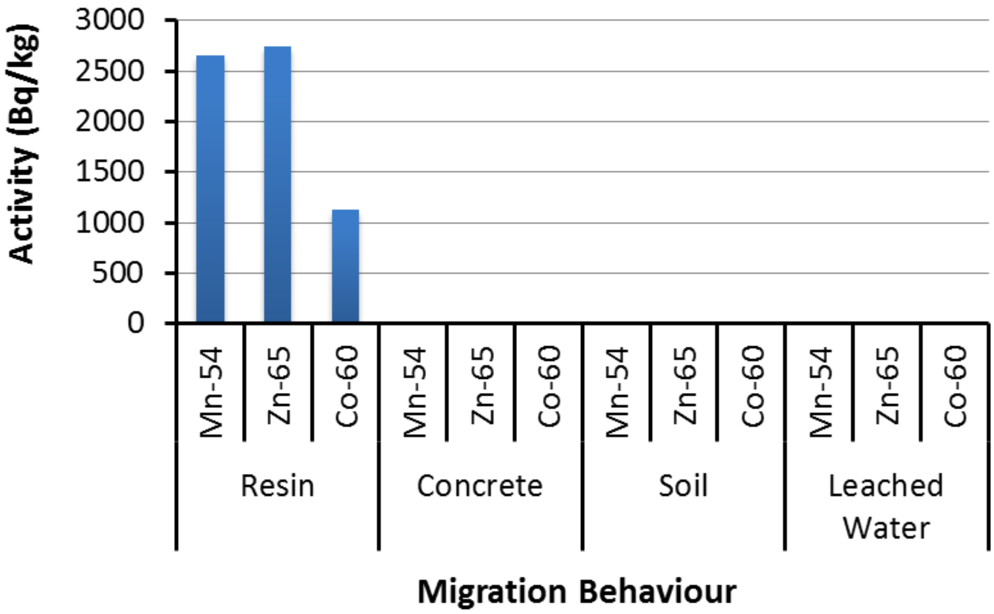

Figure 2. Adsorption characteristic of radionuclide from spent resin

3.2. Assessment of Adsorption for ^{60}Co Solution

The adsorption behavior of the ^{60}Co nuclide is important in the case of assessment of a disposal facility due to the presence of ^{60}Co in the nuclear reactor's waste stream. Although dissolution of ^{60}Co nuclide was not observed in spite of its presence in the spent resin, however, it needs to evaluate the adsorption behavior of ^{60}Co nuclide. Thus, the confirmation check of the migration behavior, and hence the adsorption affinity of ^{60}C was further verified with liquid ^{60}Co source. For this purpose, experiments were performed with gravity driven discharge of liquid ^{60}Co solution through the experimental column materials. The leached out water then collected in a pot and its activity was analyzed. In this experiments, the activity level of ^{60}Co isotope in the entire column mediums was analyzed in terms of the activity counts of two separate energy peaks as ^{60}Co radioisotope possess two energy peaks. The activity concentration of ^{60}Co at 1173.2 keV was found relatively higher than that of 1332.5 keV in all the column materials. The activity concentration of the initial solution was gradually reduced when it passed through the concrete and soil medium in the experimental runs. After each experiment, the activity concentration of the column materials and the leached out water was analyzed to quantify the variation of adsorbed activity concentration in the respective medium. Relatively low activity concentration in the leached out water indicates a considerable adsorption in the concrete and soil medium. A descending trend of the activity concentration in the column materials (i.e., concrete, soil and leached out water) was observed, as shown in Fig.3.

3.3. Assessment of Adsorption for ^{60}Cs Solution

To verify the adsorption affinity of ^{137}Cs in a similar manner of ^{60}Co, column experiments were performed with the standard active solution of ^{137}Cs. The activity concentration of ^{137}Cs in various medium is presented in Fig. 4. From this graphical view, the activity concentration of ^{137}Cs in the soil medium was found relatively higher than that of the concrete medium. The comparative assessment of the activity level between the concrete and soil medium indicates an adsorption affinity of ^{137}Cs in the soil medium. Relatively low activity concentration in the leached out water indicates a considerable adsorption in the concrete and soil medium.

3.4. Activity Distribution of ^{137}Cs, ^{60}Co in the Absorbents

The comparative adsorption behavior of ^{60}Co and ^{137}Cs radionuclide was assessed to verify the variation of adsorption affinity in the soil and cement layers, as shown in Fig. 5. From this graphical view it is recognizable that the adsorption affinity of ^{60}Co is relatively low in concrete medium than that of ^{137}Cs. The highest activity distribution of ^{60}Co (1173.2 keV) was found to be 53.49% in concrete. In the case of ^{137}Cs adsorption behavior, relative adsorption affinity in soil medium was found higher than that of the ^{60}Co nuclide. The highest activity distribution of ^{137}Cs was found to be 46.82% in the soil medium. The activity concentration in the leached out water was found relatively low for both ^{137}Cs and ^{60}Co in all the experiments. The highest activity distribution in the leached out water was found to be 15.17% and 15.59% for ^{60}Co and ^{137}Cs, respectively. This indicates relatively good adsorption capacity of the radionuclides in the studied mediums. In general observation, the distribution of activity concentration in the column materials indicates a variable adsorption affinity of the respective nuclides.

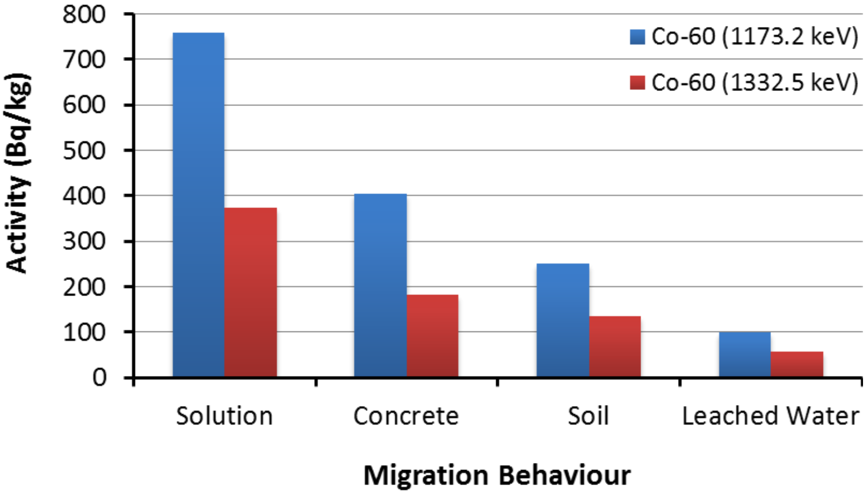

Figure 3. Migration characteristic of ^{60}Co in adsorbents and leached water

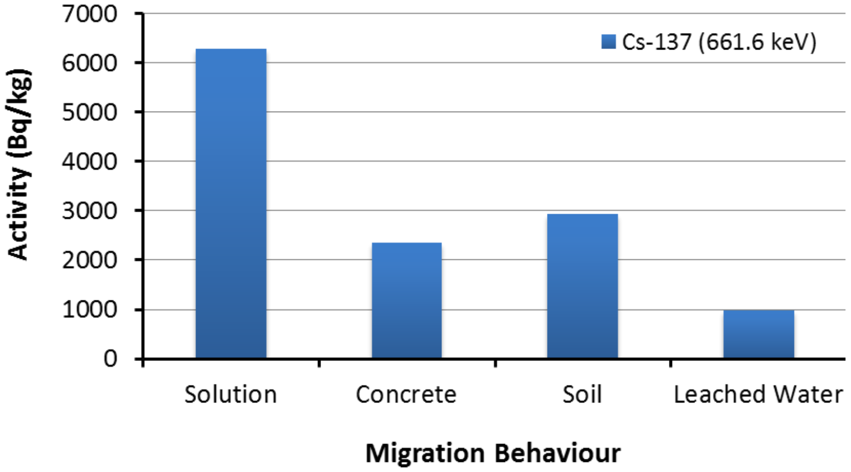

Figure 4. Migration characteristic of ^{137}Cs in adsorbents and leached water

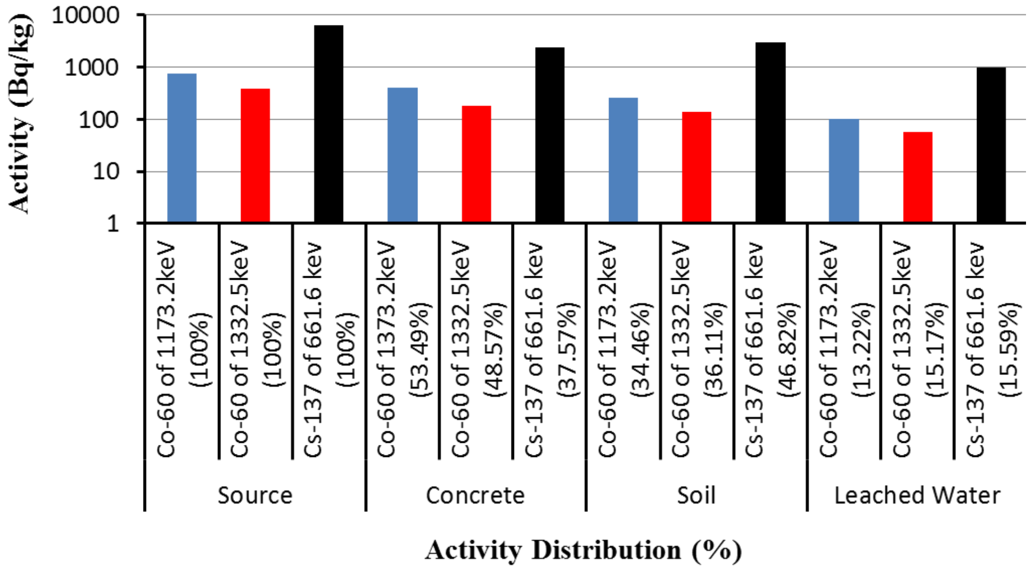

Figure 5. Activity distribution of ^{60}Co and ^{137}Cs in different layer of the column

4. Conclusions

Radionuclide-release was not identified from the spent resin in the present column materials. Thus, non-migration of radionuclide from the spent resin indicates no radionuclide dissolution. This observation indicates that the disposal of the spent resin used in the primary coolant circuit of the research reactor could be managed in a cost effective simplified cementation method. The migration, and hence adsorption of ^{137}Cs and ^{60}Co in the cemented and soil medium has been observed. This indicates radionuclide adsorption characteristic of the soil and cemented materials. Thus, cemented engineered barrier could be useful for the retardation of the L/ILW in a near surface disposal facility. The soil medium exhibits a good capacity of radionuclide adsorption, and hence can potentially be useful as a backfilling material for a disposal facility. The present experiments indicate relative low release of radionuclide in the leached water. Thus, concept of combined adsorption in the soil and cemented material could be useful to achieve a good retarding capacity for slow down the radionuclide migration in the biosphere to ensure the environmental safety. The methodical approach and experimental views of the present study could be useful for the preliminary safety assessment of a candidate site for a near surface repository.

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