Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2013.818923

52 (2014) 6817–6821
October

Taylor & Francis
Taylor & Francis Group

Migration test column for decontamination of radioactive liquids by using natural tuff

Erol Kam[a,*], Ahmet Erdal Osmanlioglu[a], Ahmet Bozkurt[b]

[a]*Cekmece Nuclear Research and Training Centre, Altinsehir Yolu, 5. Km, Halkali, Istanbul 34303, Turkey*
Tel. +90 2124732600x1050; Fax: +90 2124732634; email: erolkam@hotmail.com
[b]*Faculty of Arts and Sciences, Department of Physics, Harran University, Sanliurfa 63300, Turkey*

Received 8 November 2012; Accepted 13 June 2013

ABSTRACT

This study aims to evaluate the feasibility of using natural tuff found in the middle Anatolian region of Turkey for the removal of radioactivity from liquid radioactive wastes. Block samples that were collected from four different tuff deposits were first crushed to obtain particles with various sizes. The samples were then treated with low-level liquid radioactive waste in laboratory conditions. The effect of particle size on the adsorption performance of the tuff samples was studied for four types of radionuclides (241Am, 134Cs, 137Cs, and 60Co). The removal rates of radioactivity by the tuff samples having five different aggregate sizes (diameters of 4, 2.8, 2, 1, and 0.7 mm) were determined by the decrease in radioactivity concentration of the passed liquid discharge. In addition, the specific retention, saturation, and adsorption rates of the samples were also investigated. For most of the radionuclides, up to 50% saturation level and 70% specific retention were observed while the optimum results were acquired for the samples with 2 mm particle size.

Keywords: Aqueous radioactive waste; Natural tuff; Radioactivity removal

1. Introduction

For the past several decades, the world has seen the rapid accumulation of radioactive wastes produced as a result of industrial or medical uses of nuclear technologies. The waste is usually classified as high level or low level depending on the degree of radioactivity of its contents and is either in the solidified or the liquid discharge form. The biggest challenge in waste management activities, which has become an issue of health and environmental concern, is now finding appropriate methods of handling radioactive wastes before shipping them for the final disposal in a long-term repository. Removing the radioactive substances from the waste solution is necessary since the radioactive ions pose dangers to human health and the environment due to their high toxicity, and long half-lives. Among the treatment options in use today, adsorption by a sorbent material is known to be a significant method available especially for liquid wastes [1]. There are various candidates that can be used as sorbents and usually naturally found materials are chosen for their high ion exchange capacities, abundance, and low cost. Tuffs, usually referred to as zeolitic tuffs, have been proposed as ion exchangers to remove polluting cations from wastewaters, as an alternative to the

*Corresponding author.

methods based on precipitation. In general, it can be concluded that both zeolitic materials have good properties for the sorption of cesium from aqueous solutions [2].

In several countries, tuff deposits are being investigated as one of the geological formations that could possibly host an underground repository for radioactive waste particularly due to their favorable isolation performance [3]. The properties and behavior of tuff depend on its grain-scale structure and characteristics. In a final waste repository, transport by groundwater will be the principal mechanism for the migration of radionuclides to the biosphere. This study aims to evaluate the feasibility of using natural tuff from some Turkish deposits as an adsorbent in removing radioactivity from low-level liquid radioactive wastes.

2. Sampling locations

The natural tuff samples investigated in this study were collected from four different deposits located in the middle Anatolia region of Turkey (Fig. 1) with the following main specifications [4].

Selime tuff: It is formed by volcanic particles that are smaller than 4 mm in diameter. Being generally white, the color changes from white to pink according to alteration. The thickest formation is located in Selime and Yaprakhisar villages (60–100 m) and extends toward western and northern directions.

Kizilkaya ignimbrites: The deposit is located in the village of Kizilkaya. They are the most common volcanic rocks in the region (about 5,000 km^2) and are more stable than the Selime tuff.

Hasan Mountain: This tuff is also formed by volcanic particles smaller than 4 mm. The deposit is generally located at the north, west, and south outskirts of Hasan Mountain.

Melendiz Mountain: The deposit is formed by volcanic ashes from the outskirts of Melendiz Mountain.

3. Materials and methods

3.1. Sample preparation

The specimens were cored from block samples that were taken from each deposit. They were NX sized (5.4 cm diameter and had a length to diameter ratio 2:1) [5]. Uniaxial strength tests were applied on the air-dried specimens at room temperature [6]. Some properties of the samples are shown in Table 1. To investigate the effect of grain size on migration of radionuclides, different aggregate sizes were prepared from each tuff sample. For this purpose, the block samples were crushed and mechanically sieved through meshes to obtain five different particle sizes (with diameters of 4, 2.8, 2, 1, and 0.7 mm) for the adsorption experiments.

Migration tests were applied by using a test column (Fig. 1). The column was filled by grinded tuff as ion exchange media. Radioactive liquid waste was poured into this column and passed liquid from the column was taken and analyzed. Comparison of pH dependency of sorption between tuff and its constituent minerals suggested that it contributed to Am-241, Cs-134, Cs-137, and Co-60 sorption on tuff. Through diffusion experiments of Am-241, Cs-134, Cs-137, and Co-60, the prepared tuff samples were carried out at the pH values of 8 and 10. Effective diffusion coefficients obtained at both pH values were almost the same.

3.2. Migration parameters

Porosity is a physical property of a soil or rock that is defined as the ratio of the amount of pore space present in the sample to the total volume of the sample [7]. It is a dimensionless quantity denoted by n and is given as

$$n = V_v/V_T \quad (1)$$

where V_v is the volume of voids and V_T is the total volume (Often, porosity is expressed as a percentage by multiplying the ratio by 100).

Depending on the porosity of the tuff samples, effective parameters for radionuclide migration can be listed as the following.

Effective porosity (n_e): porosity available for flow; fraction of interconnected pore space. For massive rocks, it can be 1/10 of total porosity.

Saturation: Ratio of $V_w/V_v = 1$, where V_w is the volume of liquid waste.

Specific release (S_y): The volume of liquid waste that is released from the saturated tuff under gravity divided by the total volume of rock.

Specific retention (S_r): Volume of liquid waste remaining after release divided by the total volume of rock.

So for tuff in saturated condition, porosity may be given as

$$n = S_y + S_r \quad (2)$$

A spectrometric analysis system was used to determine activity concentrations. The system consisted of a coaxial-type high-purity germanium detector that was linked to a multi-channel buffer

Fig. 1. Natural tuff sampling locations.

consisting of an analog-to-digital converter. For data acquisition, ACCUSPEC was used as the multi-channel analyzer software. The energy resolution (FWHM) observed in the measurements was 1.8 keV at the 1.33 MeV Co-60 reference transition. Counting times were arranged up to 20,000 s to provide sufficient counting statistics. The detector was surrounded by lead and copper that provide an efficient suppression of background γ radiation present at the laboratory. Specific radioactivities (Bq/l) of Am-241, Cs-134, Cs-137, and Co-60 were measured as; 2,400, 4,200, 4,800 and, 3,600 Bq/l, respectively.

4. Results

Table 1 provides some essential information on the removal rate of radioactivity by natural tuff under experimental conditions. The porosity of each tuff sample was determined to be between 25 and 47% where the average porosities were roughly the same (32–38%). The Kizilkaya samples had higher water absorption values (38%). Based on the generic tests of volcanic tuff deposits in Turkey, Kizilkaya ignimbrites have the highest water absorption ratio and porosity. Because of these reasons Kizilkaya ignimbrites are the most suitable site for further research. The compression

Table 1
Properties of tuff samples

	Selime	Kizilkaya	Hasan	Melendiz
Density, g/cm^3	1.602 ± 0.07	1.76 ± 0.06	1.37 ± 0.08	1.375 ± 0.09
Volume mass, g/cm^3	2.52 ± 0.3	2.57 ± 0.2	2.39 ± 0.3	2.57 ± 0.2
Porosity, %	32.9 ± 3	38.6 ± 2	37.4 ± 1	37.6 ± 2
Water absorption, %	27.6 ± 5	37.4 ± 4	28.4 ± 4	30.3 ± 5
Uniaxial compression strength, MPa	19.5 ± 5	25.4 ± 5	19.4 ± 5	13.3 ± 6

strength of this formation (25 MPa) was found to be higher than the other samples.

The removal of radioactivity from the liquid waste by the natural tuff was examined by measuring the amount of radioactivity remaining in a real radioactive solution. This low-level radioactive waste contained four different radionuclides: ^{241}Am, ^{134}Cs, ^{137}Cs, and ^{60}Co. In the study, the effects of the parameters such as heat, pressure, flow rate, salt content, etc. on the removal rate were not investigated. The adsorption performance of the samples was characterized for all of the four radioisotopes. Experimental setup and the corresponding procedure used in the study were previously outlined by Osmanlioglu [8,9]. The low-level radioactive liquid was injected into the tuff samples at a constant flow rate. At the end of each injection, radioactivity concentration of the passed liquid was determined for different particle sizes under saturated conditions of tuff samples.

Specific retention of the samples was determined (for 2 mm particle size) according to the saturation levels of each sample (Fig. 2). The saturation retention tests showed that up to 50% saturation, the specific retention of the radioisotopes was 60–70%. Laboratory tests showed that the best result for the removal rate varied depending on the type of radioisotope in the solution. At 2 mm particle size, the removal of radioactivity by the tuff samples seemed sufficient (Fig. 3).

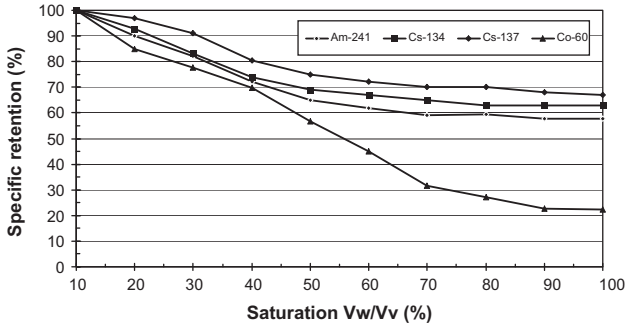

Fig. 2. Saturation vs. specific retention for 2 mm crushed tuff media.

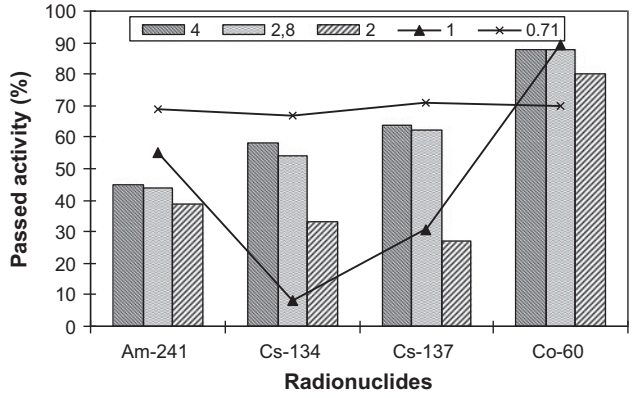

Fig. 3. Passed radioactivity vs. particle size.

Although the best particle size was 1 mm for ^{134}Cs isotope, this was not convenient for the other radionuclides. However, for particles with larger or smaller sizes, the isolation performance of the natural tuff samples deteriorated. It can be deduced from this result that the tuff samples with 2 mm grain size offer adequate isolation performance for migration of radionuclides up to 100 ml/min flow rates. In addition, the smaller particles (less than 1 mm) performed similarly for all the radionuclides in the study and were not efficient sorbent materials for radioactivity removal.

5. Conclusion

Optimum particle size for the natural tuff samples collected from different deposits in Turkey was determined for radionuclide removal tests. Saturation, retention, and migration tests were applied on natural tuff samples. The results of radionuclide migration studies indicate that the particle size of 2 mm has the best performance and the highest specific retention for most of the radionuclides studied. Further decrease in particle size of the tuff samples was not ensured by an increase in effectiveness of the removal of radioactivity from the liquid waste. Based on the generic tests of volcanic tuff deposits in Turkey,

Kizilkaya ignimbrites are considered to be the most suitable site for further research.

References

[1] C.J. Roberts, Management and disposal of waste from sites contaminated by radioactivity, Radiat. Phys. Chem. 51(4–6) (1998) 579–587.
[2] R. Cortés-Martínez, M.T. Olguin, M. Solace Rios, Cesium sorption by clinoptilolite-rich tuffs in batch and fixed-bed systems, Desalination 258 (2010) 164–170.
[3] International Atomic Energy Agency, Classification of Radioactive Waste: A Safety Guide, Safety Series No. 111-G-1.1, International Atomic Energy Agency, Vienna, 1994.
[4] Environmental Status Report, Provincial Directorate of Environment and Forestry (in Turkish) Turkey, Nigde (2009).
[5] American Society for Testing and Materials, ASTM, D4646–03 Standard Test Method for 24-h Batch-type Measurement of Contaminant Sorption by Soils and Sediments, ASTM, West Conshohocken, PA, 2003.
[6] American Society for Testing and Materials ASTM, D7012–04 Standard Test Method for Compressive Strength and Elastic Moduli of Intact Rock Core Specimens under Varying States of Stress and Temperatures, ASTM, West Conshohocken, PA, 2004.
[7] S.W. Lohman, Ground-Water Hydraulics, Geological Survey Professional Paper 708, US Department of the Interior, Washington DC, 1979.
[8] A.E. Osmanlioglu, Treatment of radioactive liquid waste by sorption on natural zeolite in Turkey, J. Hazard. Mater. B137 (2006) 332–335.
[9] A.E. Osmanlioglu, Natural diatomite process for removal of radioactivity from liquid waste, Appl. Radiat. Isot. 65 (2007) 17–20.