



An Efficient Radiochemical Method for Extraction of ^{226}Ra From the Soil Samples

Amir Mehdizadeh¹, Sedigheh Sina^{1,2*}, Reza Faghihi¹, Mohammad Hosein Sadeghi¹

¹Department of Nuclear Engineering, School of Mechanical Engineering, Shiraz University, Shiraz, Iran

²Radiation Research Center, School of Mechanical Engineering, Shiraz University, Shiraz, Iran

***Corresponding author:**

Sedigheh Sina, Department of Nuclear Engineering, School of Mechanical Engineering, Shiraz University, Shiraz, Iran
Tel: +989172228349
Fax: +987116473474
Email: samirasina@shirazu.ac.ir

Received: 5 December 2019
Accepted: 20 December 2019
ePublished: 30 December 2019

Abstract

Background: Radium-226 is a radioactive element, with a very long half-life of 1600 years, producing radon gas. According to the United States Environmental Protection Agency, radon gas is the second most important factor causing lung cancer.

Objectives: The purpose of this study was to separate ^{226}Ra from the soil of high background radiation area by a radiochemical method for using in radon calibration chamber.

Methods: ^{226}Ra can be used in standard calibration chambers for calibration of radon detection systems. For this purpose, radiochemical method was used to extract radium from the soil with a high concentration of ^{226}Ra . Four soil samples used in this study were selected from high background radiation areas of Ramsar, north of Iran. Equal amounts of samples were gathered from each region and ground. The specific activity of radium-226 was measured with HPGe detector. The highest specific activity of radium-226 (44.8 Bq/g) belonged to Talesh-Mahaleh. After radiochemical separation of ^{226}Ra , the specific activity of extracted radium crystals was measured with the HPGe detector.

Results: According to the results, the specific activity of ^{226}Ra was found to be 94.97 Bq/g. Therefore, the specific activity of ^{226}Ra was 2.12 times greater in the extracted crystals than in the original soil samples.

Conclusion: The results indicate that using the radiochemical method proposed in this study, ^{226}Ra can be extracted with an efficiency of 42%.

Keywords: Soil sample, ^{226}Ra , HPGe detector, Radiochemical method, Extracted crystals



Background

Radium has 4 radioactive isotopes, ^{226}Ra (half-life=1600 y), ^{228}Ra (half-life=5.75 y), ^{223}Ra (half-life=11.4 d), and ^{224}Ra (half-life=3.7 d) originating from the decay of ^{235}U , ^{238}U and ^{232}Th isotopes (1,2). The abundance of each radioisotope in environmental samples depends on several important factors such as the specific activity concentration, mobility of its parent nuclide, and its half-life. Radium radioisotopes are highly mobile in the environment and are important radionuclides to be considered in the radiological protection of the environment due to its prevalence (3,4). ^{226}Ra is an alpha-emitting element, with a very long half-life of 1600 years, which decays into ^{222}Rn gas. The chemical and biological behaviors of ^{226}Ra are similar to those of the other alkaline earth metals (5). Radium-226 is an important naturally occurring radionuclide in the oil and gas industry. This is mainly due to its long half-life and radiological effects owing to the similarity of its chemical and biological behaviors to those of other alkaline earth metals (6-8).

Radon is a naturally occurring radionuclide in the earth's crust, and its concentration is different in different geographic areas. The alpha particles produced in the decay of ^{222}Rn gas are very dangerous as radon enters the body through breathing. Although the half-life of radon gas is short, it can accumulate in buildings or spring water and increase the risk of internal exposure. According to the United States Environmental Protection Agency (EPA), radon gas is the second most important cause of lung cancer (9). Therefore, the use of detectors with accurate response is necessary for the measurement of radon concentration in dwellings. Standard Radium sources are used for calibration of radon detectors. The purpose of this study was to use radiochemical method for soil decontamination and extraction of ^{226}Ra from the soils with a high concentration of ^{226}Ra .

Materials and Methods

Sample Collection and Preparation

There are a few regions in the world having high levels

of natural background radiation. Some areas of Ramsar have been found to have the maximum amount of natural radiation in the world. In this study, different soil samples were selected from different regions of high background areas in Ramsar, north of Iran. These regions are Talesh-Mahaleh, Khake-Sefid, Sadat-Mahaleh, and Shahrake-Golha. Equal amounts of samples were gathered from each region and ground. To measure the radioactivity of the samples and select the soil with the highest specific activity of radium-226, the soils were transferred to Marinelli containers. The containers were sealed and kept for a month in order that the radioactive secular equilibrium is reached. The concentration of ^{226}Ra in all samples was obtained using High Purity Germanium Detector (P-type Coaxial HPGe, USA) and a Multichannel analyzer (System 8000, USA). The sample which was gathered from Talesh-Mahaleh had the highest specific activity and was selected for radiochemical process and extraction of radium-226. The activity of the radioactive elements in the samples was calculated according to equation 1 (10).

$$A = \frac{net_{cps}}{\epsilon \times p} \quad (1)$$

where A is the activity of a specific element in the soil sample, net_{cps} is net detector counts per second in a particular energy, ϵ is the detector efficiency in the given geometry and energy which was obtained using a sample with known activity concentration and the same geometry as the sample with unknown activity, and p is the probability of decay of the radioactive elements in the sample. These samples are the AQCS reference materials prepared by the international atomic energy agency (IAEA) which include thorium (RGTh), uranium (RGU), potassium (RGTh), and some other reference materials. Such reference materials are used for obtaining the efficiency and also for quality assurance of our measurement system.

To ensure the quality of the gamma spectroscopy procedure in our laboratory, we participated in inter-comparison proficiency test which is performed by the Iranian atomic energy organization each year.

The gamma peaks of 242, 295, 352 keV of ^{214}Pb and 609 and 1765 keV of ^{214}Bi were used for determining ^{226}Ra concentration. Finally, the specific activity of the elements may be obtained by the following equation (10):

$$A_s = A / m \quad (2)$$

Where A_s is the specific activity of ^{226}Ra in the sample, A is the ^{226}Ra activity, and m is the mass of the sample.

Radiochemical Separation of Radium-226

Soil samples can be dissolved using a special flux.

Radium-226 in solution is determined by precipitation of the sample with barium sulfate.

The extraction method (11) is as follows:

1) Flux preparation

a) 15 mg of BaSO_4 , 32.9 g of K_2CO_3 , 15.3 g of Na_2CO_3 , and 16.8 g of $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ were mixed in a platinum crucible.

b) The mixture was heated.

c) The mixture was rotated occasionally after melting.

d) The solution was transferred to a porcelain mortar after 30 minutes.

e) The solution was then dried and passed through a 10-mesh screen.

f) It was stored in an airtight bottle.

(1) 1 g of the soil sample was moved to a platinum crucible.

(2) 8 g of the powdered flux was added to and mixed with the soil.

(3) The platinum crucible was placed for 30 minutes on the burner until the material is melted. During this time, it was rotated occasionally to blend well.

(4) The crucible was removed from the burner and rotated so that the solution was solidified.

(5) The following solution was prepared in a beaker: (120 mL distilled water + 10 mL 18M H_2SO_4 + 5 mL 30% H_2O_2)

(6) The platinum crucible was placed in the beaker, and let the sample be separated from it.

(7) The magnetic stirring bar was transferred to the beaker and started stirring. Then 50 mL of diluted BaCl_2 solution was added to it.

Diluted BaCl_2 solution is:

(Dissolve 0.3558 g $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ in 100 mL distilled water.)

(8) The contents of the beaker were stirred for 90 minutes, then, the magnetic stirring bar was removed, and let the BaSO_4 precipitate overnight.

(9) The clear liquid above the beaker was removed so that the sediments remained.

(10) The sediments were completely poured into a 0.7-micrometer filter and rinsed twice with a vacuum pump and 0.1 mol sulfuric acid solution.

Sulfuric acid solution (0.1 mol): Diluted 0.6 mL of 96% sulfuric acid solution to 100 mL with distilled water.

(11) The filter was taken off carefully and placed inside the platinum crucible.

(12) 25 drops of 48% HF and 0.3 mL of 10% ammonium sulfate solution were added to the crucible.

Ammonium sulfate solution (10%): (Dissolved 0.5 g ammonium sulfate in 5 mL distilled water.)

(13) The crucible was placed on a hot plate at 100°C until the contents evaporated and dried.

(14) 2 mL of acetone-ethanol mixture was added.

Acetone-ethanol mixture: (50% Acetone and 50% ethanol)

(15) The crucible was placed on the burner for 10 minutes until the ashes were left.

(16) The crucible was removed from the heat and 1 mL of 15M H₃PO₄ was added to it.

(17) The crucible was put on a hot plate for 15 minutes at 100°C. For 30 minutes, the temperature was increased every 10 minutes.

(18) The crucible was placed on the burner until the white matters were dissolved and the bubbles and foam were seen.

(19) The crucible was placed in a hot water bath and filled with 3M HCl.

3M HCl: (5 mL of 37% HCl +15 mL distilled water)

(20) The liquids finally evaporated after about 3 hours and white crystals remained.

Sample Measurement

The obtained crystals were transferred to the petri dish container. The petri dish was sealed well to prevent the leakage of radon gas and kept for a month to reach the secular equilibrium. The primary soil sample was also transferred into another petri dish and completely sealed. After reaching the equilibrium, the soil and crystal containers were measured for 24 hours. As the activity of the primary sample was determined in the previous step, the detector efficiency for petri dish container can be calculated according to equation 1. According to the obtained data and the existing relationships, the activity and specific activity of the extracted crystals were calculated.

Results

The Activity of Radium-226 in Soil Sample

The activity of radium-226 in the soil samples was calculated using equation 1, as the average of activities of several peaks of radon progenies, i.e., 242, 295, and 352 keV of ²¹⁴Pb and 609 and 1765 keV of ²¹⁴Bi. The measured data of the soil samples are given in Tables 1, 2, 3, and 4. The spectrum of the sample with the highest activity is shown in Figure 1.

The ²²⁶Ra activity in the primary soil sample was found to be 5.37 kBq. The specific activity of the primary sample was found to be 44.8 Bq/g according to equation 2.

The Activity of Radium-226 in Extracted Crystals

Gamma ray spectroscopy was performed for the extracted crystals. The activity of radium-226 in extracted crystals was calculated as the average of activities of several peaks of radon progenies, as described in the previous section.

The ²²⁶Ra activity in the crystals was found to be 735 Bq. The specific activity of the extracted crystals was found to be 94.97 Bq/g according to equation 2. Table 5 compares the activity of the primary soil and the extracted crystals.

Table 1. The Activity of Soil Sample from Talesh-Mahaleh (Sample Weight: 120 g)

Energy (keV)	Nuclide	Soil activity, Bq
242	Pb-214	5110
295	Pb-214	5682
352	Pb-214	5291
609	Bi-214	5152
1765	Bi-214	5633
$A_s = 5374/120 = 44.8 \text{ Bq/g}$		

Table 2. The Activity of Soil Sample From Khake-Sefid (Sample Weight: 120 g)

Energy (keV)	Nuclide	Soil activity, Bq
242	Pb-214	4992
295	Pb-214	5364
352	Pb-214	5010
609	Bi-214	5163
1765	Bi-214	5239
$A_s = 5154/120 = 42.9 \text{ Bq/g}$		

Table 3. The Activity of Soil Sample From Sadat-Mahaleh (Sample Weight: 120 g)

Energy (keV)	Nuclide	Soil activity, Bq
242	Pb-214	4527
295	Pb-214	5008
352	Pb-214	4625
609	Bi-214	4883
1765	Bi-214	4902
$A_s = 4789/120 = 39.9 \text{ Bq/g}$		

Discussion

The specific activity of the soil samples collected from Ramsar was between 40-45 Bq/g, which is about 1000 times greater compared to the normal soil (12).

In this study, an average of 0.2 g crystal was extracted from each gram of soil. By comparing the activity of radium-226 for 1 g of soil and 0.2 g of crystals, the average efficiency of the ²²⁶Ra extraction from the soil sample was found to be 42% according to equation 3. Based on the following equation, this efficiency was calculated.

$$\varepsilon_{ext} = \frac{0.2 \times (A_s \text{ of } ^{226}\text{Ra in extracted crystals})}{1 \times (A_s \text{ of } ^{226}\text{Ra in soil sample})} \quad (3)$$

By comparing the specific activity of radium-226 for the soil sample and extracted crystals, the quality of the crystals was determined using the following equation.

$$Q = \frac{(A_s \text{ of } ^{226}\text{Ra in extracted crystals})}{(A_s \text{ of } ^{226}\text{Ra in soil sample})} \quad (4)$$

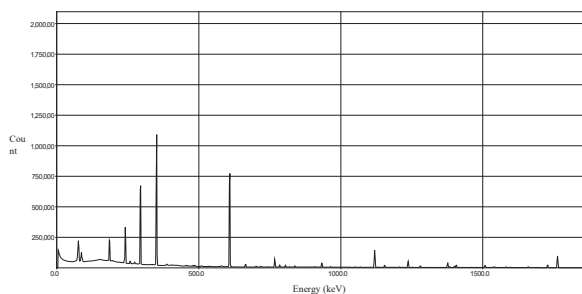


Figure 1. Spectrum of the Primary Soil Sample Measured by HPGc Detector (24 Hours).

Therefore, it can be concluded that the radiochemical method used in this study is a very effective method for extracting ^{226}Ra from the soil samples.

Conclusion

Radium-226 is usually used as a calibration source for radon detectors. It can be used in both liquid and solid forms in the calibration chambers. Currently, no standard radium-226 source exists in Iran to use as a calibration source. The aim of this study was to extract radium-226 from the soil with the highest specific activity so that it is usable as a calibration source.

In this study, a very effective radiochemical method was used for extracting ^{226}Ra from the soil samples. The specific activity of the soil samples was between 40–45 Bq/g, which is about 1000 times greater compared to the normal soil. An average of 0.2 g crystal was extracted from each gram of soil. The average efficiency of the ^{226}Ra extraction from the soil sample was found to be 42% according to equation 3. The specific activity of ^{226}Ra was 2.12 times greater in the extracted crystals than in the soil samples. Therefore, a useful Radium source was extracted from the soil sample and can be used as a calibration source in radon calibration chamber. The extracted ^{226}Ra can also be effectively used as a standard source for calibration of radon detectors.

Conflict of Interest Disclosures

None.

References

- Jia G, Torri G, Ocone R. Determination of radium isotopes in soil samples by alpha-spectrometry. *J Radioanal Nucl Chem.* 2007;273(3):779-83. doi: [10.1007/s10967-007-0948-9](https://doi.org/10.1007/s10967-007-0948-9).
- Molinari J, Snodgrass WJ. *The Chemistry and Radiochemistry of Radium and the Other Elements of the Uranium and Thorium Natural Decay Series.* Vienna, Austria: International Atomic Energy Agency (IAEA); 1990. p. 599.
- Maxwell O, Wagiran H, Zaidi E, Joel ES, Tenebe IT, Oha IA, et al. Radiotoxicity risks of radium-226 (^{226}Ra) on groundwater-based drinking at Dawaki, Kuje, Giri and Sabon-Lugbe area of Abuja, North Central Nigeria. *Environ Earth Sci.* 2016;75(14):1084. doi: [10.1007/s12665-016-5884-y](https://doi.org/10.1007/s12665-016-5884-y).
- Wallner G, Steininger G. Radium isotopes and ^{222}Rn in Austrian drinking waters. *J Radioanal Nucl Chem.* 2007;274(3):511-6. doi: [10.1007/s10967-006-6939-4](https://doi.org/10.1007/s10967-006-6939-4).

Table 4. The Activity of Soil Sample From Shahrake-Golha (Sample Weight: 120 g)

Energy (keV)	Nuclide	Soil Activity, Bq
242	Pb-214	4641
295	Pb-214	5123
352	Pb-214	4899
609	Bi-214	5095
1765	Bi-214	5171

$A_s = 4986/120 = 41.5 \text{ Bq/g}$

Table 5. Comparison of ^{226}Ra Concentration in the Primary Soil and Extracted Crystals

Energy (keV)	Nuclide	Soil activity, Bq	Extracted Crystal Activity, Bq
242	Pb-214	5110	732
295	Pb-214	5682	724
352	Pb-214	5291	728
609	Bi-214	5152	747
1765	Bi-214	5633	733
	^{226}Ra activity (Average of other nuclides)	5374 Bq	733 Bq
	Specific activity of ^{226}Ra	44.8 Bq/g	94.97 Bq/g

- International Atomic Energy Agency (IAEA). *The Environmental Behaviour of Radium: Revised Edition.* IAEA; 2014.
- Al-Masri MS. Spatial and monthly variations of radium isotopes in produced water during oil production. *Appl Radiat Isot.* 2006;64(5):615-23. doi: [10.1016/j.apradiso.2005.11.015](https://doi.org/10.1016/j.apradiso.2005.11.015).
- Al Abdullah J, Al-Masri MS, Amin Y. Dissolution of (^{226}Ra) BaSO_4 and partial separation of ^{226}Ra from radium/barium sulfate: A new treatment method for NORM waste from petroleum industry. *Appl Radiat Isot.* 2016;107:377-81. doi: [10.1016/j.apradiso.2015.11.028](https://doi.org/10.1016/j.apradiso.2015.11.028).
- International Atomic Energy Agency (IAEA). *Application of the Concepts of Exclusion, Exemption and Clearance.* Vienna, Austria: IAEA; 2004.
- United States Environmental Protection Agency (EPA). *A Citizen's Guide to Radon: The Guide to Protecting Yourself and Your Family from Radon.* Available from: <https://www.epa.gov/radon/citizens-guide-radon-guide-protecting-yourself-and-your-family-radon>.
- Knoll GF. *Radiation Detection and Measurement.* J Franklin Inst. 291, 156. 1971. doi: [10.1016/0016-0032\(71\)90034-2](https://doi.org/10.1016/0016-0032(71)90034-2).
- Lieberman R. *Radiochemistry Procedures Manual.* Eastern Environmental Radiation Facility; 1984.
- Sperrin M, Gillmore G, Denman T. Radon concentration variations in a Mendip cave cluster. *Environmental Management and Health.* 2001;12(5):476-82. doi: [10.1108/09566160110404881](https://doi.org/10.1108/09566160110404881)