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Radium micro-precipitates using alpha spectrometry and total alpha counting measurement

T. Hosseini*, A. A. Fathivand

National radiation protection department Iranian nuclear Regulatory Authority, Tehran, Iran

ABSTRACT

Background: This study consists of two parts. The first part deals with both qualitative and quantitative analysis of ^{226}Ra using alpha spectrometry measurement method. In the second part, the percent age of radioactive equilibrium between ^{226} Ra and its daughter products were determined by alpha spectrometry and total alpha measurement system after elapsed time of 15 days from precipitation.

Materials and Methods: Twelve ^{226}Ra samples as barium-radium sulfate in form of micro-precipitate on millipore and Whatman #42 filters were prepared. An alpha spectrometer with surface barrier detector and a total alpha measurement system consists of scintillation crystal assembly ZnS (Ag) were used for counting.

Results: The minimum detection limit of alpha spectrometry and total alpha counting for ^{226}Ra measurements in samples for counting time equal to 10000 seconds, were found to be 3.7 mBq and 15.8 mBq respectively. Results from total alpha counting showed that radioactive equilibrium between ^{226}Ra and its daughter products reached to about 92%±3.5, where as, in the case of alpha spectrometry radioactive equilibrium, it was destroyed due to vacuum during counting the sample. Also in case of alpha spectrometry, the optimum sample to detector distance, was found to be 0.5 centimeter.

Conclusion: From this study it was concluded that micro-precipitation can be used as a proper method for sample preparation and alpha spectrometry micro- precipitates simultaneously of ^{224}Ra and ^{226}Ra in these precipitates, prepared from different samples. Besides it is not time consuming and sources can be measured immediately after sample preparation.

***Key words:** Radium, micro-precipitate, millipore and Whatman filters, alpha spectrometry, total alpha counter.*

INTRODUCTION

^{226}Ra and ^{224}Ra the members of uranium-238 and thorium-232 series respectively are alpha emitter radionuclides and do not add directly to gamma activity of the environment. But since they are chemically similar to calcium, they are therefore, absorbed from the soil by plants and are passed up the food chain to man. Then they are accumulated in the skeleton of individuals and cause particular biological effects. Therefore, determination of ^{224}Ra and ^{226}Ra in environmental samples and foodstuffs seems to be very important for human health especially in high natural radioactivity areas.

There are different kinds of measurement methods to determine ^{226}Ra content in natural samples. Some of these methods are gamma spectrometry, emanation of radon gas (Lucas 1977), total alpha counting, solid state nuclear track detectors (Sohrabi *et al.*1995) and alpha spectrometry (Sill 1981, Lozano 1999). Among these methods alpha spectrometry, due to its lower detection limit, is preferred for determination of ^{226} Ra in trace amounts. In this study

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* **Corresponding author:**
T. Hosseini. National Radiation Protection Department Iranian Nuclear Regulatory Authority, Tehran, Iran P.O.Box: 14155-4494, Fax: +98 21 8009502.
E-mail: thosseini@msn.com

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twelve ^{224}Ra and ^{226}Ra micro-precipitates were prepared as sources according to the technique reported by Sill (1983) and counted by alpha spectrometry and total alpha counter system.

MATERIALS AND METHODS

Specific amounts (100 µl) of a diluted ^{226}Ra standard solution (11.887±3.65% Bq/ml) (Amersham), 5ml citric acid (93 g/l) and 2ml lead carrier solution (100mg Pb^{+2}/ml) were added to 400ml distilled water. Three ml of Barium-133 tracer solution (3.191±2.95% Bq/ml) was used for calculation of chemical yield. The pH of solution was changed to 9 by addition of ammonia solution. Then, 9M sulfuric acid solution was added until the pH was changed to 1. In this situation radium and lead was precipitated as $Ra/PbSO_4$ (Kim, *et al.* 2001).

The sulfate precipitate was dissolved in alkaline 0.25M EDTA (Ethylene Diamine Tetraacetic Acid, disodium salt) solution. The micro-coprecipitate of radium was prepared by addition of very small amounts of barium carrier (50 µg Ba^{2+}) to the above solution and decreasing pH to 4.2 with 3M sulfuric acid (Lim *et al.* 1989, Sill 1983). In this pH, barium and radium precipitated in form of $Ra/BaSO_4$ but lead remained in the solution. After 30 minutes the micro-precipitate was filtered on a millipore or Whatman filter.

Twelve samples were prepared by using Whatman #42 and millipore 0.45 µm filters. The thickness of micro- precipitates was about 25µg/cm^2. All filters were counted by alpha spectrometry measurement system and total alpha counter system.

Measurement systems: Alpha spectrometry measurement system consists of a surface barrier detector, model PI450251 with 450 mm^2 area, 25 keV resolution (FWHM) and 100 microns depletion dept; a vacuum chamber model 7400A (canberra) and a MCA (Multi Channel Analyzer) model 35 plus (Canberra).

Total alpha measurement system consists of scintillation crystal assembly, RD - 13 (Alpha) [A layer of ZnS (Ag) crystals adhered to a lucite light pipe which is 3/4 inch in thickness and 2.49 inches in diameter (Eberline)] and a mini scalar model MS - 3 (Eberline).

RESULTS AND DISCUSSION

The alpha spectrums of radium precipitates on Whatman #42 and millipore filters immediately after preparation are shown in figure 1 and 2 respectively. Analysis of spectrums shows that compared to Whatman #42, millipore filter has better resolution (FWHM = 80 keV) and much less tailing. So, it was found that millpore filter is much more suitable than whatman filter to prepare sources to be counted by alpha spectrometry system, since peaks due to presence of different radionuclides can be isolated better.

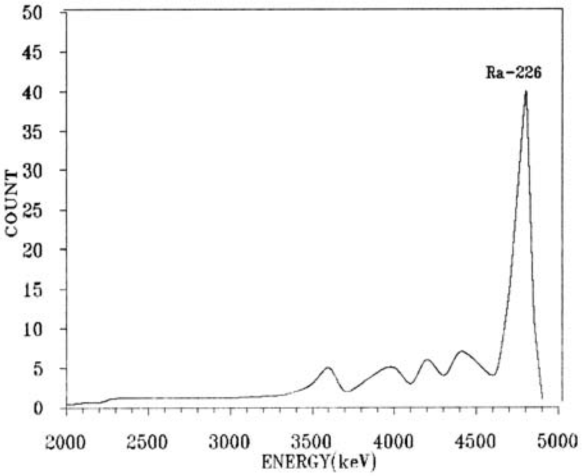

Figure 1. Alpha particle spectrum of ^{226}Ra micro-precipitate on Whatman filter immediately after preparation.

In order to obtain an appropriate distance between our samples and detector, the filters were counted in different sample to detector distances. Obtained results show that as the sample to detector distance increases, the counting efficiency decreases, so for our samples the optimum sample to detector distance at which both

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efficiency and resolution were good, was obtained to be 0.5 cm.

In order to study radioactive equilibrium between ^{226}Ra and its daughter products in the samples on millipore filters, one of the filters with known activity was analyzed by alpha spectrometry in different elapsed times after preparation. Immediately after filtration only ^{226}Ra peak was seen in the spectrum, where as, in the subsequent spectrums the area of radium daughter products' peaks grew. Considering the long half-life of ^{226}Ra, the increase of ^{226}Ra peak area in subsequent countings is assumed to be due to ^{226}Ra daughter products.

Theoretically 15 days after filtration, radioactive equilibrium should be about 93.5%, but we found it to be about 83%. It seems that vacuum process makes a part of radon gas come out from filter, which then the equilibrium destroys. In order to check this idea, after radioactive equilibrium was achieved in the precipitate, the filter was fully covered by a thin aluminium foil. The alpha spectrometry of this covered filter (with previous condition) showed radioactive equilibrium up to about 90%. So, from obtained results it was concluded that alpha spectrometry is not a good method to determine radioactive equilibrium in micro precipitates, unless special spray is used to fix and cover the precipitate. In this study the percent age of radioactive equilibrium between ^{226}Ra and its daughter products was equal to 92%± 3.5.

Results from alpha spectrometry of samples suggest that micro precipitation, as a sample preparation method is quite suitable for determination of ^{226}Ra in different samples. By this method it is possible to measure ^{226}Ra immediately after preparation of filter samples. The minimum detection limits of alpha spectrometry and total alpha counting system for ^{226}Ra measurements in our geometry and counting time equal to 10000 seconds were found to be 3.7 and 15.8 MBq respectively.

Radium precipitate, in the form of sulfate, may contain the other naturally occurring radium isotopes (i.e. ^{224}Ra and ^{228}Ra), which affect the measurement of ^{226}Ra. Among these radioisotopes ^{228}Ra is a beta emitter so it doesn't affect ^{226}Ra measurement. But the effect of ^{224}Ra on ^{226}Ra peak was determined by preparing samples of ^{224}Ra precipitate (from ^{232}Th solution) and calculating the percent of ^{224}Ra peak (about 4.6%), which lies on ^{226}Ra region [$E_{\alpha\,(Ra226)} = 4.78$ MeV and $E_{\alpha\,(Ra224)} = 5.69$ MeV]. It means that if sample contains ^{224}Ra and ^{226}Ra, 4.6% of ^{224}Ra peak region will lies in ^{226}Ra peak's region. This addition should be diminished while analyzing samples for ^{226}Ra contents. Figure 3 shows the spectrum of ^{224}Ra micro - precipitate 24 hours after filtration.

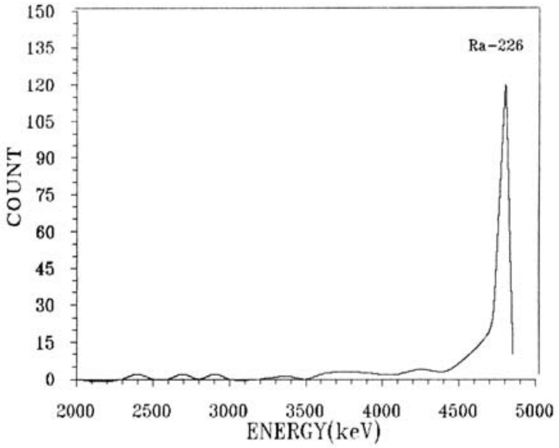

Figure 2. Alpha particle spectrum of ^{226}Ra micro-precipitate on millipore filter immediately after preparation.

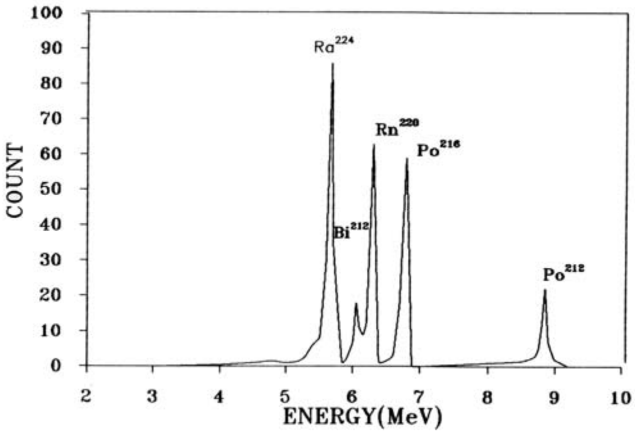

Figure 3. Alpha particle spectrum of ^{224}Ra micro-precipitate on millipore filter.

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CONCLUSION

Measurement of ^{224}Ra and ^{226}Ra contents in micro-precipitate samples were studied by alpha spectrometry and total alpha counting measurement methods. It was shown that it is possible to prepare micro-precipitate of radium in sulfate form from various environmental and food samples and measure it immediately after filtration process, without waiting to achieve radioactive equilibrium between radium and its daughter products. Also it is a fast method to measure radium in different samples. Analysis of alpha-particle spectrums of radium micro-precipitates on both millipore and Whatman # 42 filters showed that the former is more suitable for radium determination. For our samples the optimum sample to detector distance, which gives us both good efficiency and resolution, was found to be 0.5 cm. The percent of ^{224}Ra peak's region which lies in ^{226}Ra peak's region, due to its tailing, was found to be 4.6% of ^{224}Ra peak's that should be diminished while analyzing ^{226}Ra in the samples.

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