

Determination of Uranium Isotopes in the Ground Water

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ABSTRACT

The concentrations and activity ratios of uranium isotopes were determined in the ground water taken from 29 sites in Chungnam area. The concentrations of uranium isotopes at the different sampling points were variable, ranging between 0.8 to 70.1 ppb. The concentrations of uranium isotopes in the ground water at the Yusong area were higher than those at other regions. The activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the ground water was not unit, ranging between 1.0 to 3.3. The Yusong area samples showed lower $^{234}\text{U}/^{238}\text{U}$ activity ratios than the other region samples

1. Introduction

Natural radionuclide existing in the environment is an important in the general study of radionuclide migration, and the results of behavior of radionuclides may be used as a reference data in the case of an accident in a radioactive waste repository or in a nuclear fuel plant. Among the natural radionuclides occurrence in the environment, uranium isotopes are good tracers to study the behavior of these radionuclides, because natural uranium can be easily detected in nearly all materials from the environment.

Most natural waters contain a few ppb of dissolved uranium. In natural waters, uranium concentrations greater than 100 ppb are quite rare. The ground water quality largely depends upon the geological characteristics of the aquifer. The dissolved uranium content of continental ground water is a complex function of numerous variables; among these, aquifer rock type and ground water composition are probably most important [1].

The present work aims to determine the concentrations and activity ratios of uranium isotopes in the ground water. This result may be used as a reference data in the case of an accident in a radioactive waste repository or in a nuclear fuel plant, and elucidated the behavior characteristics of uranium isotopes in the ground water under natural conditions.

2. Materials and methods

Sampling points were chosen around Chungnam region. Ground water samples were taken at 29 sites from the spring. The pH was measured with a glass electrode. After adding ^{232}U as a yield tracer, 20 liters of ground water were evaporated to dryness until 100 ml using an evaporating system. Then, uranium isotopes were extracted from dissolved sample materials into TBP (tributyl phosphate) in carbon tetrachloride [2]. This extraction step separates uranium isotopes very effectively from most of the matrix elements, such as Si, Fe, alkali and alkaline earth elements. The radiochemical purification of the U fraction was performed with hydrochloric acid (1+1). Back extraction was done using HCl (1+11). Finally, purified uranium isotopes was electrodeposited from electroplating solution [3] on a polished stainless steel disc and measured by α -ray spectrometry.

3. Results and discussion

Typical uranium isotopes spectra obtained from the ground water samples were presented in Figs 1 and 2. As shown in Figs 1 and 2, the peaks of ^{238}U , ^{235}U , ^{234}U and ^{232}U were well separated from the radionuclides. However, ^{235}U activity must be determined very carefully. When determining ^{235}U in environmental samples, the normally poor accuracy due to counting statistics and the wide band energy (from 4.2 MeV to 4.7 MeV), which makes the associated error higher than that of the other uranium isotopes, is to be considered in the correct determination of ^{235}U isotope.

The results of concentrations and activity ratios of uranium isotopes in the ground water were shown in Fig. 3. The concentrations of uranium isotopes at the different sampling points were variable, ranging between 0.8 to 70.1 ppb. The concentrations of uranium isotopes in the ground water at the Yusong area were higher than those at other regions. The increase of the concentrations of uranium isotopes at the hot springs area could be attributed to physicochemical reaction between the ground water and the mother rocks. Owing to a violent material exchange between the subterranean hot waters and the rock stratum, the erosion of the rocks has been greatly increased. Hence, uranium isotopes of underground in hot springs area easily dissolved in the ground water and increased the concentration of uranium isotopes.

Activity ratios of uranium isotopes vary with the source and can be utilized to identify the different sources of release. In general, the activity of ^{238}U is nearly equal to that of ^{234}U in natural conditions, because of radioactive equilibrium. However, the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the ground water was not unit, ranging between 1.0 to 3.3, depending on the reactive character of water and rock and residence time of water. The main mechanism which may contribute to this disequilibrium in the water includes α -particle recoil ejection of ^{234}Th into solution, preferential solution of ^{234}U due to radiation damage, and the change of ^{234}U to a more soluble U^{6+} in the associated rocks [4,5]. Also, the hot spring area samples showed lower $^{234}\text{U}/^{238}\text{U}$

activity ratios than the other region samples, although the concentrations of uranium isotopes were higher than those of other regions. This indicates that in hot springs waters much of the ^{238}U located in the lattice is dissolved and the superiority of being leached owing to daughter's recoil is reduced. Hence, the activity ratio of $^{234}\text{U}/^{238}\text{U}$ is close to that in the rock.

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4. References

1. A. P. Lopatkina, "Characteristics of Migration of Uranium in the Natural Waters of Humid Regions and their Use in the Determination of the Geochemical Background for Uranium. *Geochem. Int.*, 788 (1964).
2. Environmental Radioactivity Analysis and Measurement, JCAC Manual, Japan (1990).
3. M. H. Lee, M. Pimpl, "Development of a new electrodeposition method for Pu determination in environmental samples, In press (1998).
4. J. N. Rosholt, V. R. Shields, E. L. Garner, "Isotopes Fractionation of Uranium in Sandstone", *Science*, 139, 224 (1963).
5. J. K. Osmond, J. B. Cowart, M. Ivanovich, "Uranium Isotopes Disequilibrium in Ground Water as an Indicator of Anomalies", *Int. J. Appl. Radiat. Isot.* 34, 283 (1983).

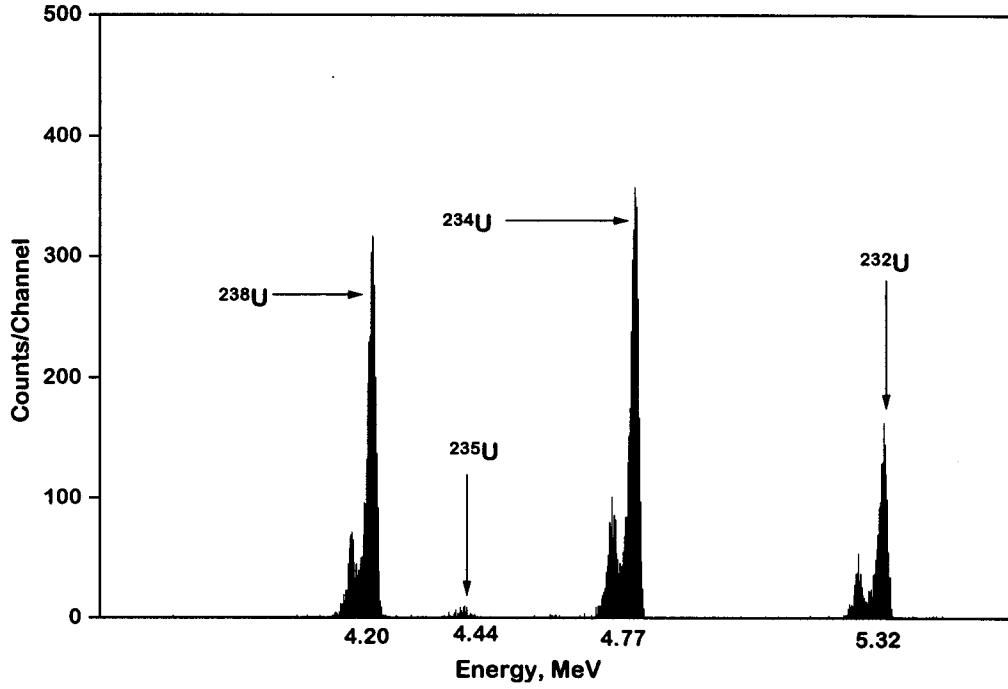


Fig. 1. Concentration of uranium isotopes in the ground water at the Yusong area

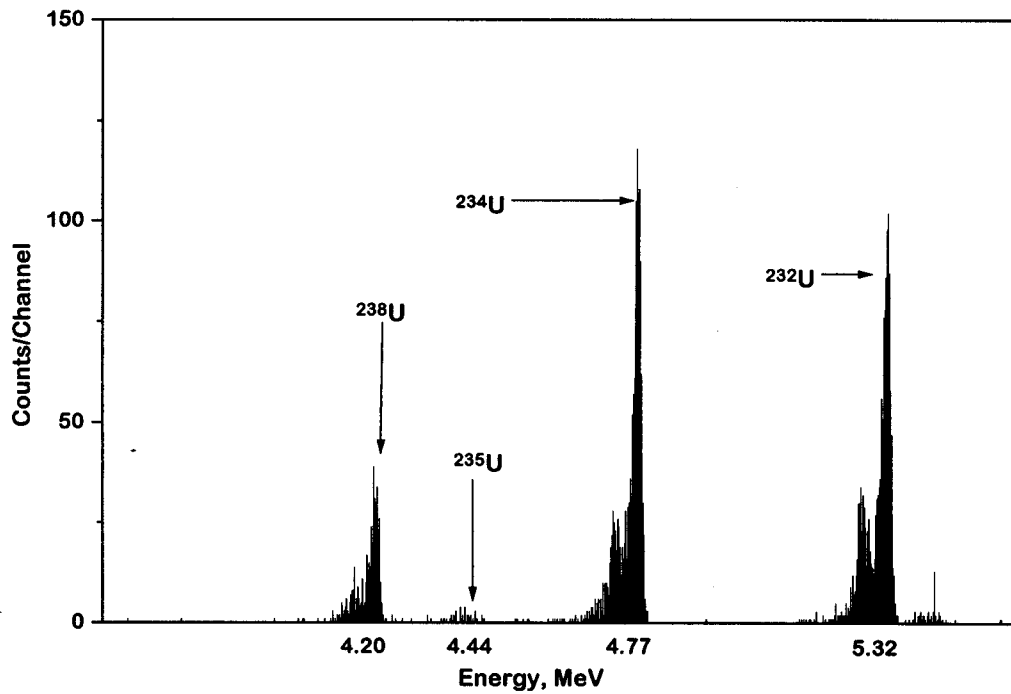


Fig. 2. Concentration of uranium isotopes in the ground water at Chungnam area

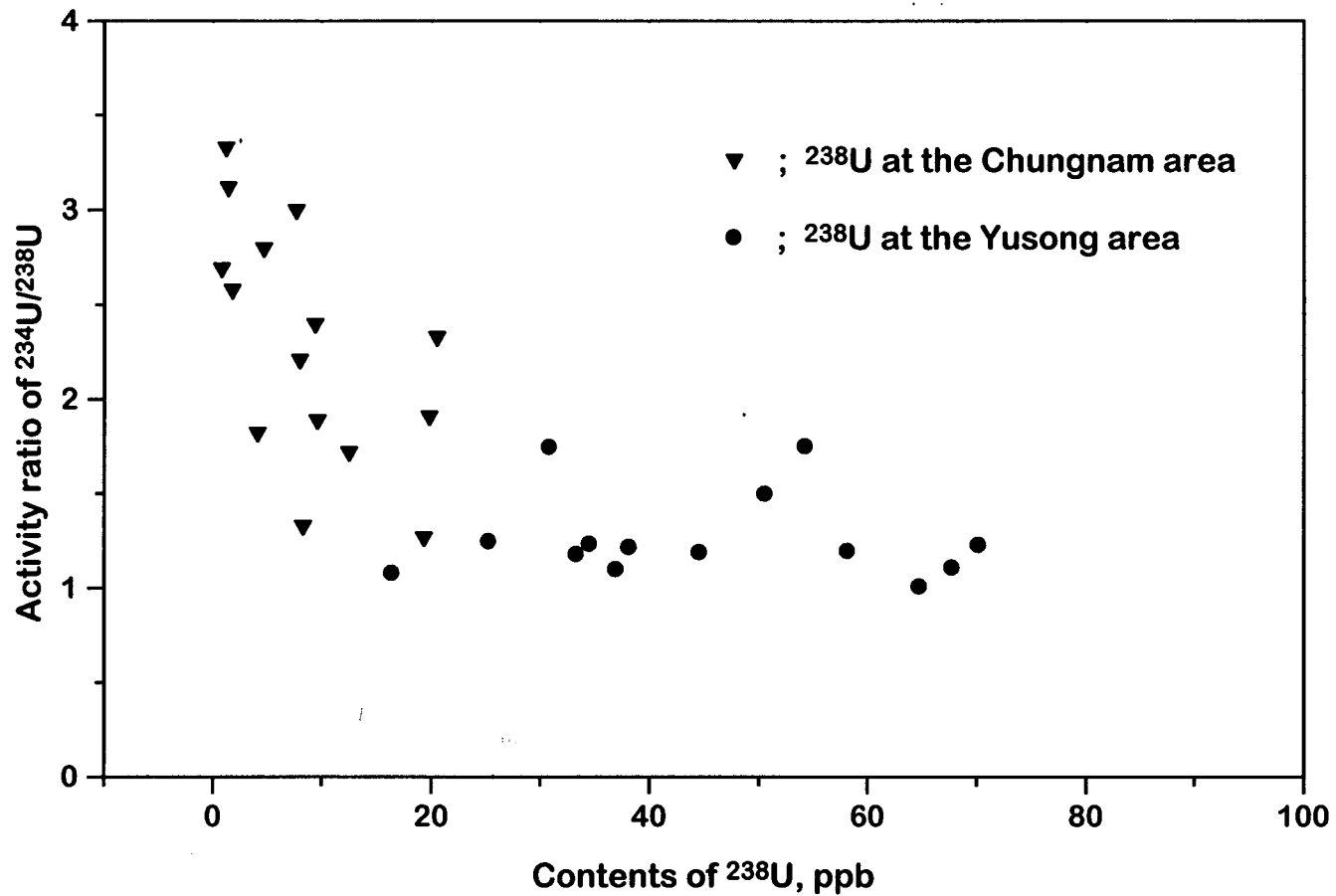


Fig. 3. Contents of ^{238}U vs. activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the ground water

22