Proceedings of the Korean Nuclear Society Autumn Meeting
Seoul, Korea, October 1998

Determination of Natural Alpha Radiation Effect: Application of ESR Spectroscopy

Young Hwan Cho, Jinho Jung , Sung Pil Hyun, and Pilsoo Hahn

Atomic Energy Research Institute
150 Dukjin-dong, Yusong-gu
Taejon, Korea 305-153

Abstract

Alpha radiation leaves a stable defect center to certain minerals. Electron Spin Resonance(ESR) spectroscopy is a powerful tool for quantifying these defect center. ESR method has been applied to trace alpha-radiation effect around the uranium ore deposit. The results show that ESR technique can be used to measure rapidly and nondestructively the defect center produced by natural alpha radiation.

Introduction

ESR spectroscopy has been widely used for radiation research in many fields, and extended to mineralogy[1]. Recently, it is being applied to neclear fields such as food irradiation, radiation dosimetry, etc.

It was known that certain minerals host a paramagnetic defect center within the lattice by irradiation. The migration of radionuclides through the uranium orebody(Koongarra) has been studied internationally under the OECD/NEA joint project[2,3]. While studying the actinide migration around uranium ore deposit, we have noticed that ESR method would be useful to see the effect of alpha-radiation of natural actinide elements (U, Th) to geomedia over geological time scale. This study is based on the ESR detection of paramagnetic defect produced by natural alpha radiation

Study Site

The uranium ore deposit studied(Koongaarra) is located in the Northern Territory of

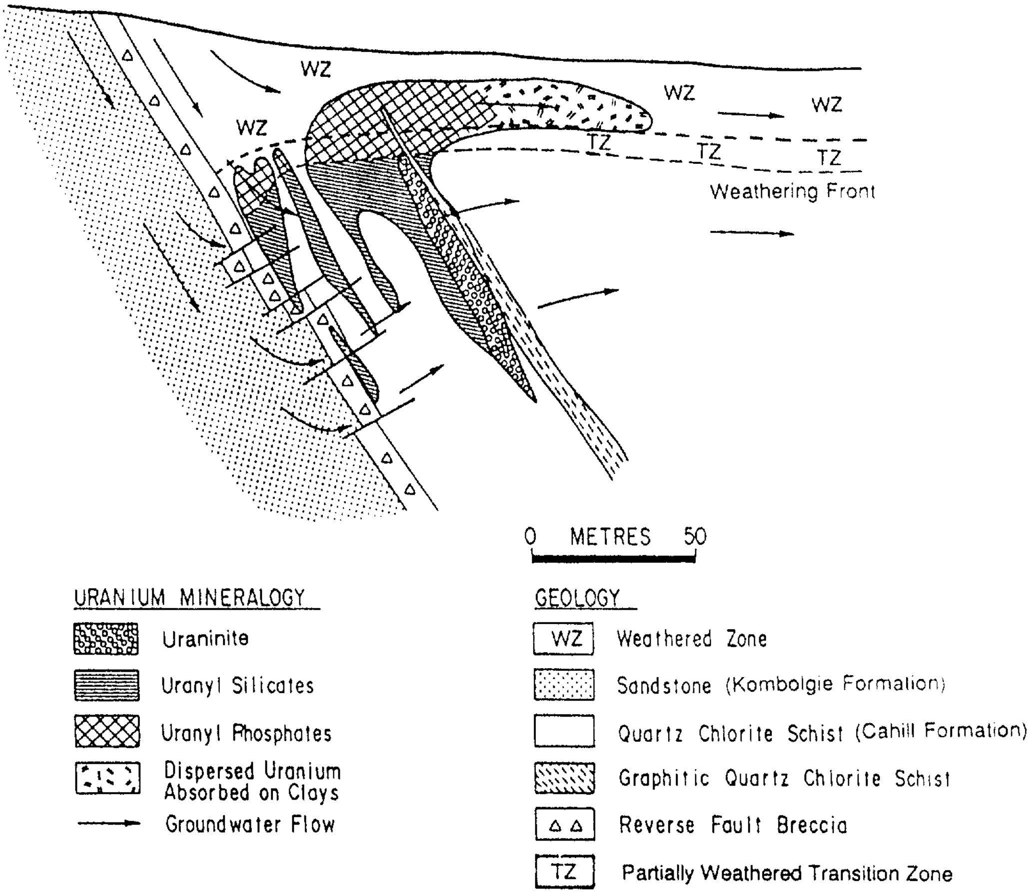

Figure 1. Vertical geological cross section of Koongarra uranium deposit.

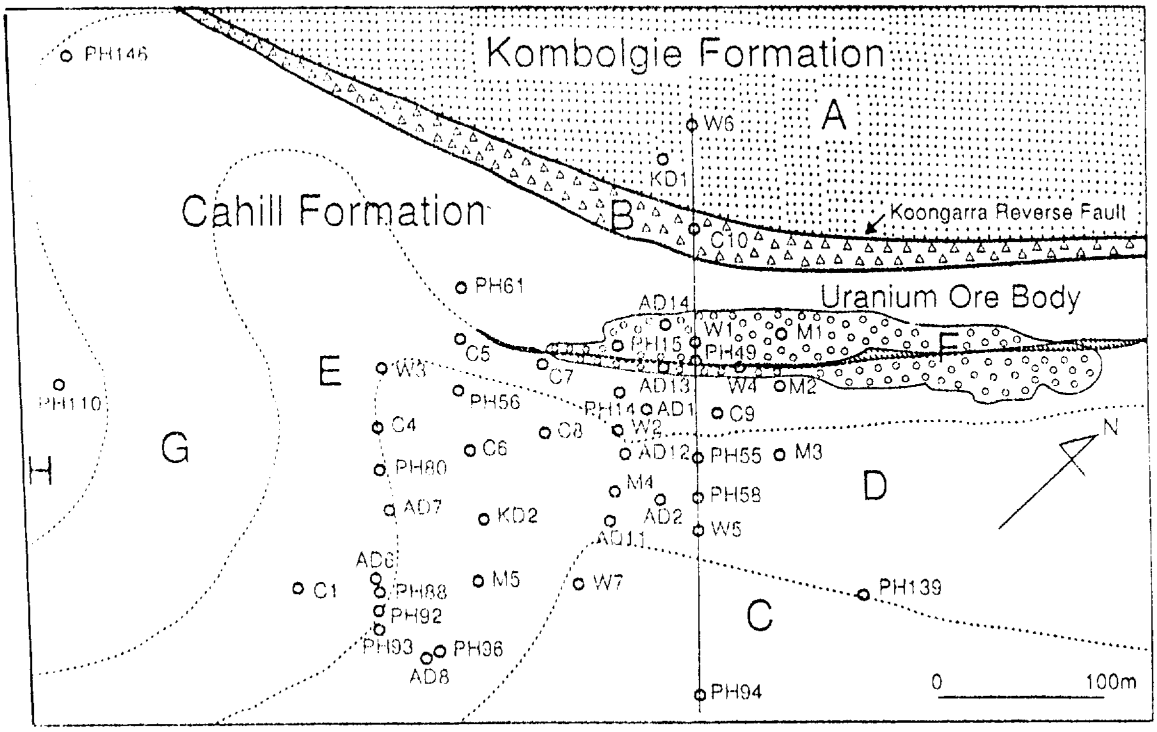

Figure 2. Geological map and location of boreholes (plan view)

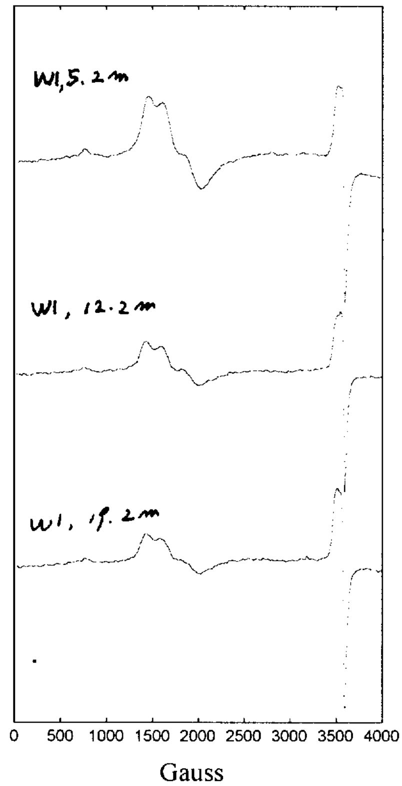

Figure 3. ESR spectra of W1 borehole samples with varying depths.

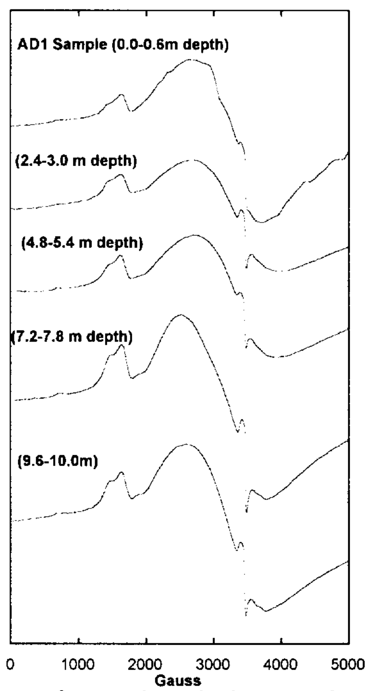

Figure 4. ESR spectra of AD1 borehole samples with varying depths

Australia, about 200 km east of Darwin. The orebody has been studied as an natural analogue for the migration behavior of actinides in radioactive waste. The orebody consists of a primary orezone in quartz chlorite schist, and a secondary orebody in the weathered zone, which was formed by the leaching of uranium from the primary orebody(Figures 1,2).

ESR Measurement

The ESR spectroscopy was used to measure the concentration of defect center in the study site samples. All ESR measurement were made at X-band (9.6 GHz) and room temperature on a Bruker EMX spectrometer. About 50 mg of samples from various locations were measured in a quartz tube without any sample pretreatment.

Results and Discussions

Figures 3,4 show the typical ESR spectra of some natural samples around Koongarra uranium orebody. In general, samples exhibit a characteristic ESR spectra consisting of two main group of resonances: (I) a group of lines at low magnetic field values, centered at g～4.2 and (II) lines at higher field values, centered at g～2.0. This resonance line(II) is attributed to lattice defect center produced by alpha-radiation. The intensity of absorption peak in g～2.0 region is closely related to defect center concentration. The resonance lines(I) are attributed to be arising from Fe^{3+} ion impurity in the mineral lattice[4]. We need to pay attention to the absorption lines at g～2.0 which arises from defect center in the samples. Figure 3,4 show ESR spectra of W1 and AD1 boreholes samples, respectively, with varying depths. The intensity of defect center peak at g～2.0 is very strong in all W1 samples. However, ESR spectra from AD1 borehole samples exhibited relatively weak compared to those of W1. W1 borehole is located inside of the primary uranium orebody zone(Figure 2). AD1 borehole is located near the orebody. This means W1 borehole samples experienced much alpha irradiation from actinide elements in the orebody than those of AD1 samples. The samples far away from the orebody exhibited virtually no defect center peaks. These results imply that the concentration of defect center is a measure of alpha radiation dose over long time scale. In general, a good correlation was achieved between defect center concentration and actinide elements(U, Th). Gamma irradiation up to 10^6 Gy had little effect on the formation of defect center, which means that defect center is created only by alpha irradiation. Some samples exhibit appreciable defect center concentration even in the

absence of actinide concentration, which can be explained by the alpha irradiation in the past time. This means that some minerals in the sample act as a sensitive dosimeter to external alpha-irradiation over geological time scale. The possible mechanism of hosting defect center is by close contact of actinide elements with geomedia, such as sorption process. The detailed analysis of ESR defect center data and actinide concentration together with mineralogical, geological studies may reveal the long-term migration behavior of actinides in the nature. That is an aim of the natural analogue study of long-term safety from high-level radioactive waste dispoal.
Further study is underway.

Conclusions

- It was found that geological samples are keeping the record of past migration history of actinides within the defect center.
- ESR spectroscopy was useful to quantify defect center produced by natural alpha irradiation.
- ESR method may provide a unique tool for studying past history of actinide migration in the geosphere over geological time scale.

Acknowledgement

This project has been carried out under the Nuclear R&D Program by MOST.

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