*Journal of the Korean Magnetic Resonance Society* **2001**, *5*, 13-18

# Determination of Alpha Defect Center in the Nature Using EPR Spectroscopy

Young Hwan Cho*, Sung Pil Hyun, and Pilsoo Hahn

Atomic Energy Research Institute, 150 Dukjin-dong, Yusong-gu,
Taejon, Korea 305-353
*Received April 4, 2001*

***Abstract :*** Natural alpha radiation produced a stable defect center to certain minerals. Electron Paramagnetic Resonance(EPR) spectroscopy is a powerful tool for quantifying this defect center. EPR method has been applied to trace alpha-radiation effect around the uranium ore deposit. The results show that EPR technique can be used to measure rapidly and nondestructively the defect center produced by natural alpha radiation. In general, a good correlation was achieved between defect center concentration and actinide elements(U, Th). These results imply that the concentration of defect center is dependent on the alpha radiation dose over long time scale.

## INTRODUCTION

EPR spectroscopy has been widely used for radiation research in many fields, and extended to mineralogy.[1] Recently, it is being applied to nuclear fields such as food irradiation, radiation dosimetry, etc. However, studies on the defects produced by alpha radiation are extremely rare.

It was known that certain minerals host a paramagnetic defect center within the lattice by irradiation. The migration of radioactive elements through the uranium orebody(Koong-arra) has been studied internationally under the OECD/NEA joint project.[2] While studying the actinide migration around uranium ore deposit, we have noticed that EPR 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 EPR detection of paramagnetic defect produced by natural alpha radiation.

## EXPERIMENTAL

***Study Site :*** The sample of Koongarra uranium ore deposit was collected from the Northern

*To whom : yhcho@kaeri.re.kr

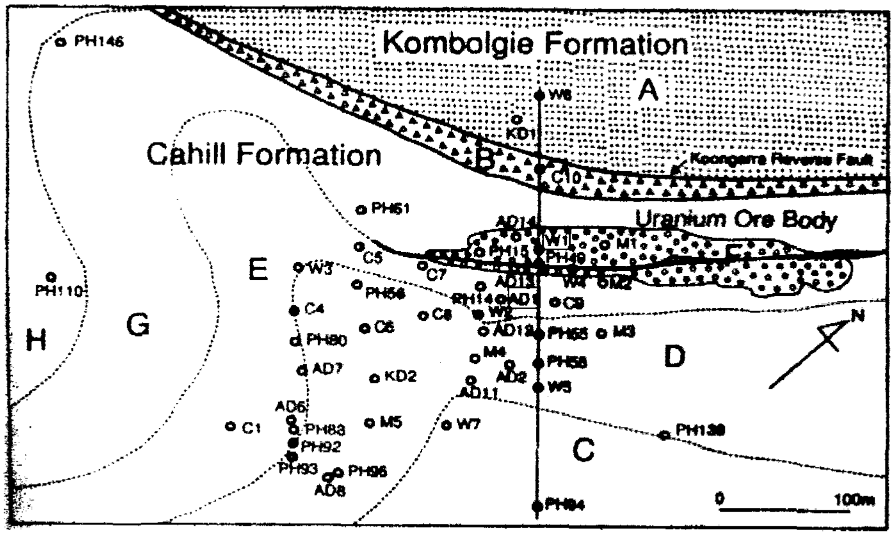


**Fig. 1.** Surface geological map and the location of boreholes in Koongarra uranium orebody area.

Territory of Australia, about 200 km east of Darwin. The orebody has been studied as a 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). The chemistry and mineralogy of rocks and soils in this region is well documented.[3,4]

***EPR Measurement :*** The EPR spectroscopy was used to measure the concentration of defect center in the site of studied samples. All EPR measurements 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

Although the EPR method is restricted to a certain range of paramagnetic species, it can yield a wide range of detailed information relating to the occurrence and location of paramagnetic substitutional impurities and lattice defect (trapped hole or electron centres) inclays. Figures 3,4 show the typical EPR spectra of some natural samples around Koongarra uranium orebody. In general, samples exhibit a characteristic EPR spectra consisting of two main groups of resonance: (I) a group of lines at low magnetic field values, centered at $g \sim 4.2$ and (II) lines at higher field values, centered at $g \sim 2.0$. This resonance line(II) is attributed to lattice defect center produced by alpha-radiation. The intensity of

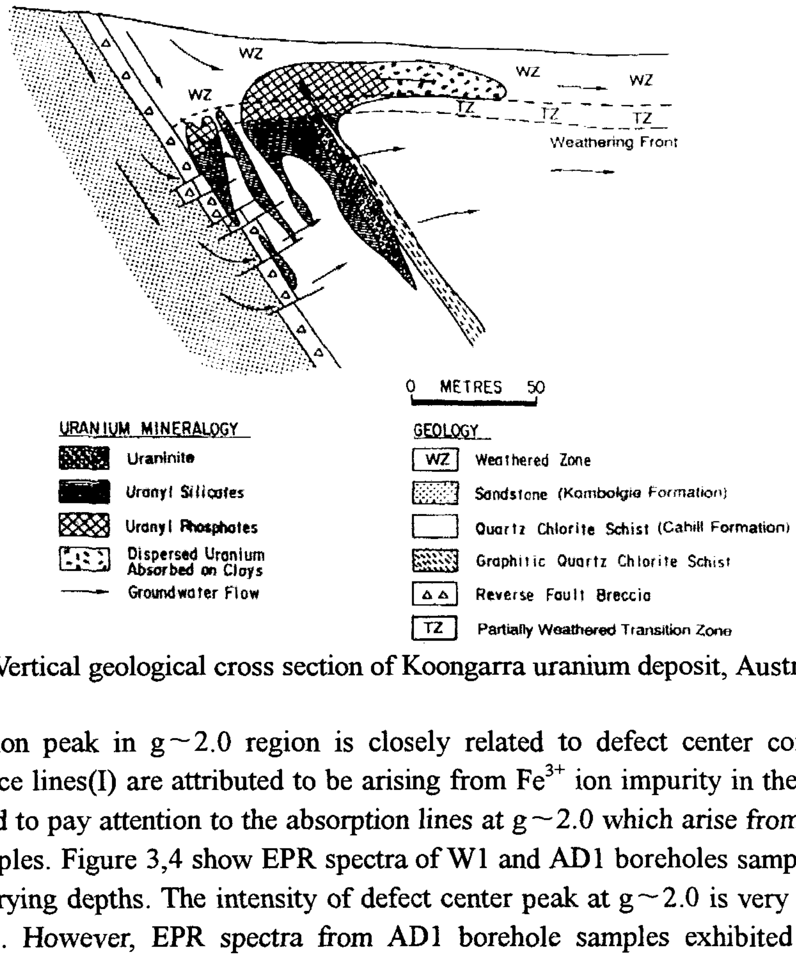


**Fig. 2.** Vertical geological cross section of Koongarra uranium deposit, Australia.

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.[5] We need to pay attention to the absorption lines at g~2.0 which arise from defect center in the samples. Figure 3,4 show EPR 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, EPR spectra from AD1 borehole samples exhibited relatively weak intensity 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 had more 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 dependent on the 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 using Co-60 source 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 caused 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 mostly possible mechanism of hosting defect center is by close contact of actinide ele-

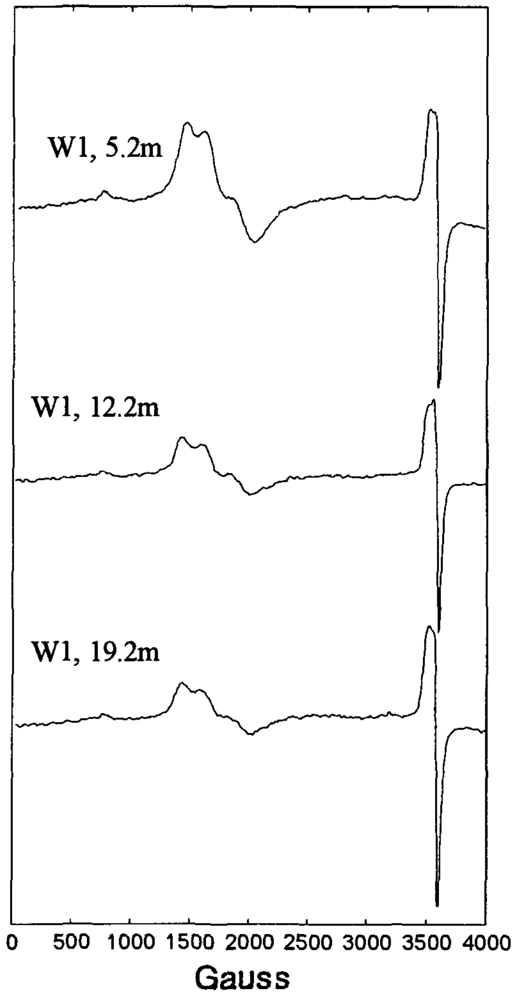


**Fig. 3.** EPR spectra of W1 borehole samples with varying depths

ments with geomedia, such as sorption process. The detailed analysis of EPR defect center data and actinide concentration together with mineralogical, geological studies may reveal the long-term migration behavior of actinides in the nature. This means that EPR spectroscopy can provide a unique tool for studying past history of actinide migration in the geosphere over geological time scale. That is an aim of the natural analogue study of long-term safety from high-level radioactive waste disposal. Further study is underway.

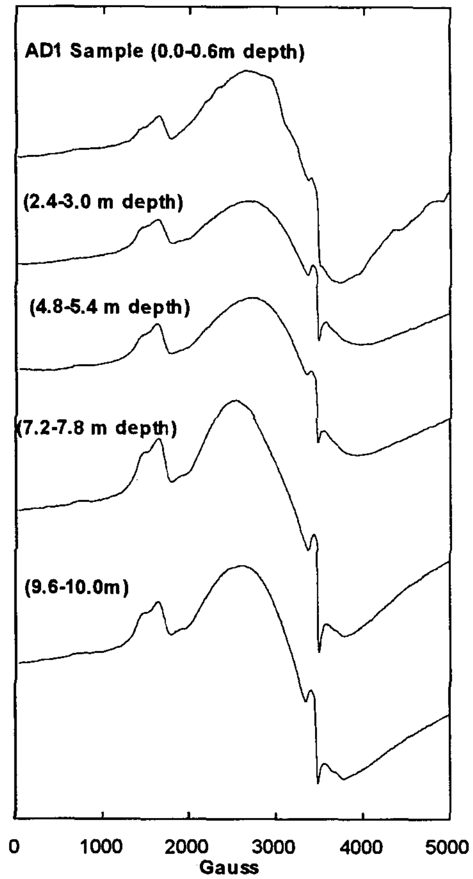


**Fig. 4.** EPR spectra of AD1 borehole samples with varying depths

## CONCLUSIONS

The following conclusions can be drawn from this study:

- It was found that geological samples are keeping the record of past migration history of actinides within the defect center.
- EPR spectroscopy was useful to quantify defect center produced by natural alpha irradiation.
- EPR 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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