

## Examination on geological potential for long-term supply of uranium resources

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## Introduction

According to "Long-term Energy Supply and Demand Outlook" released on July 2015, the dependence on the nuclear power plants is planned to be 20 to 22% of the electric power supply in FY2030. Because uranium is limited resource, it is necessary to consider the long-term supply.

Information on uranium resources is periodically reported by international and national organizations. For example, OECD/NEA and IAEA have jointly prepared periodic updates on world uranium resources, production and demand. These updates have been published in what is commonly known as the "Red Book" (OECD/NEA-IAEA, 2014). In this presentation, supply and demand of uranium are reviewed based mainly on the Red Book, and then future possibility on increase in uranium resources in Canada and Australia, the principal supplying countries, is discussed.

## Resource and Demand

According to Red Book 2014 edition, total identified resources (reasonably assured and inferred) as of 1 January 2013 amount to about 7.6 million tonnes U in the less than USD 260/kgU (USD 100/lb U<sub>3</sub>O<sub>8</sub>) cost category. Annual uranium requirements was 61,600tU in 2012. Identified resources are sufficient for over 120 years based on the 2012 requirement. Annual uranium requirements by 2035 are projected to rise to between 72,205tU and 122,110tU, so that relation between supply and demand could be changed in future.

## Uranium deposit type

OECD/NEA-IAEA (2014) classifies uranium deposit types into following 15 categories, which are arranged according to their approximate economic significance;

1. Sandstone deposits
2. Proterozoic unconformity deposits
3. Polymetallic Fe-oxide breccia complex deposits
4. Paleo-quartz-pebble conglomerate deposits
5. Granite-related
6. Metamorphite
7. Intrusive deposits
8. Volcanic-related deposits
9. Metasomatic deposits
10. Surficial deposits
11. Carbonate deposits
12. Collapse breccia-type deposits
13. Phosphate deposits
14. Lignite and coal
15. Black shale

## Potentiality of increase in uranium resources

Canada produced 8,998tU in 2012. All amounts of uranium were mined from the Proterozoic unconformity deposits. The Proterozoic unconformity deposits are generally large scale and are characterized by their high ore grades. For example, present principal uranium mines (Cigar Lake and McArthur River deposits) contain more than 100,000tU with the ore grade of more than approximately 15%U<sub>3</sub>O<sub>8</sub>. The plane extension is 1,700m long and 30m wide in the McArthur River deposit, and 1,950m long and 20 to 100m wide in the Cigar Lake deposit. The deposits locate in the

depth of more than 400m below surface.

Many uranium deposits have been discovered by extensive exploration activities since 1960s.

However, many target areas remain to be unexplored, because the distribution of uranium deposits is so narrow and because drilling is only method to detect the deposits in deep underground. After the discovery of the McArthur River deposit in 1989, excellent deposits have been discovered such as the Millennium deposit in 2000 and the Phoenix deposit in 2008. New findings of uranium occurrences are continuously reported (e.g. Government of Saskatchewan, 2015). Taking these points into consideration, the discovery of new deposits and the increase in resources may be quite possible. Australia produced 7,009tU in 2012. Uranium was mined from the Proterozoic unconformity, polymetallic Fe-oxide breccia complex and sandstone deposits. Development of the previously discovered deposits would be proceeded. Discovery of new deposits is quite possible by progress of exploration, because Australia has much unexplored land.

#### Reference

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OECD/NEA-IAEA, 2014, Uranium 2014: Resources, Production and Demand.

Keywords: Uranium Resource

## Geosphere Stability Project

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### 1. Japan Atomic Energy Agency

#### Introduction

Geosphere stability project is being carried out by Japan Atomic Energy Agency in order to establish techniques for investigation, analysis and assessment of the long-term evolution of geological environments, taking into account long-term geological phenomena such as volcanism, faulting, uplift, denudation, climate change, and sea-level change. This project is being conducted under a contract with Agency of Natural Resources and Energy, part of Ministry of Economy, Trade and Industry of Japan as part of its R&D supporting program for developing technology of geological disposal of high-level radioactive waste.

The geological disposal secures the safety in the time scale over tens of thousands of years by the multiple barrier system which consists of an engineered barrier and a natural barrier together. Crustal movement in and around Japanese Islands is active, because Japanese Islands is located in the mobile belt. Therefore it is important that identification of the natural phenomenon that may have a remarkable influence on geological disposal system and understanding of the behavior pattern and scale of the geological environments with the natural phenomenon. The project has the development of a numerical model "Geological-Evolutionary Model" expressing a change of the geological environments and the development of "Innovative Elemental Technique" that is elemental technology required for model construction.

#### Overview of Project

##### Geological-Evolutionary Model

The purpose of the Geological-Evolution Model is to establish modeling techniques, which can express a long-term change of the geological environments. In addition, the project contains the R&D such as the methodology for verification and an uncertainty estimate of the models, and the visualization technology of the model.

In the R&D, FEP (Features, Events and Processes) analysis and scenario development for geological-evolutionary modeling of mountain area and plain area have been carried out. Paleo-hydrogeological model was constructed in consideration for long-term geological environments such as topography, geology and surficial environment, and spatial distribution of long-term stability of groundwater flow conditions were numerically assessed with the standard deviation and the variation coefficient.

##### Innovative Elemental Technique

The Innovative Elemental Technique consists of Provenance analysis techniques, Groundwater recharge estimation techniques, Carbonate mineral measurement techniques and Crustal movement estimation techniques as the technique required for the modeling on a long-term scale and the analysis evaluation.

The technique to identify a supply source of the sediment using the ESR signal of the quartz, technique to estimate a river spillage in consideration of the topography, technique to analyze the microscopic area of the carbonate mineral, distorted speed distribution based on the displacement speed of the active fault were developed in the R&D.

#### Future studies

In future work, geomorphological, geological, hydrological, and geochemical models will be integrated, and uncertainties of geological-evolutionary modeling will be analyzed based on difference in geological environments of the mountains and plains. In addition, it is necessary for continuation of the development of the elemental techniques and to develop the modeling method using the techniques.

Keywords: Long-term stability of the geological environments, Geological-Evolutionary Model, Innovative elemental techniques, High-level radioactive waste, Geological disposal

## Downward erosion near the recent shoreline since Late Pleistocene

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We did the risk analysis of the downward erosion of about future 100,000 years for geological disposal. We have investigated the base of the alluvium in Japan. The depth of alluvium is a good indicator of the downward erosion caused by both river process and sea-level change. On the basis of these data, we showed the relationship between the maximum depth of downward erosion near the recent shoreline since Late Pleistocene (y) and the uplift from Late Pleistocene to the recent (x) in the uplift areas are as follows:  $y > -x - 100$ . Based on the uniformitarian view, we can estimate the maximum depth of downward erosion of the future in the uplift area using the above relationship. However, evaluation of subsidence areas is a future challenge.

[ Reference ] Hataya R., Yanagida M., Torigoe Y. and Sato M., Journal of the Japan Society of Engineering Geology, Vol.55, No.1, 2016 (in Japanese with English abstract).

Keywords: downward erosion, alluvium, risk analysis, geological disposal

## Grouping of Japanese Islands based on spatial-temporal tendencies of natural phenomena

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Post closure safety of a HLW repository is assessed by dose calculations based on comprehensive scenarios which describe likelihood of natural events and potential impacts on the functions for isolation and containment of radionuclides. In principle, likelihood of natural events which may cause adverse impacts on the safety functions, such as fault activities, is evaluated by extrapolating the past tendency into the future. The extrapolation assumes that the tendency of natural events in the past have been maintained no shorter than the period of assessment in the future. It has also been acknowledged that the tendencies of natural phenomena vary depending on the local tectonic evolutions in Japan. NUMO prepares a technical report which develops a pre-selection site-specific safety case where no specific area is specified. Spatial-temporal distribution and tendencies of natural phenomena in Japanese Islands were compiled and grouped to provide basic information for development of natural event scenarios.

The following results were derived based on the latest literature information. The island arc of Japan is divided into the forearc/backarc areas of the volcanic fronts with respect to the existence of Quaternary volcanoes. The backarc areas are subdivided into the closer area from the volcanic front where Quaternary volcanoes are densely distributed and the farther ones where the volcanoes are sparse or absent. The volcanic fronts have almost been fixed since the Pliocene or older ages. Various volcanic activities and spatial-temporal evolutions are identified in the backarc areas during this period. Active faults tend to distribute along the boundary between mountains/hills and basins/planes. Areas are also divided into three with respect to the spatial density of active faults (high/moderate/low). Various styles of faulting and periods of activities are identified within each group. With respect to the average uplift rate during the last 100,000 years, areas with high uplift rate and that with low rate or subsidence are identified. During the longer period of Quaternary, some areas indicate possible occurrence of inversion from subsidence to uplift within several tens of hundred thousand years.

Some implications for the scenario development are described below. Areas in the forearc areas do not have to expect any volcanism, those in the backarc areas need to consider occurrence of volcanic activities, and those far away from volcanic fronts require careful assessment including magma generating processes. With the assumption that the higher the density of fault distribution is, the more likely the area is to be influenced by disruptive events, such area needs to consider the occurrence of events such as branching, extension and induced movement of active faults. Areas with possible inversion from subsidence from uplift during the Quaternary need careful assessment even though the uplift rates during the last 100,000 years are low.

This kind of grouping will help to understand relative position of a certain area within Japan and provide basic information on future occurrence of natural events for scenario development.

Keywords: geological disposal, volcano, fault, uplift/subsidence, likelihood, scenario

## Groundwater flow simulation related to flow barrier fault for geological disposal

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### Introduction

Not a few fault or shear zone have fault core of its center with damaged zone on its both side (Evans et al.1997). There are lots of faults in Japan since it is located at convergent plate boundary. If there is a fault block surrounded by low permeable fault, groundwater cannot easily flow into the fault block from its surroundings, causing a huge drawdown in the block, whilst very small drawdown at the outside of the block. On the other hand, the ground water recovery could be very slow of its recovery phase, causing long duration of unsaturated field.

Previous our study are as follows; a) simple simulation of groundwater drawdown behavior due to a withdrawn of ground water from the shaft, b) distribution characteristics of the fault block in Japan, and c) relationship between the trace length of fault and thickness of the fault core. As a result, for the a) above, a large difference occurs in drawdown inside and outside of the fault block by simple simulation in the case of 4 orders of magnitude difference in hydraulic conductivity of the host rock and the fault. In particular, seldom drawdown has calculated at the outside of the fault block. For the b) above, the area of the fault block based on the seamless geological map in Japan (AIST) is about 2 % of the entire Japanese area. Moreover, the area of a fractured rock such as pre-Neogene deposit and plutonic rocks is more than 90% for the area of the fault block in Japan (Takeuchi other 2015). In addition, the histogram of the area of the fault block in each district in Japan is almost the same as the area of geological disposal facilities (Takeuchi et al., 2015). For the c), we found out general relationship between the thickness of the fault core and the trace length of the fault based on the existing data. Namely the thickness of the fault core has plateau as about 20cm against the trace length of few hundred meters or more (Takeuchi et al., 1995). In this study, we analyzed drawdown behavior inside and outside the fault block by changing the hydraulic conductivity and thickness of the fault, respectively.

### Result

In the case of the hydraulic conductivity is  $1E-10$  (m/s), the drawdown is uniformly about 155m from the ground level for the 50m in fault thickness, and about 250m for the 80 and 100m, respectively. In the case of the hydraulic conductivity is  $1E-11$  (m/s), the drawdown is uniformly about 355m for each thickness of the fault. Moreover, in the case of the hydraulic conductivity is  $1E-12$  (m/s), the drawdown is uniformly about 355m for the 50 and 80m, and about 455m for the 100m. In any case, the obvious drawdown was not observed at outside of the fault block.

### Conclusion

The simulation has revealed that the drawdown behavior inside the fault block could vary with the hydraulic conductivity of the fault and its thickness. Moreover, it has been suggested that the ground water level could be deeper than the shaft depth in some cases. In the future, it should be simulated the ground water behavior in the recovery phase and fault property such as hydraulic conductivity of the fault core for the major fault in the field.

Keywords: Fault Core, Flow Barrier Effect, Fault Block, Groundwater flow, Geological Disposal

## Alteration process along a small-scale fault at the depth of -500m in the MIU site

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In order to understand the hydrological feature of underground environment, it is important to clarify the analogous history of water-rock interaction and function of flow-path. In particular, it is considered that fractures and faults play an important role on elemental migration in underground environment. Although the example of single fracture has been studied, the hydrological feature of a small-scale fault is still not well understood. Therefore, structural, mineralogical and geochemical characteristics and formation process of a small-scale fault and alteration process along the fault with fracture fillings at -500 m identified in Toki granite distributed in Central Japan was investigated. Investigation was mainly conducted by observation of a borehole core sample excavated in the JAEA's Mizunami Underground Research Laboratory. Various studies on geology, geochemistry and hydrology in and around the fault were carried out including the data analysis provided by hydrological borehole logging. Based on the results, faulting and alteration processes due to groundwater flow along the fault are considered as follows: 1; Intrusion of the pluton and its cooling.2; Formation of the fault and associated fractures.3; Hydrothermal alteration through the fault and fractures in deep underground.4; Calcite precipitation and smectite formation by relatively low temperature meteoric groundwater in shallow underground. This result implies that such investigations can be applied for to know the history of water-rock interaction and hydrological feature of a fault and associated fractures.

Keywords: fault, Toki Granite, alteration



## Development of Technology on Long Term Monitoring in the Borehole and it's Application

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In Japan, there are three stages of site-selection for the deep geological disposal of HLW. Literature surveys, followed by preliminary investigations (PIs) and finally, detailed investigations (DIs) are carried out in successive selection stages. Underground survey facilities are constructed in the final selection stage. Geological, environmental and radiological conditions might be affected by the construction of these underground survey facilities and the final depository. It is necessary to obtain the initial-state conditions of pore water pressure and water chemistry through long term monitoring of boreholes during the PIs in order to estimate the influence on geological, environmental and radiological conditions by the construction of underground facilities and the final depository.

Since 2000, CRIEPI has been conducting a project on the development of directional drilling and measurement/logging technologies. In FY2006, we began applying these technologies to the Omagari fault, distributed at the Kami-Horonobe area in Horonobe-town, Hokkaido (site no. HCD-3). After drilling the borehole of length 1000 m, a steel pipe was inserted to support the borehole wall. In FY2013, considering the hydro-geological conditions along the borehole, a Standpipe Multi Packer (SPMP)-type monitoring system was installed in the borehole. In FY2014, we began to measure the pore water pressure at three measurement intervals and in FY2015 ground water collection for water chemistry and ground water dating was started.

The data recorded on data loggers were collected every 3-4 months on site and the batteries were replaced at the same time. At the PI stage, we assume that long term monitoring for several boreholes will be achieved in vast areas where power supply and/or communication networks are not available. Thus, we are developing an automatic data acquisition system for long term monitoring, which enables us to access the data remotely.

The research and development for this study are funded by the Agency for Natural Resources and Energy, a subsidiary of the Ministry of Economy, Trade and Industry (METI) and have been carried out as a program for the systematization of the controlled drilling technology and measurement method used in boreholes. In situ drilling and surveying were conducted in collaboration with the Horonobe Underground Research Center of the Japan Atomic Energy Agency (JAEA).

Keywords: Pore Water Pressure, Water Chemistry, Stand-alone Data Acquisition System

## In-situ tracer experiments for granitic rock mass

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For safety assessment for geological disposal of high-level radioactive wastes, it is very important to estimate transport properties of host rock accurately. Therefore the authors have developed an apparatus for in-situ tracer test and the method to evaluate transport properties from the results of tracer tests. Tracer tests were conducted with the testing apparatus at an experimental site in granite area in Japan.

The experimental site was a horizontal gallery at -300 m Stage in the Mizunami Underground Research Laboratory of Japan Atomic Energy Agency (JAEA). Tracer tests were conducted at two boreholes, 12MI31 and 13MI37, excavated from the tunnel wall. From the observation of borehole walls with a borehole TV camera, the response of pore water pressure at 12MI31 during excavation and hydraulic test of 13MI37, the fracture intersecting at 21.90 mab (meter along the borehole) with 12MI31 is probably identical with the one at 23.14 mab with 13MI37. The distance between the boreholes was 2.95 metres. According to the prior single-hole hydraulic tests, the transmissivity of the target fracture was  $6.9 \times 10^{-8}$  to  $1.1 \times 10^{-6}$  m<sup>2</sup>/sec. Fracture filling materials such as chlorite, clay and pyrite were observed in rock core samples.

Single-hole tracer test is a test where injection and recovery of tracers was conducted at the same section of a single borehole. At first groundwater sampled at the test site was injected into a borehole. After the flow field in the fracture reached a steady state, tracer solution was injected into the borehole. Subsequently groundwater was injected into the borehole as chaser. Finally pumping was conducted at the borehole and tracers were recovered.

Dipole tests were conducted as cross-hole tracer tests. At first sampled groundwater was injected into a borehole and pumping was conducted at another borehole. After the flow field reached a steady state, tracer solution was injected into a borehole and the tracers were recovered at the other borehole. Injection rate was set at one fifth or one tenth of pumping rate to attain high recovery rate.

Fluorescent dyes, Uranine, Amino-G acid, deuterium and iodine as non-sorbing tracers and rubidium and barium as sorbing tracers were used. Water samples were taken from the pumped water with fraction collector and analyzed in a laboratory. Concentration of uranine was measured with online sensors.

During single-hole tracer tests, in the case where pumping was started just after injection of chaser ended, recovery rates of non-sorbing and sorbing tracers were 80 to 90 percent and 70 to 80 percent, respectively. By contrast, in the case where pumping was started some time after injection of chaser ended, recovery rates were decreased from the influence of the background groundwater flow. In both cases, peak concentrations of sorbing tracers were lower than ones of non-sorbing tracers.

During crosshole tracer tests, the recovery rates of non-sorbing tracers were 54 to 63 percent at 10 hours after the tests started in consequence of the background groundwater flow. The recovery rates of adsorptive rubidium were 24 to 25 percent after 30 hours. Compared to the non-sorbing tracers, decay and time lag of peak concentration were observed from the influence of adsorption on rock mass. The recovery rates of more strongly adsorptive barium were only 5 percent after 30 hours. Concentration of barium at the pumping borehole was nearly equal to the background concentration throughout the testing time and no significant breakthrough curves could be obtained. Fracture aperture, dispersion length and distribution coefficients on rock matrix will be evaluated

through numerical simulations on the basis of breakthrough curves.

This study was commissioned by the Agency for Natural Resources and Energy in the Ministry of Economy, Trade and Industry. The tracer tests were conducted as a part of a collaborative research with JAEA.

Keywords: tracer test, granite, fracture

## Influences of faulting and hydrothermal alteration on matrix diffusion in granitic rocks

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## 1. Japan atomic energy agency

Matrix diffusion is an important process for controlling mass transport in crystalline rocks such as granitic rocks. The matrix diffusion rate is controlled by porosity. The influence of hydrothermal alteration on matrix diffusion is one of the important issues in Japan, because the porosity in plutons in an orogenic belt such as Japan has the potential to increase associated with faulting and / or hydrothermal alteration. Thus, this study focuses on the relationship between faulting, hydrothermal alteration and matrix diffusion.

Six granitic rock samples (three samples are altered rocks and others are non-altered rocks) are obtained from ground level -300m in Mizunami Underground Research Laboratory (MIU) located in Mizunami city Gifu prefecture, central Japan. Detailed observations and through-diffusion experiments to use uranium as a tracer material provide two results, 1) Porosities and effective diffusivities of samples located near faults show high values compared with samples from sites which are unaffected by faulting and 2) The decrease in porosities and effective diffusivities of samples located near faults is related to the increase in the percentage of secondary minerals.

This result suggests that faulting and hydrothermal alteration have potentials of controlling the distribution of microscopic voids and having an effect on the matrix diffusion rates.

Keywords: matrix diffusion, fault, hydrothermal alteration, granitic rocks

## Spatial characterization of permeability distribution around faults through in-situ permeameter measurements

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In safety assessment for geological disposal of high-level radioactive waste, radionuclide migration caused by the action of groundwater is examined (e.g., JNC, 1999), so it is necessary to understand groundwater flow properties around disposal site. Faults are known to behave as conduit, barrier, or combined conduit-barrier system (Caine et al., 1996) dependent on their internal structure. Because faults can be dominant pathways of groundwater flow and mass transport, their hydrological properties are significant. However, information on spatial distribution of permeability around faults is limited and influences of the spatial distribution on groundwater flow or mass transport have not been sufficiently understood. This research aims to elucidate the spatial distribution of rock permeability around a fault and to reflect them to hydrogeological model around the fault. Study site was research tunnels of Mizunami Underground Research Laboratory, Japan Atomic Energy Agency (Mizunami, Gifu, Japan). A fault referred to as "Main shaft fault" is distributed adjacent to the main shaft and dip of the fault is nearly perpendicular at 300 meters deep (Tsuruta et al., 2010). Several horizontal tunnels extend from the main shaft at different depths, and this is advantageous to examine relationships between permeability and distance from the fault core. Permeability of Toki granite exposed on the tunnel wall was measured along horizontal tunnels at 200, 300, and 500 meters deep using gas permeameter (Mini-Permeameter MP-401, TEMCO, Inc.). In the presentation, spatial distribution of permeability around the fault is shown and cause for the distribution is discussed.

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Keywords: Fault, Permeability, Spatial distribution, Groundwater, Hydrogeological model

## Experimental study on correlation of permeability with physical and chemical properties using Toki granite samples

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Understanding of hydraulic property of rock mass is essential to implement a safe geological disposal of high-level radioactive waste for long term. In order to grasp hydraulic property of rock in detail, we clarified a spatial variation of permeability of Toki granite based on experiment and examined the relation with rock physical properties.

The samples are cores collected from 3 boreholes in Tono area (Mizunami Gifu, Japan) drilled by Japan Atomic Energy Agency. This area is mainly composed of Toki granite. First, permeability was measured using the gas permeameter (Mini-Permeameter MP-401, TEMCO, Inc.). Furthermore, the measurement of P-wave velocities, micro crack properties and chemical composition were performed. As the result, the strongly positive correlation between permeability and cumulative length of micro crack was clarified. Moreover, permeability and P-wave velocity has negative correlation. On the other hand, the correlation between micro crack properties and P-wave velocity was not strong. The reason of these results is that although both of permeability and P-wave velocity are affected by the aperture formed by crack in rock mass, P-wave velocity involves the effect of rock matrix as a medium.

Rock core samples were classified into two groups based on the Mn/Fe ratio. Mn/Fe ratio can be regarded as an indicator to stand for the lithofacies. Mn/Fe ratio corresponded with P-wave velocity variation. The correlation between permeability and P-wave velocity within each group became stronger than the case that all samples were considered.

From these results, the relationship between the permeability and physical properties of Toki-granite is discussed and suggested the method to estimate the hydraulic property from physical properties.

Acknowledgement: We would like to express our sincere thanks to the co-researchers of Japan Atomic Energy Agency for their supports and many constructive comments for this study.

Keywords: Chemical composition, Micro crack, Permeability, P-wave velocity, Toki granite

## Temporal changes of radiocesium outflow in mountainous forest of the Abukuma Mountains, Fukushima

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This paper presents outflow characteristics of Cs-137 in mountainous forest of the Abukuma Mountains, Fukushima, during 2013-2014. Cs-137 deposition via throughfall, stemflow, and litterfall processes was estimated to be on the order of  $10^3$  Bq m<sup>-2</sup>, and the outflow of Cs-137 via surface washoff was estimated to be on the order of  $10^2$  Bq m<sup>-2</sup> from April 2013 to December 2014 in the experimental plots installed in deciduous broad-leaved and cedar forests in the Abukuma Mountains. Cs-137 inventories of forest soil down to a level of 1 cm were decreasing in ridge and valley-bottom soil during the period from December 2012 to October 2014. The inventories in mountain slope showed both decreasing and increasing tendencies because of the heterogeneous transportation of Cs-137 via surface washoff on the slope. The results of outflow rate simulations using the SACT model developed in the Japan Atomic Energy Agency indicate decreasing tendency accompanied with a deeper penetration of Cs-137 into soil profile. Thus, the forest floor in the mountainous forest seems to be a sink of radiocesium contamination rather than a source for the contamination of the other ecosystems.

Keywords: Fukushima Dai-ichi Nuclear Power Plant accident, radiocesium, mountainous forest

## Comparison of sorption coefficients between powder and intact solid phase: Case study with $^{133}\text{Cs}$ and $^{87}\text{Sr}$ on pumice tuff

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Adsorption experiment was carried out with both powdered and block pumice tuff to compare the distribution coefficient ( $K_d$ ) values of stable cesium ( $^{133}\text{Cs}$ ) and strontium ( $^{87}\text{Sr}$ ) under variety of geochemical conditions. The main objective was to infer the  $K_d$  difference in laboratory and field condition. Cs and Sr were selected as two important nuclides from low and intermediate level radioactive waste, and pumice tuff as one of the host rocks of such waste repositories. Pumice tuff blocks were prepared for this study expecting that the block sample is nearly the 'natural condition', like what is found in the field- compact with intact pores. Considering minimum dependency of ionic strength, initial nuclide concentration and pH,  $10^{-4}$  mol/l Cs and Sr concentration was selected as initial experimental concentration (Rajib *et al.*, 2015, 2016) where 4, 8 and 12 were chosen as low, neutral and high pH conditions. Since ionic strength significantly influences  $K_d$  at less than 1.0 mol/l on pumice tuff (Rajib *et al.*, 2011, 2016), relatively high ionic strength of 1.0 and 3.0 mol/l was adopted. To keep the effect of particle size at minimum, homogenous grain size of 150-300  $\mu\text{m}$  was used for the powdered materials, whereas blocks were prepared at an equal size of 1  $\text{cm}^3$ . Conventional batch technique was carried out with a solid-solution ratio of 1:10. However, as the ratio cannot be kept fixed for block samples (the weight varied from 1.090 to 1.456 g), adjustment to 1:10 ratio was necessary to recalculate  $K_d$  values. After reaching equilibrium pH at the contact time of 14 weeks, Cs and Sr concentrations were measured by ICP-MS, and the  $K_d$  values were determined considering the natural dissolution of Cs and Sr during aging period (Rajib *et al.*, 2015). Pumice tuff samples were collected from below 50-100 m at subsurface where water-rock interaction occurred and a redox zone has been formed. Hence, both fresh and oxidized pumice tuffs were possible to use to observe any oxidation effect. To investigate surface area properties of pumice tuff, mercury intrusion porosimetry (MIP) and observation through scanning electron microscope (SEM) was conducted.

The results show that  $K_d$  values for block samples are considerably lower than powdered samples for both Cs and Sr. Almost all the samples showed similar tendency with the average lower values of 8.79% for Cs and 4.94% for Sr in block samples of fresh tuff. In oxidized tuff, block samples showed 14.58% and 13.65% lower values for Cs and Sr, respectively. The lower  $K_d$  values on block samples might be due to the use of lower surface area as sorption sites since many closed pores exist in intact solid which cannot be accessed. The destruction of smaller pores in oxidized tuff due to oxidation phenomena might be the cause of higher reducing  $K_d$  values. However, use of pumice tuff blocks for comparable batch adsorption experiment is possible due to its highly porous nature where much amount of surface area can be accessed. The  $K_d$  difference between block and powder samples is expected to be caused mainly by the effect of diffusion, taking very long equilibrium period, and the physical and mechanical properties of pores and fractures during experiment.

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Keywords: Distribution coefficient, Powder and block samples, Oxidation, Pumice tuff, Radioactive waste repository

## Behavior of co-precipitated Se (IV) during the transformation of Fe-oxides at alkaline conditions

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<sup>79</sup>Se is a radionuclide present in high-level nuclear wastes and is of particular environmental interest due to its long half-life. Degradation of waste repositories over time may lead to its release into the wider environment in the form of Se (IV). While Se is an essential element for flora and fauna, it is known to be toxic when present at high concentrations. One of the best-known mechanisms that immobilize Se and limit its mobility in the wider environment is by sorption to iron oxides / hydroxides. These phases may be formed from the corrosion of metal canisters used to contain waste forms under alkaline conditions, providing suitable sinks for Se. Most studies have focused on poorly crystalline ferrihydrite or hydrous goethite (e.g. Hayes et al, 1987; Manceau and Charlet, 1994; Das et al, 2013) due to their high surface areas and reactivity. However, since these phases are metastable with respect to other stable Fe-oxides phases (e.g. hematite, magnetite, maghemite), it is possible that these phases will eventually transform to more stable phases which may result in either the release or retention of previously uptaken Se. There is very limited information on the fate of Se during this transformation process. Thus, it is the objective of this study to clarify the behavior of Se during transformation of poorly crystalline Fe-hydroxides to crystalline Fe-oxides.

Se (IV) was co-precipitated with ferrihydrite by adding base to a solution containing both Fe and Se to raise the pH to 10. The resulting slurries were then aged at 80°C for 4 days to induce transformation. Solids and liquids were then separated by centrifugation and filtration. Solution samples were analyzed for Se concentrations while solids were characterized using x-ray diffraction (XRD), infrared (IR) spectroscopy and x-ray absorption spectroscopy (XAS). Results of solution analyses show that co-precipitation sequesters approximately 90% of Se from the solution. Transformation of the ferrihydrite to crystalline hematite and goethite phases at alkaline conditions results in the release of a fraction of the initially sequestered Se, such that only approximately 50% of the original Se is retained by the solids. Spectroscopic studies of the solids suggest that Se is sequestered from solution by sorption on ferrihydrite and not by the formation of an independent Fe-Se phase. During ferrihydrite transformation to crystalline hematite and goethite, Se is similarly retained by sorption on the crystalline phases. The release of Se during transformation may thus be attributed to the lower sorption capacity of crystalline Fe-oxides for Se. The results of this study thus show that post-uptake behavior of hazardous elements must be considered during long-term assessments of high-level waste repository conditions.

Keywords: Selenium behavior, iron oxide transformation, nuclear waste disposal

For development of evaluation technique for long term behavior between engineered barrier and host rock in near field

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In order to improve the engineering reliability in high-level radioactive waste disposal with respect to production and construction technique of overpack and the buffer material in terms of long-term integrity in consideration of the environment of deep underground, expansion of knowledge related to the quality and investigation related to the construction of the evaluation technique of the integrity have been carried out for the showing of the assessment guideline. Various experiments and researches have been conducted about the quality of the overpack and buffer material. On the other hand, it is necessary to include host rock around the engineered barrier to the experimental object in order to consider the actual disposal environment. Further, the method of evaluating the long term behavior of the engineered barrier include the experiments for understanding of phenomenon, the modeling and the numerical analysis. Experimental approach is possible to evaluate the actually occurring phenomena. On the other hand, it is difficult to directly evaluate the long term behavior of several decades to several hundred years. Therefore, the long term behavior evaluation requires modeling and numerical simulation. To improve the reliability of long term behavior evaluation by numerical simulation, it is necessary to verify the analysis technique by experimental support. Based on such backgrounds, the present study is aimed to construct a verification method of the long term behavior evaluation between the engineered barriers and the surrounding rock.

Constructing the method of the validation and verification of the evaluation of the long term behavior (such as the numerical simulation), it has been carried out in the following viewpoints in the present study.

- 1) Organizing of environmental factors that affect the long term integrity of engineered barrier (thermal, mechanics, hydraulic, etc.).
- 2) Employment of numerical code of coupled thermal-hydraulic-mechanical (THM) processes.
- 3) Considering various environmental conditions to which are exposed after the emplacement of an engineered barrier, the equivalent data of long term behavior is measured by a centrifuge model test which can be time acceleration test on the basis of the scaling law of centrifugal force field.
- 4) In the period of several decades in which the surrounding of the waste is headed for saturation, it is considered that mechanical behavior is the most dominant by the reflooding of groundwater in post-closure. Therefore, the evaluation period by centrifuge model test requires period in which the behavior of the post-saturation can also be partially measured.
- 5) In order to reduction of uncertainty in input value in numerical analysis, we measure the physical properties of the material used in the centrifuge model test, and enter the value in the numerical analysis code.
- 6) We conduct a sensitivity analysis in parameters of the numerical analysis code, and reveal the sensitivity of the parameters.
- 7) Then, we improve the accuracy of the analysis code by comparing the results of numerical

simulation and centrifuge model test, and contribute to the validation of the evaluation technique of long term behavior.

This research includes portions of the results of the "Study on Performance Evaluation for Engineering Components of HLW, FY2015" under a grant from the Agency for Natural Resources and Energy (ANRE) in the Ministry of Economy, Trade and Industry (METI) of Japan.

Keywords: Geological disposal, Long term behavior evaluation, Centrifuge model test, Coupled analysis

## A mechanism of container sinking in bentonite buffer hitherto not considered

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One of the major concepts of the geological disposal of high level radio active waste is to enclose a metallic container by bentonite buffer which is considered to be impermeable and chemically stable. Since the average density of the container is around 6 to 7 and very heavy, the scenario of container sinking has been evaluated because excess sinking shortcuts the pathway of nuclide migration in the bentonite and is detrimental to the bentonite buffer functions. Previous considerations of container sinking have been through the mechanical deformation of the bentonite. In this presentation, a chemical deformation mechanism is presented as another scenario of container sinking, which has not been previously considered in the rad-waste disposal field. Chemical deformation here is the deformation through the pressure solution of minerals constituting the buffer, transportation by diffusion, and precipitation. That such chemical deformation is a ubiquitous phenomenon occurring in various scales in the crust of the earth will be shown through the review of previous works. It will also be shown that such chemical deformation can occur at the range of the pressure and temperature of the geological disposal. It is deduced that container sinking scenario needs to be evaluated also from the viewpoint of chemical deformation of the bentonite, because the sinking distance can be significantly large in the time range of up to a million years for the safety assessment.

Keywords: Canister sinking, Chemical deformation, Bentonite, Pressure solution, Engineered barrier, Radioactive waste disposal

Natural analogue study on the stability of smectite under hyper-alkaline conditions  
- Exploration of active sites at Narra in Palawan Island, Philippines -

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Smectite will play an important role in engineered barriers for the geological disposal of radioactive wastes due to its low permeability that contributes to the isolation of radioactive nuclides, and its high cation exchange capacity that retards the migration of radioactive nuclides. However, it is predicted that hyper-alkaline fluids from cementitious components of the barrier could affect the smectite, resulting in the loss of its favorable properties as a barrier component. Although many experiments have been conducted to assess the stability of smectite under hyper-alkaline conditions, there is a significant disparity between time scales involved in laboratory experiments and actual disposal environments. Natural analogue studies to assess the stability of smectite under hyper-alkaline conditions may be used to bridge these differences in time-scales. In particular, active sites, where the reaction between smectite and hyper-alkaline fluids can be presently observed, may lead to the better understanding of the geological time-scales of the reaction and to the more precise assessment of the long-term stability of smectite under hyper-alkaline conditions. However, natural analogue studies at active sites are a few. Thus, the objectives of this study are to search for suitable active sites, and to understand the interaction between smectite and hyper-alkaline fluids.

We excavated trenches at Narra in Palawan, Philippines on the contact between ultramafic rocks and recent sediments. Fluids analysis of groundwater from trench sites revealed that hyper-alkaline water (pH>11) generated by serpentinization flows in the sites. XRD analysis of solid samples from trenches revealed the presence of trioctahedral Fe-smectite. SEM observation and thermodynamic modeling suggest that these phases were generated as the result of the alteration of primary ultramafic minerals by hyper-alkaline fluids. SEM observation of samples from the layer with the Fe-smectite also showed the presence of CSH phases that precipitate from hyper-alkaline fluids. These provide the evidence that hyper-alkaline fluids actively interact with the sediments overlying the ultramafic rocks. Therefore, Narra is surely the site where the interaction between smectite and hyper-alkaline fluids can be actively observed in nature. The reactions observed in this site can thus be considered as the analogues of the reactions that might occur at actual disposal sites where Fe could be supplied from the engineered barrier, wastes or microbial activities, and the Fe-smectite might be produced by hyper-alkaline fluids. Radiocarbon dating of organic material, from the layer of calcite which is the precipitation from surface water overlying the the Fe-smectite-bearing layer suggest these organic materials are formed about 4500 years ago. This implies that hyper-alkaline fluids circulation in the site may have begun approximately 4500 years ago. Therefore, this study implies that the Fe-smectite could be formed at actual disposal sites and it requires a relatively short time.

This research was initiated within a project to develop Geological Disposal Technologies in Japan using Natural Analogue, which was funded by the Ministry of Economy Trade and Industry (METI), Japan.

Keywords: Natural analogue study, Fe-smectite, geological disposal, active site, hyper-alkaline fluids



## Activity evaluation of fracture zone in granitic rock -Case study on the survey of fracture zones at the "Monju" site

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Background: In the fast breeder prototype reactor Monju of the Japan Atomic Energy Agency (JAEA), geological survey of fracture zones has been carried out at the Monju site based on the instruction received from the Nuclear Regulation Authority (NRA). JAEA submitted a complete report to NRA on March 2014. Results of the investigation, it has shown that no clear evidence was observed to indicate that on-site fracture zones are active faults. In December 2015, the field survey by the Experts Committee of NRA was carried out.

Overview of geological survey: The basement rock of the northern Tsuruga peninsula where the Monju site is located is composed of the Late Cretaceous-Paleogene granite known as Kojyaku granite. In the on-site investigation, the stripping area was expanded in the northern direction of the longest fracture zone in the reactor building foundation rock. We examined cross-cutting relationships of fractures and displacement of markers and features of the fracture zone structure.

Activity evaluation of fracture zone: In the stripping area, the fracture zones were grouped into 2 systems called  $\alpha$ -system (ENE-WSW) and  $\beta$ -system (NNE-SSW). We confirmed that the  $\alpha$ -system was formed after the  $\beta$ -system from the cross-cutting relationships. The  $\alpha$ -system fracture zones are left-lateral faults that have mesh-like clay veinlets, and the width of the  $\alpha$ -system fracture zones is several centimeters. The latest shear zone with width of about 1cm less was identified by investigation of cross-cutting relationships focusing on  $\alpha$ -system fracture zones. Minor right-lateral slip and biotite grains deformed plastically are observed on the thin-section of the latest shear zone. According to the existing experimental studies (Stesky, 1978; Lin, 1999, etc.), the temperature condition of plastic deformation of biotite grain is known to be exceeding 150 to 250 degrees centigrade. Also, biotite grains deformed plastically are universally observed along the fracture zones in the stripping area. Therefore, it can be considered that these fracture zones are older small-scale geological structure which was formed under the high-temperature environment of the deep part before exposure of the granitic body, and that these fracture zones are non-active in recent age.

In addition, the basalt dikes which have the K-Ar ages of about 19Ma are observed in the stripping area. It can be considered that basalt dikes partially intruded along the  $\alpha$ -system fracture zones later from the distribution of the chilled margin of contact boundary between the granite. Calcite veins and irregular local deformation structures are observed into the basalt dikes. Also, the calcite veins often develop parallel to the boundary between the basalt and the granite along the fracture zones. Twins which indicated the deformation at a high temperature (150 ~ 300 C, typeII; Burkhard, 1993) are observed frequently in the calcite veins of the boundary portion under microscopic observation. However, there is no damage due to shear in the calcite vein itself. From this fact, it can be also considered that the boundary portion between the basalt and the granite along the fracture zone is not sheared after the calcite veins are formed in a high temperature environment.

In the activity evaluation of the fracture zone in the granite, it is possible in some cases to estimate the formation environment (temperature conditions, etc.) of the fracture zone from the feature of structure and minerals in the fracture zone or veins along/across the fracture zone. As a result, information of consideration concerning whether the fracture zone is active or not can be obtained.



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Keywords: fast breeder reactor "Monju", survey of fracture zone, Kojyaku granite