# Micro-chemical Probe: Powerful New Tool for Assessing Radioactive Waste Geological Disposal Sites

Takuma Yoshida
Akira Sasahira, Dr. Sc.
Kenji Noshita

*OVERVIEW: NUMO's approach has been to call for municipalities to volunteer as candidate areas for the disposal of high-level radioactive wastes in Japan. The proposed sites are being subjected to a multi-step assessment process to determine their suitability starting with a survey of relevant literature, then proceeding with a surface evaluation, and finally an underground assessment of the sites. When investigating sites from the surface, deep boreholes are dug to ascertain the nature of the sites, but so far we have lacked the technology to directly measure distribution coefficients and other key geochemical properties that largely govern the migration behavior of radionuclides in rock strata, and have had to rely on measurements based on simulations of underground conditions done by test equipment on the surface. It was this fundamental limitation that motivated the present project to develop a micro-chemical probe capable of directly measuring deep subterranean conditions in a borehole.*

## INTRODUCTION

ESTABLISHING a viable nuclear fuel cycle for reprocessing and recycling spent nuclear fuel is critically important to ensure Japan's long-term energy resources. Indeed, the value of the nuclear fuel cycle has become even more apparent recently in the light of concern about global warming and scarce uranium resources. As the process is currently conceived, the nuclear fuel cycle involves first separating fission products from spent nuclear fuel through reprocessing, then depositing of the high-level nuclear material as vitrified waste in underground sites.

The Nuclear Waste Management Organization of Japan's (NUMO's) approach has been to call for

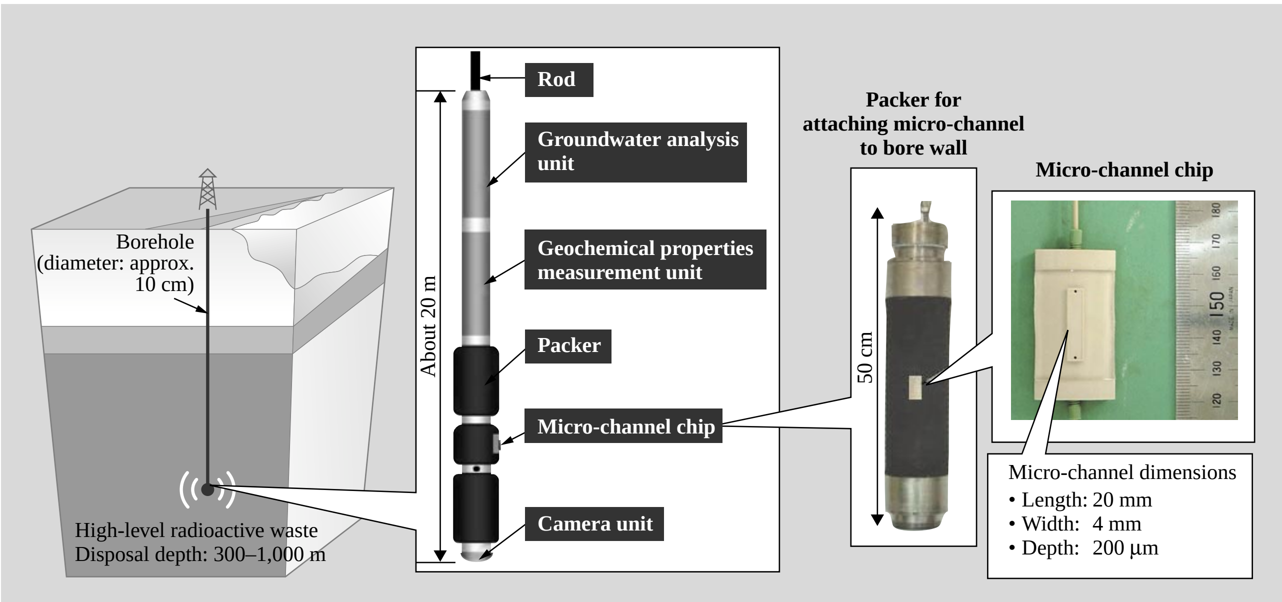


*Fig. 1—Overview of the Geological Environment Measurement System Using the Micro-chemical Probe.*
*The system measures geologic distribution coefficients, diffusion coefficients, and other geochemical properties based on solid/liquid interfacial reaction assessment technology incorporated in a unique micro-channel developed by Hitachi. This will enable in situ measurement of critical geochemical properties in deep subterranean formations by lowering a micro-chemical probe down into a borehole.*

municipalities to volunteer as candidate areas for the disposal of high-level radioactive wastes in Japan. The proposed sites are being carefully scrutinized through a multi-step assessment process—survey of all relevant literature followed by a surface evaluation and then an underground assessment—to determine which of the sites are most suitable. The actual site has not yet been determined, but 4-5 years after beginning to survey the literature on the various sites, a preliminary survey will be conduced based on boreholes, followed by a detailed investigation using underground equipment and facilities, and finally the actual disposal construction site will be selected. Although rock permeability and other hydrographic properties can be measured from the surface via a borehole, we have lacked the technology for directly measuring distribution coefficients and other key geochemical properties that reveal the migration behavior of radionuclides in rock strata, and have had to rely on measurements based on simulations of underground formation conditions done by test equipment on the surface.

This paper reports on the status of the micro-chemical probe currently under development that will enable us to directly measure distribution coefficients and other critical geochemical properties in deep subterranean formations via boreholes (see Fig. 1).

## MICRO-CHEMICAL PROBE

Radionuclides leaching from radioactive wastes in disposal sites will get into groundwater and migrate over a very long geologic period, but mobility is impeded in the process through repeated sorption and desorption where groundwater comes into contact with bedrock. The rate of radionuclide migration is thus far slower than the rate of groundwater migration, and the degree of radioactivity will be largely attenuated before such radionuclides reach the biosphere. But in order to accurately assess mobility retardation, it is necessary to measure hydrographic characteristics such as the permeability of the rock as well as the geologic distribution coefficient that reveals the extent of radionuclide absorption by the rock.

Conventionally, the distribution coefficients of actual rock strata have been obtained using testing equipment deployed above ground to simulate underground conditions using rock samples brought up from boreholes. But now with the growing need for more accurate assessment of nuclear waste geological disposal sites, Hitachi was commissioned as part of Ministry of Economy, Trade and Industry (METI)'s Innovative and Viable Nuclear Energy Technology Development Project to develop technology that dramatically improves the quality and reliability of geologic distribution coefficient data, and since 2004 has been working in collaboration with Tokai University, the University of Tokyo, the Tokyo Electric Power Company, Tokyo Electric Power Services Co., Ltd., DIA Consultants Co., Ltd., and JGC Corporation on a rugged micro-chemical probe that is capable of directly measuring the distribution coefficient from deep subterranean layers in boreholes.

The micro-chemical probe consists of two basic units: a geochemical properties measurement unit, and a groundwater analysis unit. The features of each unit are explained as follows.

(1) Geochemical properties measurement unit

The geochemical properties measurement unit features a unique thin layer channel device called a micro-channel developed by Hitachi for measuring the geologic distribution coefficient and diffusion coefficient. We will discuss how the micro-channel works in some detail later in the paper.

(2) Groundwater analysis unit

Iron ions ($Fe^{2+}$) and sulfide ions ($S^{2-}$) in groundwater are key constituents in determining the character of underground chemical environments, and also have an enormous affect on geochemical properties. Unfortunately, these ions tend to become unstable in the atmosphere when they are brought up to the surface, so this unit was developed to be lowered down into boreholes to analyze iron and sulfide ions in situ to obtain a clear picture of geochemical properties and other critical information about the underground environment.

In the rest of this paper, we will elaborate on the current development status of these key units of the micro-chemical probe.

## STATE OF DEVELOPMENT OF THE MICRO-CHEMICAL PROBE

### Geochemical Properties Measuring Unit

(1) Micro-channel for measuring geochemical properties

The conventional procedure for determining a diffusion coefficient was to measure the time it takes for a tracer radionuclide to actually pass through a thin sample of rock, as illustrated in Fig. 2 (a). And the distribution coefficient was measured by crushing a rock sample, exposing the surfaces of granules inside, then measuring the distribution coefficient from the amount of absorption.

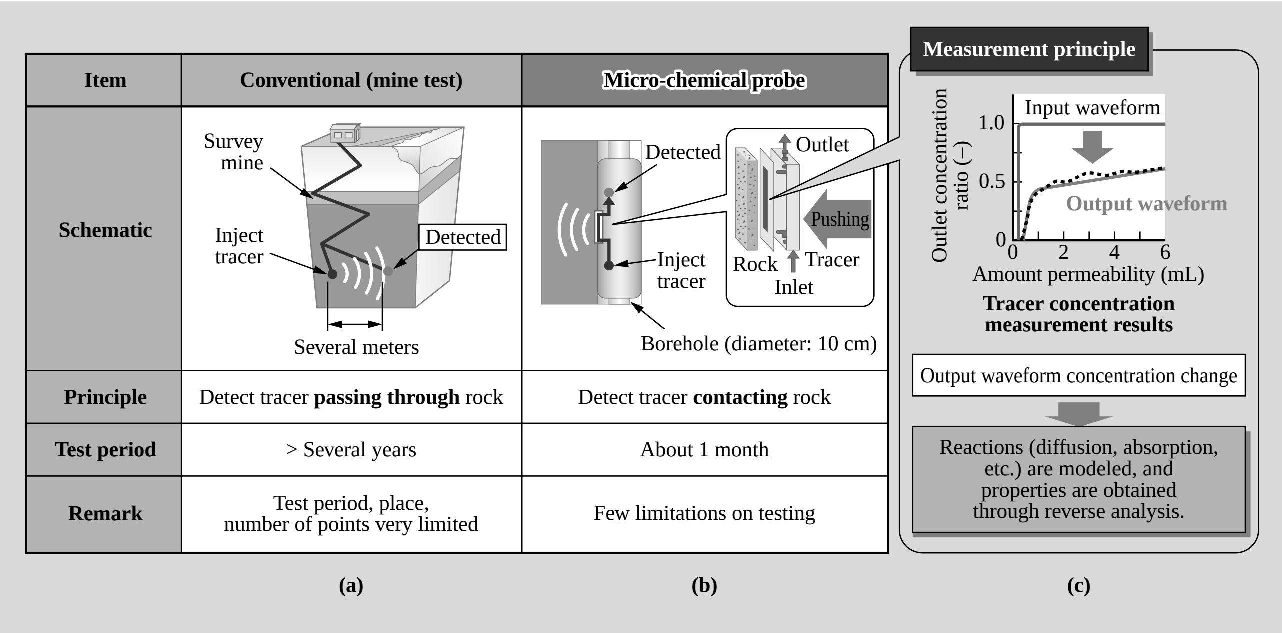


*Fig. 2—Measurement of Geochemical Properties with the Micro-channel.*
*Figure shows the conventional method of measuring geochemical properties (a), the new method using the micro-chemical probe (b), and a typical example of the micro-mockup (c) .*

Adopting the basic principle of a microreactor, Hitachi recently developed a far better method of analyzing the properties of rock called the micro-mockup method that permits the relatively rapid measurement of both the distribution and diffusion coefficients at the same time(1). A typical example of micro-mockup is shown in Fig. 2 (b) and (c). A sample is extracted from the site bedrock, and a fine 100-μm-wide channel is scribed in the surface of the sample. A response curve is then obtained by allowing tracer solution to permeate through the sample, and measuring the change over time of tracer concentration at the outlet. By analyzing the response curve, we can assess the rock retarding coefficient and the matrix diffusion coefficient. The advantages of this approach are not only that it is much faster than the conventional method, but also that it permits evaluation of retardation by sorption of crack surfaces and matrix diffusion from the same unbroken sample, variables that have been extremely difficult to directly measure in the past(1).

(2) Emplacement unit

In order to perform deep subterranean measurements in a borehole, we had to come up with a reliable mechanism for firmly attaching the micro-mockup micro-channel to the curved inside wall of the borehole. Working in collaboration with DIA Consultants, we devised a method that attaches the micro-channel board chip to the bore wall by water pressure using a flexible elastic gasket called a packer.

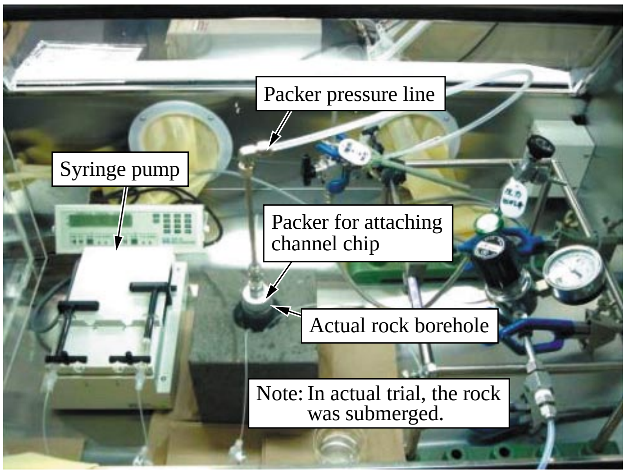


*Fig. 3—Borehole Radionuclide Permeation Test Setup.*
*Radionuclide migration tests were performed in an actual hole bored into a block of granite.*

We conducted test measurements of radionuclide migration using this method in an actual hole bored into a block of granite using the setup shown in Fig. 3. A 500 Bq/mL (becquerel-per-milliliter) solution of tritium (H-3) and 1,000 Bq/mL solution of strontium chloride (Sr-85) were used for radionuclides. The micro-channel here was 200-μm deep, 2-cm long, and 4-mm wide. We conducted the same test using a thin

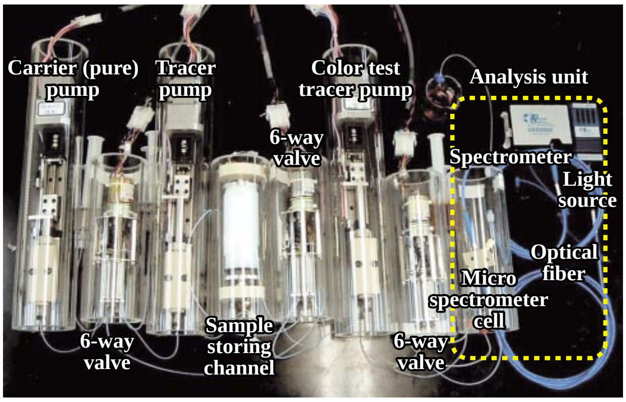


*Fig. 4—Photo of the Tracer Analysis System.*
*Spectrometer system is enclosed in housing less than 60 mm across.*

slab rock sample instead of a block of stone, and compared the results using the different shaped samples. We obtained virtually identical response curves for the two H-3 nuclides and for the two Sr-85 nuclides, despite the fact that the shapes of the two rock samples were totally different. We also conducted a numerical analysis, and obtained $1 \times 10^{-11}$ ($m^2/s$) for the effective diffusion coefficients, $D_e$, from the H-3 response curves, and $1 \times 10^2$ (mL/g) for the distribution coefficients, $K_d$, from the Sr-85 results for the two samples. It is thus apparent that we would obtain virtually identical radionuclide migration retardation characteristics from deep underground in a borehole and from lab data.

(3) Tracer analysis system

Fig. 4 shows a photograph of the tracer analysis system. The system alternately sends tracer solution and carrier solution to the micro-channel, which passes first through a micro spectrometer for online analysis, then to a sample storing channel for off-line analysis. The sample storing channel for off-line analysis consists of many narrow tubes for storing tracer solution samples over a long time frame to assess changes in concentration, and also to store samples brought up from the hole for detailed analysis that could not be analyzed on line.

Designed and built for use in deep underground boreholes, the tracer analysis system is ruggedly constructed to meet the following specifications:

(a) Compact implementation: The diameter of the system is less than 60 mm.

(b) Pressure resistance: The system is designed to

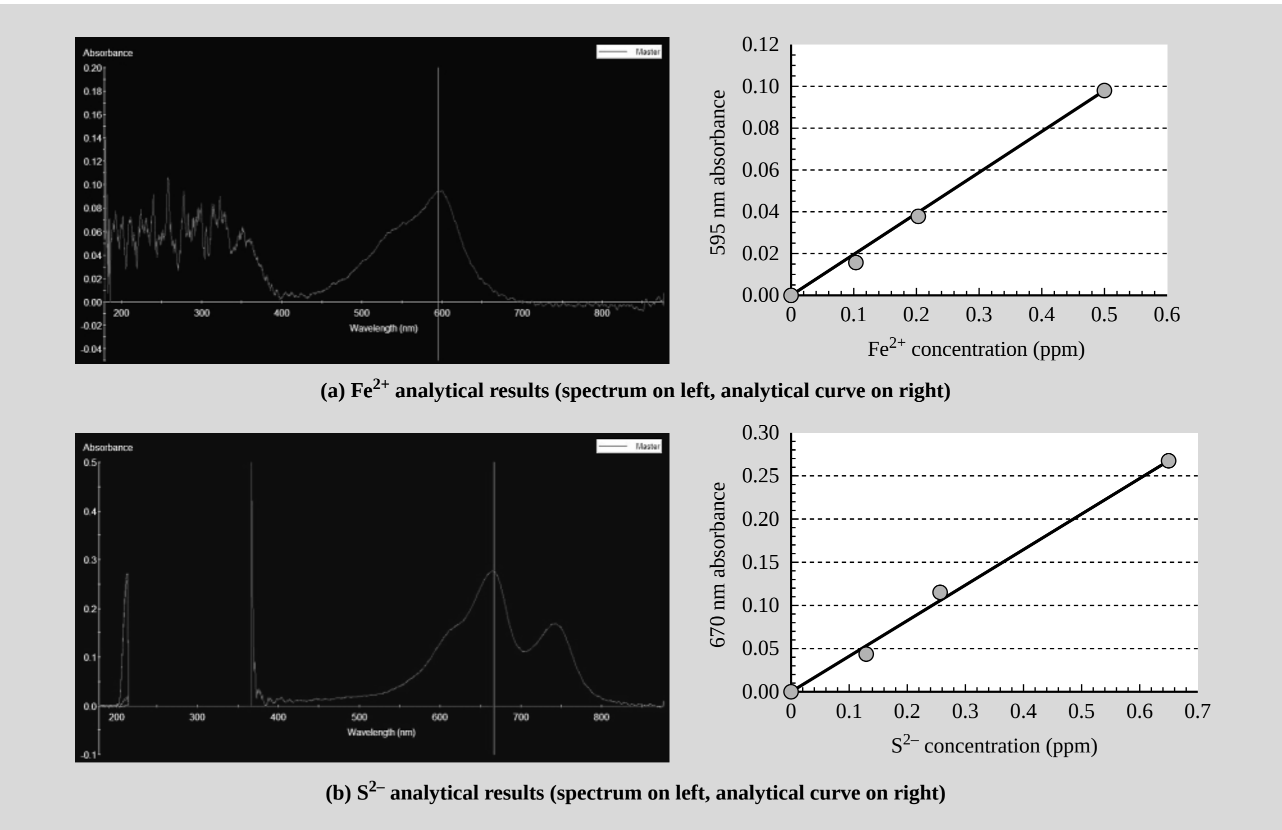


*Fig. 5—Online Analysis Verification Test Results.*
*All analyses confirmed that results were below the specified lower limit detection value of 0.1 ppm.*

withstand pressures more than 5 MPa.

(c) Analytical accuracy: Accuracy of the system is equivalent to a typical absorptiometer.

Actual online test results for the micro spectrometer indicate that the system could prospectively yield results that are below the specified lower limit detection value of 0.1 ppm.

### Groundwater Analysis Unit

The groundwater analysis unit consists of a micro spectrometer and multiple water quality sensors for analyzing groundwater samples. The micro spectrometer is capable of in situ measurements of iron and sulfide ions — reducing agents that have a disproportionately large affect on migration of radionuclides — in amounts as low as 0.1 ppm. The water quality sensors are capable of in situ measurement of time, depth, water temperature, pH (hydrogen ion concentration), Eh (oxidation-reduction potential), conductance, and dissolved oxygen. This system too was designed and built for use in deep underground boreholes, so was constructed to meet the same rugged pressure resistance and dimensional requirements as the tracer analysis system. Fig. 5 shows typical online analysis test results for the micro spectrometer. All of the analysis results are well within specified target ranges.

## CONCLUSIONS

This report details the progress made so far in developing key units for implementing a rugged micro-chemical probe for use in deep boreholes. We are currently finalizing the overall design of the micro-chemical probe so it will stand up to the harshest conditions as we approach the final milestone—full-scale trials of the probe in an actual borehole. The objective of this work has been to develop a powerful new tool that in the near future will contribute to assessment of geological disposal sites for radioactive waste that are safe and reliable. This development is part of the work achieved in the Innovative and Viable Nuclear Energy Technology Development Project supported by METI in 2004 and 2005.

## REFERENCE

(1) K. Okuyama et al., “A fast and sensitive method for evaluating nuclides migration characteristics in rock medium by using micro-channel reactor concept,” *Physics and Chemistry of the Earth*, Vol. **32**, Iss. 1-7, pp. 463-468 (2007).

## ABOUT THE AUTHORS

Takuma Yoshida

*Joined Hitachi, Ltd. in 1986, and now works at the Nuclear Fuel Cycle Project Management Group, the Nuclear Power Business Development & Management Division, Hitachi-GE Nuclear Energy, Ltd. He is currently engaged in bringing together back-end operations of the nuclear fuel cycle. Mr. Yoshida is a member of the Atomic Energy Society of Japan.*

Akira Sasahira, Dr. Sc.

*Joined Hitachi, Ltd. in 1985, and now works at the Nuclear Power Chemical Project Fuel Cycle Group, the Power & Industrial Systems R&D Laboratory, the Power Systems. He is currently doing work in nuclear fuel cycle area. Dr. Sasahira is a member of the Chemical Society of Japan and the Atomic Energy Society of Japan.*

Kenji Noshita

*Joined Hitachi, Ltd. in 1991, and now works at the Nuclear Power Chemical Project Fuel Cycle Group, the Power & Industrial Systems R&D Laboratory, the Power Systems. He is currently engaged in research on radioactive waste disposal. Mr. Noshita is a member of the Atomic Energy Society of Japan.*