Application of Inert Gases as Tracers
in a Multiple Well System

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ABSTRACT

Gas chromatographic analyses of dissolved inert gases were applied in tracer tests of groundwater flow in fractured rock masses. Nine wells drilled in granite were used for the tracer test and the test with He gas clearly disclosed mass transport of the tracer solution between the test wells but the result with Ar was less satisfactory. The attainable signal-to-noise ratio in the groundwater may be responsible for the unsuccessful result in the Ar test and the success of He in the hydrological test. Since inert gases do not interact with the chemical environments, experiments concerning dissolution and migration of chemical species and the present tracer test for the water flow can be carried out in the groundwater simultaneously without mutual interference. This method may be feasible for in situ characterization and monitoring of a repositories for high level radioactive wastes.

INTRODUCTION

Rock masses usually contain systems of cracks or failures whose anisotropy and inhomogeneity can complicate the simple hydrological model. In this context, in situ tracer studies in controlled field experiments may provide information concerning the direction and/or velocity of the water in complicated flow systems in rock masses. Many kinds of substances have been used as tracers, but most substances such as radio-isotopes inevitably pollute waters. Inert gases, however, present no health and safety hazards, and are neither lost nor added under the ground (an exception being radiogenic He and Ar in water). Thus, inert gases are potentially useful tracers (Carter et al., 1959) and measurements of the concentration of inert gases dissolved in groundwater have been used for various purposes, such as the calculation of infiltration temperatures (e.g. Sugisaki, 1961; Mazor, 1972) and groundwater dating (Zaikowski et al., 1987). Nevertheless, tracer tests with inert gases have been beyond the the scope of most routine hydrological studies because of technical and labor intensive obstacles in the measurement.

The current method for the determination of gases dissolved in water is extraction from a water sample by evacuation and analysis with a mass-spec-

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trometer (e.g. Carter el al., 1959; Mazor, 1972; Heaton and Vogel, 1981), which is quite complicated labor intensive and expensive. Sugisaki et al. (1982), however, overcame the preconception that inert gases in trace amounts can only be analyzed with a mass-spectrometer and showed that they can easily and precisely measured using a gas chromatography, a much less expensive technique. Sugisaki and his colleagues successfully applied this method to tectonic problems such as earthquake prediction (Sugisaki, 1978, 1981; Sugisaki and Sugiura, 1985, 1986; Sugisaki et al., 1983). Sugisaki and Taki (1987) devised a simple method for extraction and analysis of gases dissolved in natural water, that does not need a vacuum line but still has a high enough sensitivity to measure He content as low as 50 ppb. This technique is applicable to routine analyses of inert gases dissolved in water and would be adopted for monitoring purposes.

In the present study, we use this method to examine the feasibility of inert gas tracers in field tests of hydrological studies in fracture dominated rock masses. Further, we evaluated the efficacy of each inert gas in the tracer test. This study is a part of a project for evaluation of Tono uranium mine, central Japan, as an analogue to a nuclear waste repository.

GEOLOGICAL SETTINGS

Tono uranium mine, Mizunami City, Central Japan, (Power Reactor and Nuclear Fuel Development Corporation) provided a site for the present experiments. The ore body in the mine is covered with Tertiary sedimentary rocks (consisting mainly of sandstone, shale, conglomerate and tuff) and underlain by Cretaceous granite. The geology around the place is schematically shown in Fig. 1. The present test was conducted in biotite granite of medium to coarse grain (Toki Granite), at a corner of the gallery about 5 m east from the bottom of a main shaft. The fracture system with both horizontal and vertical cracks filled with clay may be responsible for the groundwater flow around the site.

METHODS

Nine wells for the tracer test were drilled at 50 cm apart (Fig. 2). Each well is 74 mm in diameter and 1.5 m in depth. A plastic pipe 40 cm long was inserted into the upper part of each well to protect the well-wall from collapse. Preliminary tests showed that water levels in the wells rose at rates of 0.1 to 3.5 cm/min after removal of the 1.5 m water column present in the wells. Under equilibrium conditions, all the wells spontaneously discharged water at a rate that varied daily. This suggests that the fluid flow rock mass around the site cannot be treated as a fixed and homogeneous medium in the scale on this investigation.

We tested the flow direction and velocity of the groundwater in the multiple well system; the central well (denoted as No. 0 in Fig. 2) was used for

the tracer injection and 8 surrounding wells (No. 1 to No. 8) were designated for the withdrawal. The concentration of tracer in the injected/withdrawn water was recorded as a function of time.
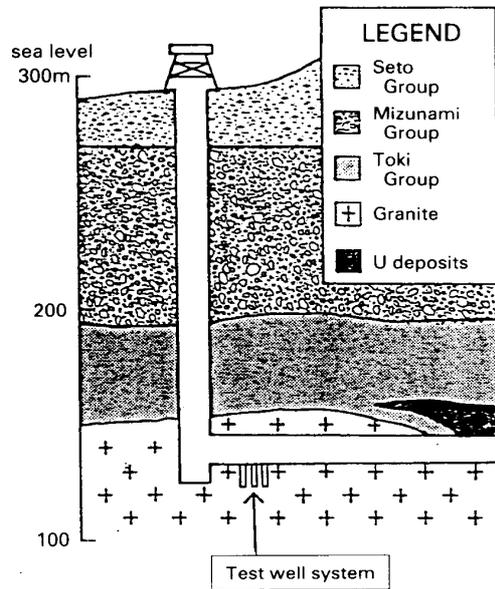

Fig. 1   Diagrammatical section showing geology and the experimental site in Tono uranium mine. The uranium deposits are contained in Toki Group of sandstone and conglomerate in the Miocene. Mizunami Group and Seto Group mainly consisting of sandstone, shale, conglomerate and tuff are the Miocene and the Pliocene, respectively. The wells for the present test were drilled on the floor of the basal granite.
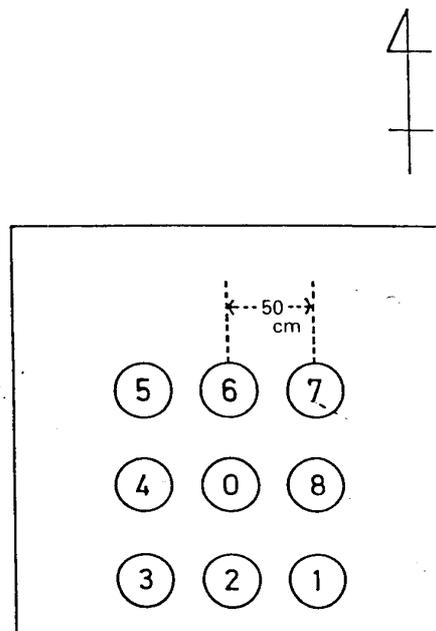

Fig. 2   Ground plot showing arrangement of the test wells and their numbers.

The injection procedure for the conservative inert gas tracer solution is diagrammatically shown in Fig. 3. Minute bubbles of the gas were blown into in situ collected groundwater in a bottle through a sintered glass ball from a gas tank. The surplus gas that was not absorbed in the water was exhausted into open air. The gases originally dissolved in the groundwater were replaced by the tracer gas within about 10 min. Then the stopcock attached to the bottle was opened and the water charged with the tracer gas flowed down slowly into the bottom of the injection well through a rubber tubing while gas at a little higher than one atmosphere pressure was being supplied into the bottle. The injected volume of the tracer solution (6.5 1) is approximately the same as that of the injection well. The excess water in the well (No. 0) replaced by the solution during this procedure was drained out of the upper part of the well through the other rubber tubing. It takes about 10 min to inject the tracer solution. After injection was complete, both rubber tubings were closed with pinchcocks to prevent contact between the well water and air; otherwise, the inert gas in the injection well will escape into air before the tracer solution permeates into the surrounding rocks. This procedure is different from that of tracer tests using ionic substances such as halogens which do not readily escape from the injection well.
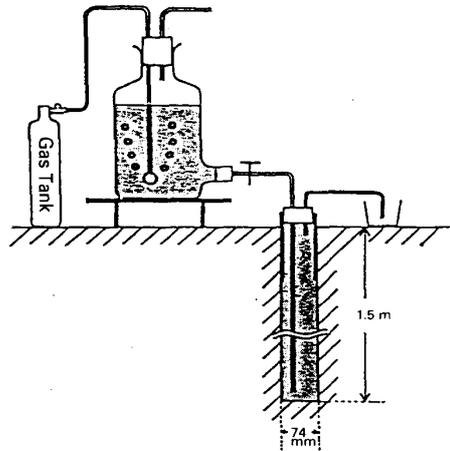

Fig. 3   Schematic diagram showing injection procedure for tracer solution.

After injection, water samples of 100 ml each were collected from a depth of 1.0 m in each well, although only 7 ml of water is necessary for the analysis. The tracer gas dissolved in samples was analyzed according to the method of Sugisaki and Taki (1987) with a gas chromatograph at a field laboratory on the surface near the shaft. Since this mine, a test plant of the corporation, is only accessible at day except holidays, the sampling intensity was limited. Approximately 20 min were required to analyze each sample; it takes about 3 hrs to analyze 9 well samples at each time. Determination errors are estimated to be ±0.015 ml/l and ±0.0001 ml/l for Ar and He, respectively,

which may be derived from the fluctuation of atmospheric temperature and sampling procedures. The initial conditions and background levels of He and Ar are shown in Table 1.

Table 1.   Atmospheric inert gases dissolved in water

	He	Ar	Kr
Abundance in atmosphere (ppm)	5.2	9170	1.1
Concentration in water equilibrated with atmospheric air at 10°C (ml/l)	4.8×10^{-5}	0.38	9.2×10^{-5}
Saturated value in water at 1 atm inert gas pressure and 10°C (ml/l)	9.3	42	84

RESULTS

Test using Ar gas as the tracer

The concentrations of Ar dissolved in well waters are plotted against time (Fig. 4). The diagram for each well in Fig. 4 is arranged in correspondence with the location of the well (Fig. 2) at the experimental site.
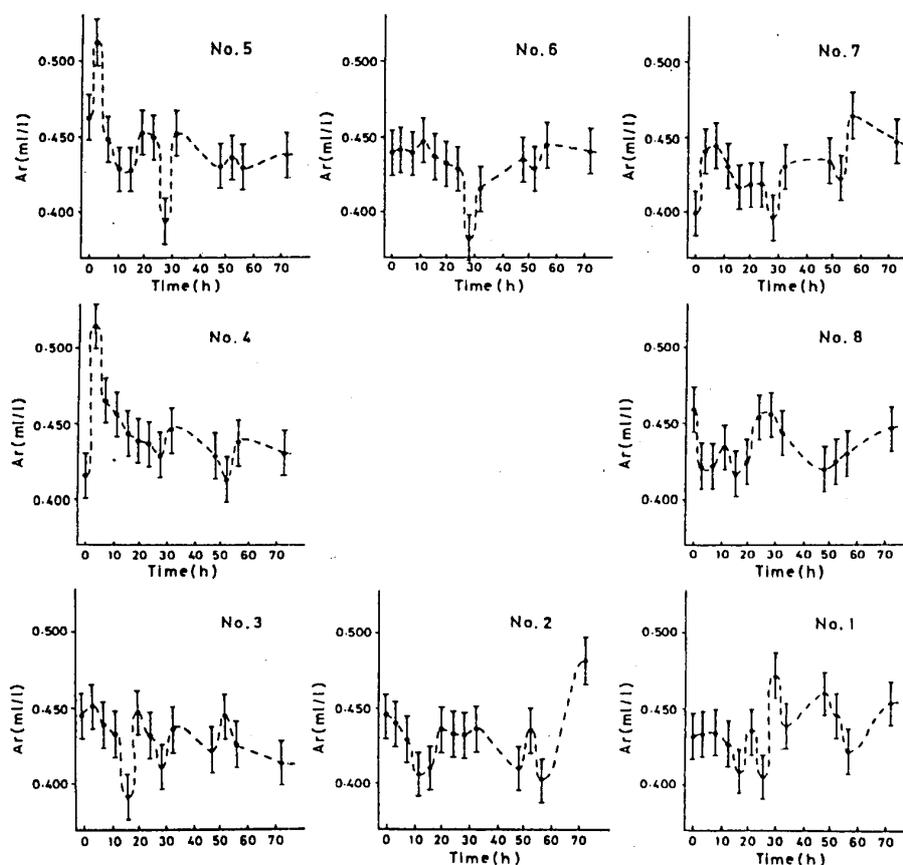
Fig. 4   Temporal variation of Ar concentration in each well. Dotted curves showing Ar variation were arbitrarily drawn. The arrangement of each diagram in this figure corresponds to that of wells in the site (Fig. 2).

As shown in Fig. 4, Ar peaks in wells No. 4 and No. 5 appeared 3 hrs after the injection. This shows that the groundwater flows westerly or northwesterly at a rate of about 6×10^{-3} cm/s (20 cm/hr). The increase in Ar concentration at the peak, however, amounts to at most 20% in the wells, and the change was not conspicuous. Furthermore, the sampling interval (3 hrs) seems too long to decide whether the highest Ar values observed (3 hrs after the injection) occurred before or after the actual peak of Ar. Thus, the direction and the rate of the groundwater flow is only loosely constrained on the basis of the present result. The sampling interval was therefore shortened in the further experiment using 3 wells (Fig. 5), but a clear trend in Ar-concentration change for the 3 wells is difficult to recognize. It should be noted that average Ar concentrations increased in the additional test which was carried out 3 weeks after the previous test. This suggests that the groundwater in the site was contaminated by Ar injected through the tests.
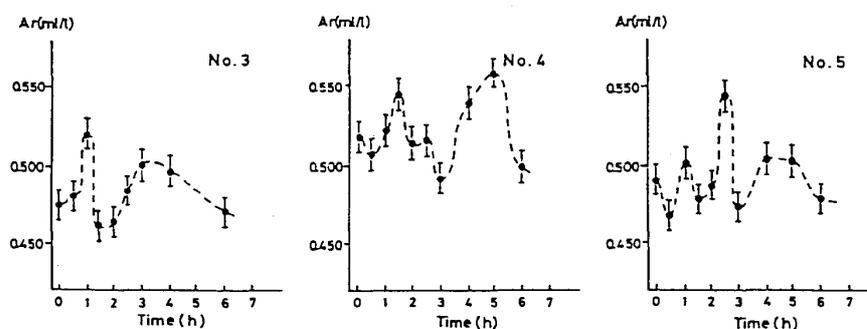

Fig. 5   Temporal variation of Ar concentration at the additional test using Ar as the tracer. Note increased concentrations of Ar as a whole compared with the previous test shown in Fig. 4.

Test using He gas as the tracer

With He as a tracer gas, the temporal variation of He concentrations in the injected well as well as the surrounding wells was measured and gave clear peaks of He at several wells (Fig. 6). In well No. 4, a conspicuous peak appeared 75 hrs after the injection of the tracer solution. The highest concentration measured at the peak amounts to 7 times the background value of 10^{-3} ml/l. Meanwhile, well No. 0, as the source of tracer solution, exponentially decreased its He concentration; the initial He concentration was as high as 9.5 ml/l, saturated value, and at 170 hrs since the start of the test, it had fallen to the background value. This shows that the original water charged with He was swept away from well No. 0 and flowed westerly at the rate of 1.9×10^{-4} cm/s (50 cm/75 hrs). During the test with He, a small and later peak appeared at well No. 7 and also positive increases over the background level of He emerged at wells Nos. 2, 5 and 8. This implies that groundwater slowly spread in every direction besides the main and westward flow. Further, the hydraulic conductivity should have been different at each fracture which plays an important role in conducting fluids through the rock mass.

DISCUSSION

Since the main purpose of this study is to evaluate the feasibility of routine tracer test by means of inert gas in fields, hydrological parameters such as permiability, hydraulic gradient and others were not measured and the hydraulic evaluation of the present results is beyond the scope of this paper. Several technical points are discussed below.

Tracer tests by means of Ar and He suggest a main and westward flow of groundwater in this site. The travel time of Ar solution between well No. 0 and No. 4 appeared to be shorter than that of He (Figs. 4 and 6). This discrepancy could be ascribed to changes in the hydraulic conductivity of each fracture by plugging with clay at various degrees during and between the tracer tests. This is supported by the following observation. Water was removed from a well by pumping and then recovery rates of water levels at other wells were measured. The mutual relation of water-level changes between wells was not fixed but varied between tests: In the first test, pumping water from a well resulted in level drops in several wells around it. In the next test, pumping water from the same well caused water level drops in wells other than those which lowered their level in the previous test. This suggests that the connection through fractures between wells was modified by transfer of clays filling in fractures; the transfer was caused by intensive flows of the groundwater during pumping. Consequently, hydraulic conductivities between wells changed between the tests. Furthermore, it should be noted that these hydrological tests for water level recovery were repeated during the interval between the time for the Ar tracer tests and that for He test: In this interval, the hydrological condition around the site must have changed as a result of
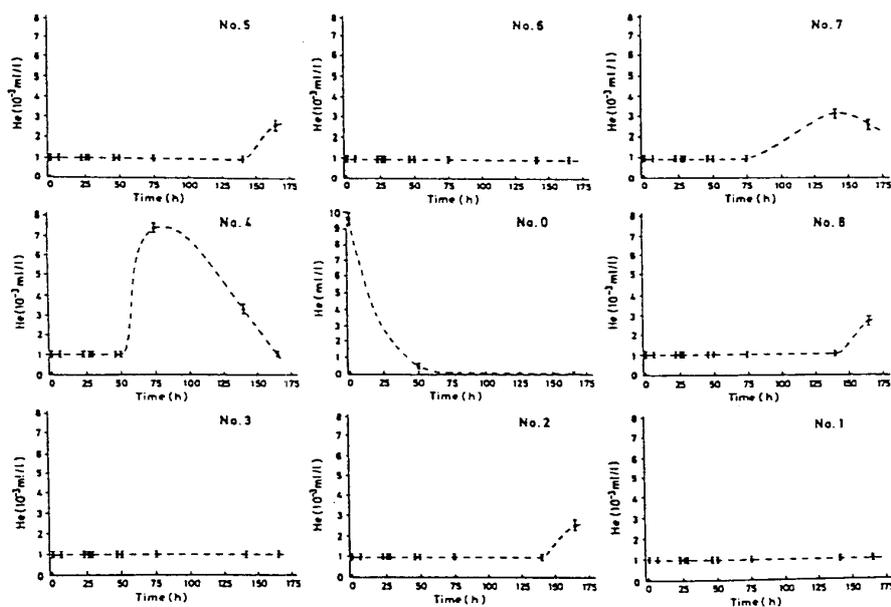

Fig. 6   Temporal variation of He concentrations in the tracer test. The scale of He concentration for well No. 0 is different from the rest.

pumping a large volume of groundwater from the wells. This modification of the condition may have inevitably resulted in the discrepancy between flow rates measured with Ar and He. Thus, the groundwater flow in a rock mass is likely to be a complex process involving many sets of fissures of varying size which are sometimes opened and sometimes plugged owing to transfer of clays in fissures.

The results of the tracer tests showed that He provided the clear peak and good test (Fig. 6), whereas the Ar results were more equivocal (Figs. 4 and 5) even though Ar is more soluble than He in water (Table 1), and might be expected to provide a clearer peak in the test. The difference between He and Ar in the tests can be attributed to their ratios of peak (signal) to background (noise) concentrations in the groundwater. The background He-concentration is about 10^{-3}ml/l, which is higher than that of surface waters due to radiogenic contribution from the uranium deposits. The highest He concentration was 7×10^{-3} ml/l at well No. 4, whereas the initial concentration in the water injected at well No. 0 was 9.4 ml/l, saturated value at 10°C (Table 1). The water at the peak observed at well No. 4 represents only about one-thousandth, by volume of the conservative tracer solution injected in well No. 0. For an equivalent dilution of Ar-saturated solution, (42 ml/l: Table 1), the Ar concentration is lowered to 0.042 ml/l, or only low a tenth of background Ar (0.4 ml/l). Thus, the low signal-to-noise ratio (as low as 0.042/0.4) in Ar variation, due to the high Ar background, inevitably resulted in the equivocal concentration of Ar in the tracer test. Furthermore, Ar injected in the first test irregularly raised the background level in the site and may have spoiled the trend of Ar in the subsequent test (Fig. 5).

Argon background concentrations in normal groundwaters are slightly higher than equilibrium with atmospheric air (Table 1). This shows that Ar in the groundwater was derived from atmospheric air and the excess Ar may be ascribed either to the solution of air bubbles trapped by infiltrating water in the unsaturated zone (Heaton and Vogel, 1981) or to air trapped in the fissures of hard rock masses (Mazor et al, 1983; Sugisaki, 1987). In contrast, the He background in the present groundwater is higher than that of the usual groundwater whose He level is estimated from equilibrium between water and atmospheric air. Most of the excess He here can be attributed to radiogenic He rather than the trapped air (Sugisaki, 1987). The radiogenic He does not however contribute to the background as much as does atmospheric Ar. The high background of Ar in groundwater results from the high abundance of Ar in atmospheric air (0.917%). An inert gas of trace concentration in atmospheric air could therefore be useful as a tracer in similar tests. Because Kr-abundance in atmospheric air is less than He and is more soluble in water than He (Table 1), Kr gas might be a candidate for tracer tests. Nevertheless, we hesitated to examine it, because of its low sensitivity in gas-chromatographic analysis and because of its high cost.

In this study, a single gas chromatograph demonstrates the usefulness of the technique but the obtained data were not always sufficient for practical

use. Use of two or three gas-chromatograph apparatus and shorter intervals of measurements would be achieved. This is not unrealizable in the field, considering cost, size and maintenance of the apparatus.

CONCLUSIONS

The present study was originally undertaken as a part of the project for a geologic repository of nuclear wastes. In the wells used for this study, experiments on the dissolution of glasses containing chemical analogues for radionuclides and the migration of these substances in groundwater have been conducted. Injection of most other types of tracers would have chemically altered the groundwater at the site and interfered with the experiment for nuclear wastes. Injection of inert gases does not. Thus, both the experiments — dissolution-migration of chemical analogues and tracer test with inert gases — are simultaneously workable.

Groundwater generally contains He as low as 50 ppb (Table 1), the concentration of dissolved He in water equilibrated with air. Whereas the groundwater for the present test carried out at a corner in the uranium mine showed higher He background (10^{-3} ml/l). Nevertheless, this background level did not hinder the test. Thus, use of He as the tracer test has a twofold advantage — chemical inertness and low background; hence this method may be widely applicable to general hydrological works.

ACKNOWLEDGMENTS

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