Plan and progress of a cooperative research program on field migration test between JAERI and CIRP (Phase-2)

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A six-year cooperative research program between the Japan Atomic Energy Research Institute (JAERI) and the China Institute for Radiation Protection (CIRP) has been conducted to establish a safety evaluation methodology for shallow land disposal of low level radioactive waste. The program consists of tracer experiments in aquifer and aerated layers at a CIRP's field test site, large scale simulation experiments with undisturbed soil, laboratory experiments, survey of the site characteristics, and migration analysis with an appropriate model.

To perform the tracer experiments in aquifer layer, an Underground Research Facility (URF) has been constructed to directly access to the aquifer layer of 31 m deep in the site. The URF provides two experimental pits and one experimental shaft. The pit experiment tests three dimensional migration of both stable and radioactive tracers under natural groundwater flow condition, while the shaft experiment tests tracer migration under various flow conditions. All the tests use H-3, Sr-90, Np-237, Pu-238 for radioactive tracers, or Br, Sr, Ce, Nd for stable tracers. Groundwater samples for H-3 and Br concentration have been analyzed regularly. Soil samples will be taken after the completion of the experiments in the year of 2000.

Tracer experiment in aerated layer is performed to test tracer migration behavior in both engineered materials (cement, bentonite, mortar) and natural barrier material (loess soil) with the same tracers for the aquifer experiment. Nine pits are provided at the ground surface in the site. Four of them are tested under artificial rainfall condition by showering water regularly and the rest is for natural rainfall condition. Soil samples have been taken several times and the preliminary result shows some extent of migration in stable Sr distribution.

Large scale simulation experiments have been conducted with undisturbed soil from the aerated and aquifer layers in the site under a controlled flow condition. The size of columns for the aerated layer experiment is up to a diameter of 28 cm and 120 cm long, and experimental tanks for the aquifer experiment are 30×30×80 cm.

Conventional migration model will be verified by comparing the migration distribution of nuclides obtained from the field migration experiments with that evaluated by using the values of necessary parameters analyzed and obtained from the laboratory experiments, large scale simulation experiments, and the survey of site characteristics.
Keywords: field test, tracer test, nuclide migration, natural condition, aerated layer, aquifer layer, Np-237, Pu-238, Sr-90, cooperative research

1 Introduction

A six-year cooperative research program between the Japan Atomic Energy Research Institute (JAERI) and the China Institute for Radiation Protection (CIRP) has been conducted as the phase-2 program to establish a safety evaluation methodology for shallow land disposal of low level radioactive waste. The phase-1 program was conducted from 1989 to 1994 to test radionuclide migration behavior of Sr-90, Co-60 and Cs-134 in aerated loess layer under both artificial and natural rainfall conditions, and to evaluate the validity of the migration model combining advection-dispersion equation with instantaneous equilibrium model[1].

Followed by the phase-1 program, the phase-2 program has been started with three additional test items, such that TRU nuclides migration, performance of engineered barrier materials, and nuclide migration in aquifer zone. The phase-2 program consists of tracer experiments in aerated and aquifer layers at a CIRP's field test site, large scale simulation experiments using undisturbed soil, laboratory experiments, investigation of the site characteristics, and migration analysis with an appropriate model.

Tracer experiment in aerated layer is performed to test tracer migration behavior in both engineered materials (cement, bentonite, mortar) and natural barrier material (loess soil) using H-3, Sr-90, Np-237, Pu-238 for radioactive tracers, or Br, Sr, Ce, Nd for stable tracers. For performing the tracer experiments in natural aquifer layer, an Underground Research Facility (URF) has been constructed to directly access to the aquifer layer of about 31 m deep in the site[2]. Groundwater samples for H-3 and Br concentration have been analyzed to obtain data on groundwater flow. Large scale simulation experiments are conducted with undisturbed soil taken from the aerated and aquifer layers in the site under a controlled flow condition. Conventional migration model will be verified by comparing the migration distribution of nuclides obtained from the field migration experiments with that evaluated by using the values of necessary parameters analyzed and obtained from the laboratory experiments, large scale simulation experiments, and the survey of site characteristics.

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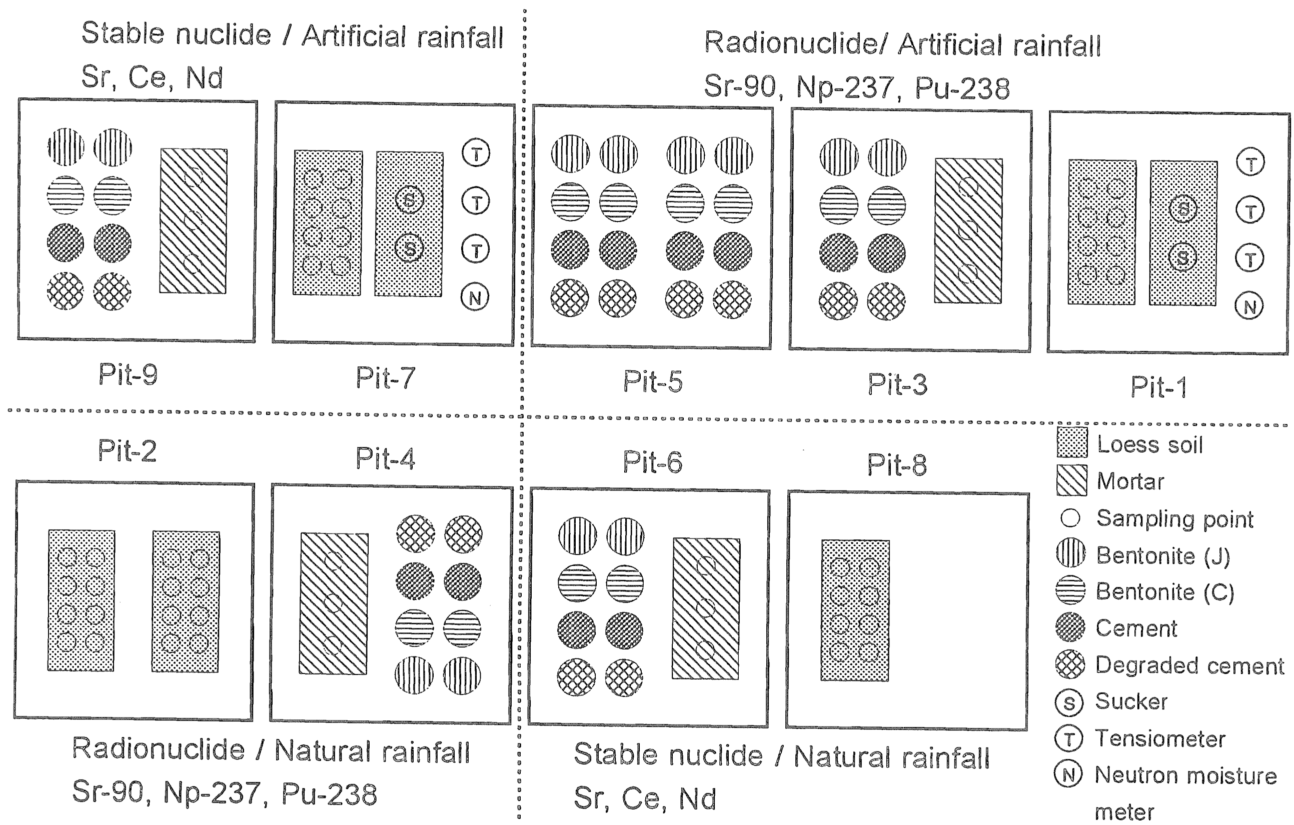

Fig. 1　Plane view of pit arrangement for aerated field tracer test.

2 Field migration test

2.1 Migration test in aerated zone

The aerated migration test is performed in nine experimental pits dug in the ground surface at the CIRP's field test site. Test condition of the tests can be summarized in a combination of tracer type, rainfall condition, and testing material as shown in the fig. 1. All the tests use Sr-90, Np-237, Pu-238 and H-3 for radioactive tracers, or Sr, Nd, Ce and Br for stable element tracers. Tritium and Br are used as a tracer for soil water movement because of their negligible adsorbability. Four of the nine pits are tested under artificial rainfall condition by showering water, and the rest is for natural rainfall condition. The pits for the artificial rainfall condition are dug in ground surface in a experimental housing constructed beside the pits for natural rainfall condition. To apply artificial rainfall at a constant amount a day with a reasonable homogeneity, water showering device has been developed by arranging a lot of injection needles overlaid the pit to apply water drops onto it. The intensity of the artificial rainfall is controlled at a rate of 5 mm/hr and each pit is showered for 3 hours a day. Materials for the test are loess soil for natural barrier, cement, bentonite, and mortar for engineered barrier.

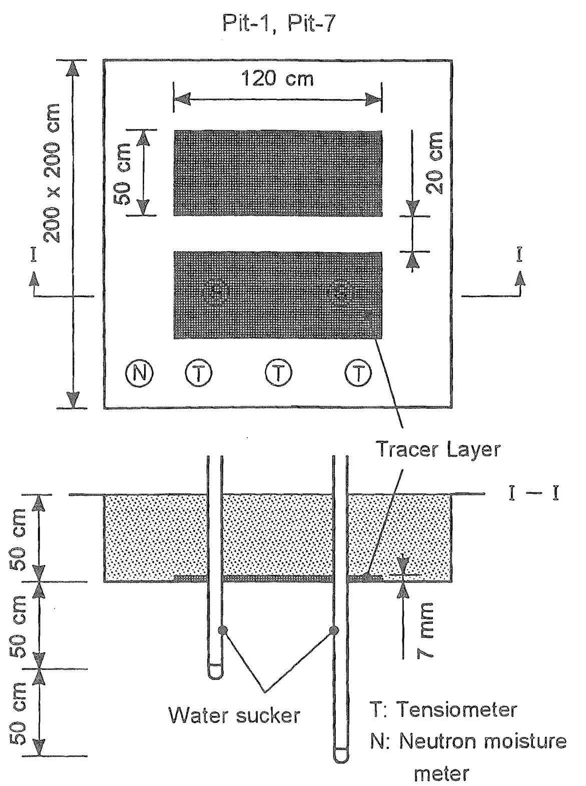

Fig. 2　Cross-sectional view of the pit for natural barrier material.

2.1.1 Test for natural barrier material

Figure 2 shows a cross section of the pits for natural barrier. Pits were dug to 50 cm deep from the ground surface, and the bottom of the pits was leveled. Then, a pipe for neutron moisture observation, tensiometers, and soil water suckers were installed. To place tracer source material at a proper position and size, a polyethylene frame with 120×80 cm inside length and thickness of 7 mm was laid on the bottom and then tracer source material was filled up to the rim of the frame. Quartz sand with 60 ~ 80 mesh is used as the tracer source material for the pits under artificial rainfall condition while loess soil for the pits under natural rainfall condition.

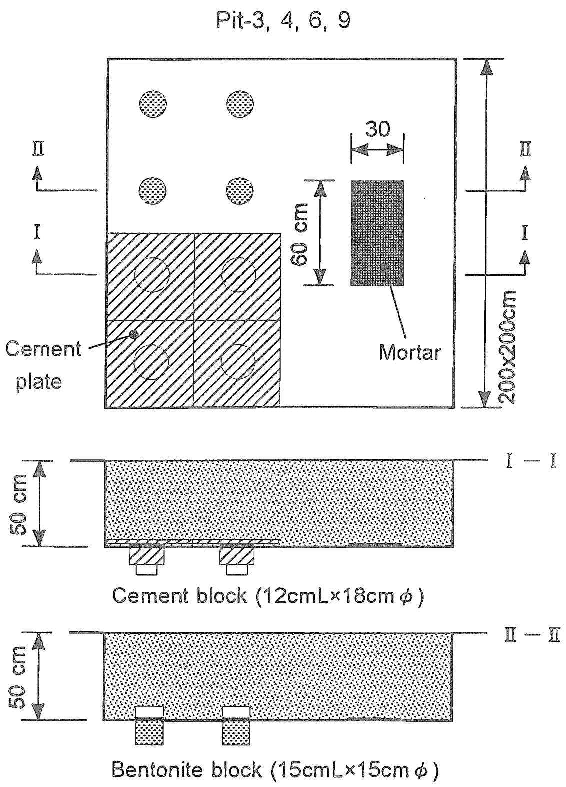

Fig. 3 Cross-sectional view of the pit for engineered barrier material.

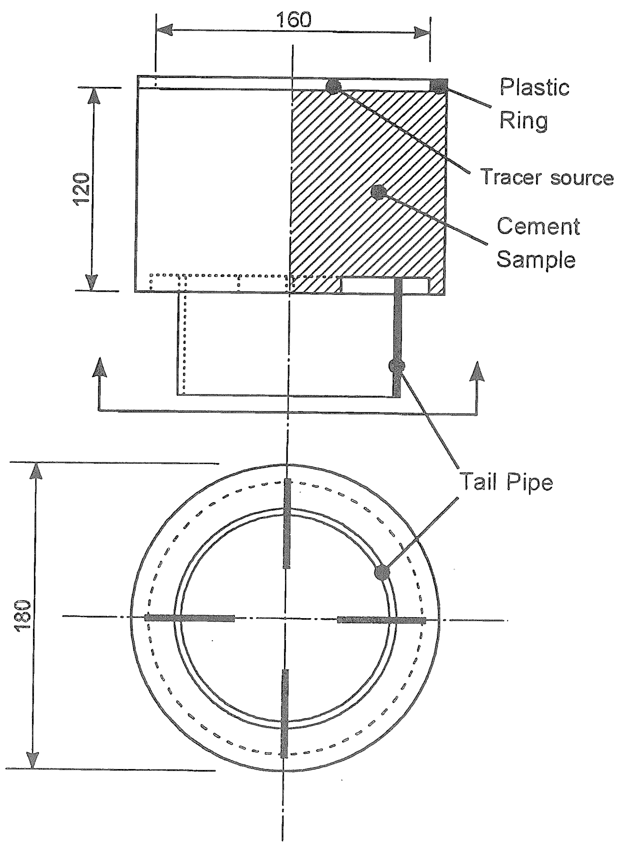

Fig. 4 Structure of cement block sample.

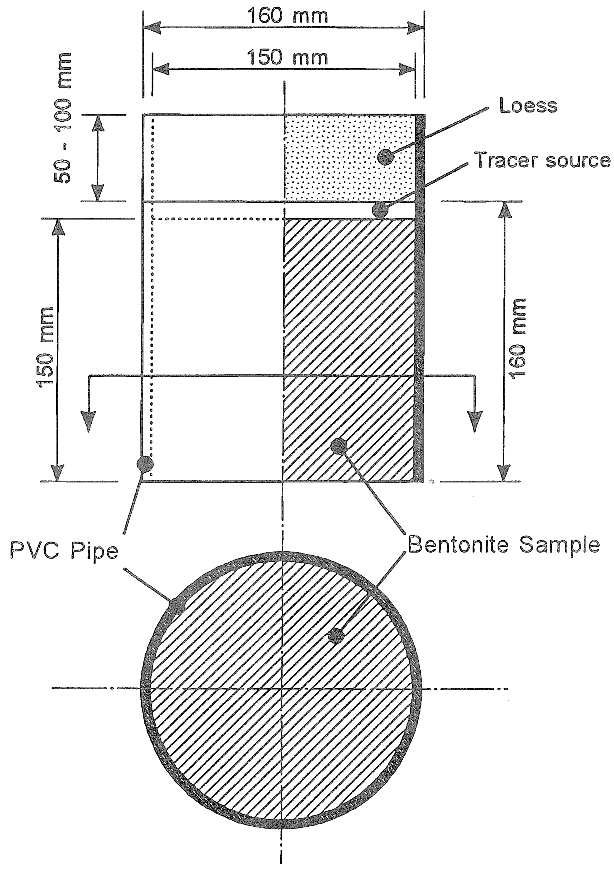

Fig. 5 Structure of bentonite block sample.

2.1.2 Test for engineered barrier material

Figure 3 shows a cross section of the pits for engineered barrier. Placement procedure for mortar is the same as that for the tracer source applied to the pits for natural barrier material. Mortal is tested as the form of powder while cement and bentonite are tested as the form of block. Figure 4 and fig. 5 show schematic illustration of cement and bentonite block samples, respectively. Tracer source material used for the both blocks is quartz sand with grain size of 60 ~ 80 mesh.

The cement block for the test is made by pouring cement slurry, which is mixture of common silicate cement and water at a ratio of 1 : 0.3, into the mold with an inside diameter of 18 cm. Degraded cement, which is made by mixing 3% $NaHCO_3$ and 3% Na_2SO_4 solution and the cement at a ratio of 0.34 : 1, is also tested to observe the effect of degradation on tracer migration for long term performance.

Two kinds of bentonite are tested in the research program. Physico-chemical properties of the two bentonite are listed on the table 1. Japanese bentonite (Kunigel V1) is Na-bentonite, while Chinese one is Ca-bentonite. The blocks of two kinds of bentonite are prepared in the same way by mixing bentonite and common construction sand at a ratio of 15 : 85 and pouring into a PVC pipe with an inside diameter of 15 cm and 16 cm in hight.

Table 1 Phisico-chemical properties of bentonite.

ITEMS	Japanese bentonite	Chinese bentonite
Particle density		2.61
Water content (%)	6.68	30.44
Dry bulk density (g/cm^3)		1.31
Saturation ratio (%)		80.25
Liquid limit (%)		99.8
Plastic limit (%)		41.0
Plastic index		58.8
Total surface area (m^2/g)		617.48
Particle distribution (mm,%)		
> 0.25		3.09
0.25-0.10		0.92
0.10-0.05		22.69
0.05-0.01		8.4
0.01-0.005		5.2
< 0.005		59.7
pH	10.06	7.32
Cation exchange capacity (mmol/100g)	60.0	83.40
Exchangeable K^+ (mmol/100g)	0.9	0.34
Exchangeable Na^+ (mmol/100g)	55.6	1.60
Exchangeable Ca^{2+} (mmol/100g)	24.70	52.92
Exchangeable Mg^+ (mmol/100g)	8.25	27.86

Apparent density of the bentnite sample after compaction is calculated to be 1.80 g/cm^3.

Test material of mortar is prepared by mixing cement, sand, and water at a ratio of 1 : 2.5 : 0.55, and crashed into powder after 28-day solidification. The mortal powder with grain size smaller than 2 mm is sieved and used in the test.

2.1.3 Sampling of soil water

Sampling of soil water is performed in the pits for tracer migration in the natural barrier material under the artificial rainfall condition. Soil in the pits for natural rainfall condition is too dry to suck soil water sample. The suckers are installed at depths of 1.0 m and 1.5 m deep in the pit-1 and pit-7. Pit-1 is for the migration test of radionuclide tracer and pit-7 for stable element tracer. Tritium concentration in water samples is measured using low-background scintillation counter, Br concentration determined by colorimetric method.

Figure 6 shows the temporal change of H-3 concentration in the sucked soil water at the two depths in the pit-1 and that of Br in the pit-7. From the time difference between two concentration peaks observed at 1.0 m and 1.5 m deep, water flow rates in the two pits can be calculated. The water flow rate is found to be 2.17 cm/day for the pit-1 and 2.04 cm/day for the pit-7.

2.1.4 Sampling of test material

Core sampling method is employed for soil sampling in the pits for natural barrier material and mortar. Sampling in the pits for natural barrier material is conducted basically two times a year. In the pits for natural barrier and mortar, core

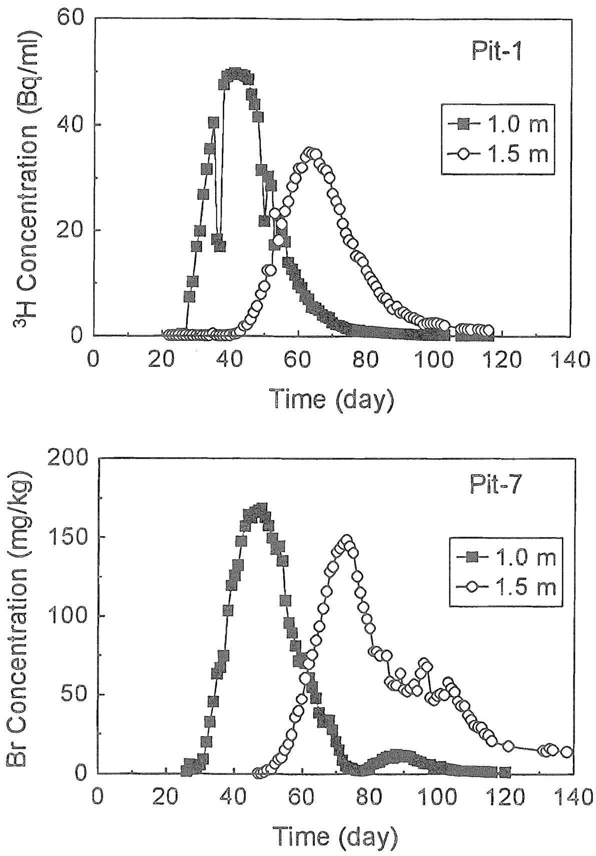

Fig. 6 Change of tracer concentration in soil water sucked in the pit-1 and pit-7.

sample is taken by inserting a sampling sleeve using a manual operation device into the pits to measure detailed vertical distribution of tracer migration. In the sampling sleeve, ten sampling tubes with an inside diameter of 4 cm and a length of 4 cm are set to collect and hold soil sample material. After sampling, each sampling tube is taken out of the sampling sleeve, and then the soil sample is successively pushed out to be sliced at a certain thickness. Measurements of Np-237 and Pu-238 are made mainly by gamma spectrometry in conjunction with radiochemical analysis and alpha spectrometry. Sr-90 is measured by radiological separation followed by beta counting,

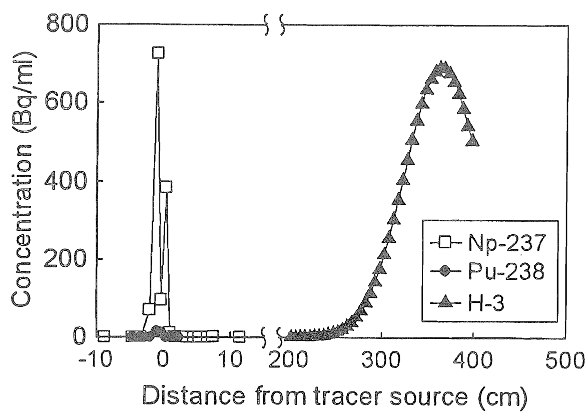

Fig. 7 Longitudinal distribution of Np-237 and Pu-238 in soil layer after 143 days.

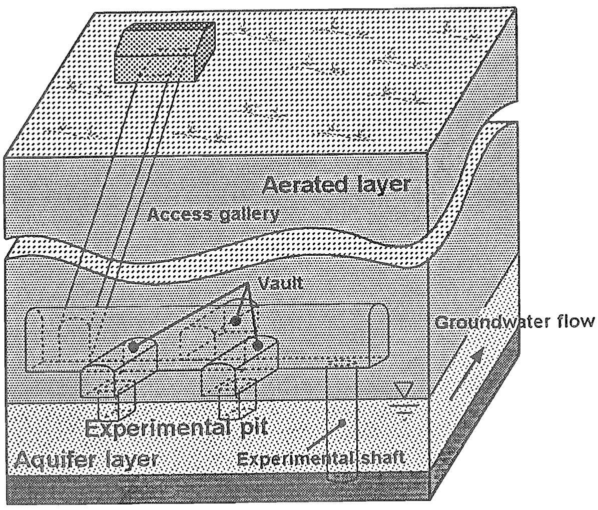

Fig. 8 Schematic view of the Underground Research Facility (URF).

which is standardized in China. All stable element tracers are measured by using ICP-AES (inductively coupled plasma atomic emission spectrometry). Figure 7 shows longitudinal distribution of Np-237 and Pu-238 concentrations obtained from the first soil sampling at 143 days after from the start of artificial rainfall in the pit-1. Concentration of H-3 shown in the fig. is relative value calculated using flow rate obtained by peak analysis described in 2.1.3 to compare water movement with the migration of Np-237 and Pu-238. These radionuclide tracers have remained at the initial position.

Cement and bentonite blocks have been periodically taken out of the pit to measure longitudinal distribution of tracer concentration. Since tracers migrate within a very small extent in both cement and bentonite blocks, a cutting device has been developed utilizing a lathe to take sample with a precise thickness control of up to sub-millimeter order. The device is set in a hood with ventilation to avoid unexpected contamination.

2.2 Migration test in aquifer zone

2.2.1 Underground Research Facility (URF)

To perform the aquifer migration test in the actual aquifer layer under the natural groundwater flow condition, the URF has been constructed in the CIRP's field test site. Schematic view of the URF is shown in the fig. 8. To minimize the construction impact upon the quality of the surrounding groundwater and avoid direct contact between concrete and soil layer, polyethylene sheets and water-proof coating material were applied to the wall of the URF. The URF provides one experimental shaft and three experimental vaults at the depth of about 29 m underneath the ground surface. The pit experiment tests three dimensional migration of both radioactive and stable element tracers under natural groundwater flow condition, while the shaft experiment tests tracer migration under various flow conditions.

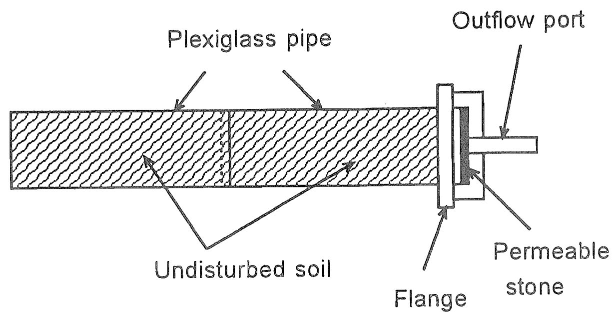

Fig. 9 Schematic illustration of the assembly.

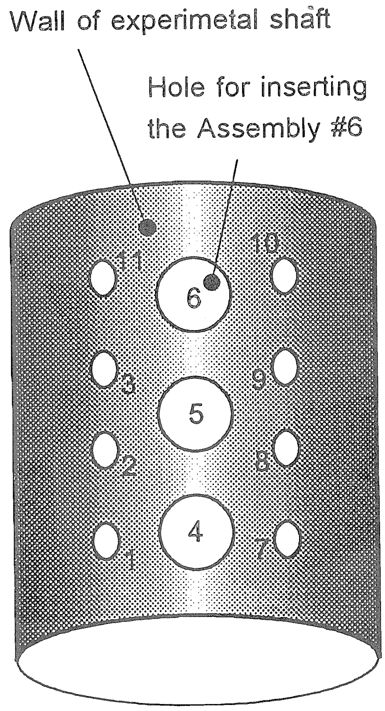

Fig. 10 Arrangement of testing holes on the wall of experimental shaft.

2.2.2 Assembly test

The lower part of the experimental shaft is constructed under groundwater table for the in-situ column experiment using experimental column set of undisturbed soil referred as assembly. The assembly is composed of two section of plexiglass pipes and an outflow port connected with flange at the end of the pipe (fig. 9). To place the tracer source in the soil column, the assembly column is cut along the gap between the two sections. On the opened surface of the soil column, two sizes of tracer source were applied. One is point source with a length of 1.8 cm and a diameter of 2.3 cm, and the other is plane source with a thickness of 5 mm. The both are mixture of tracer and quartz sand with 60 ~ 80 mesh.

Tracer migration in the assembly is tested by installing the

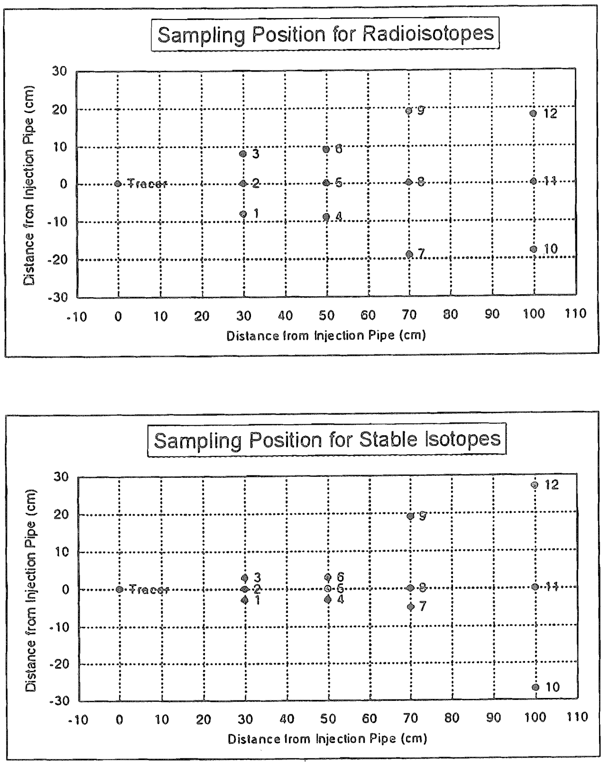

Fig. 11 Arrangement of injection and sampling pipes in the experimental pits.

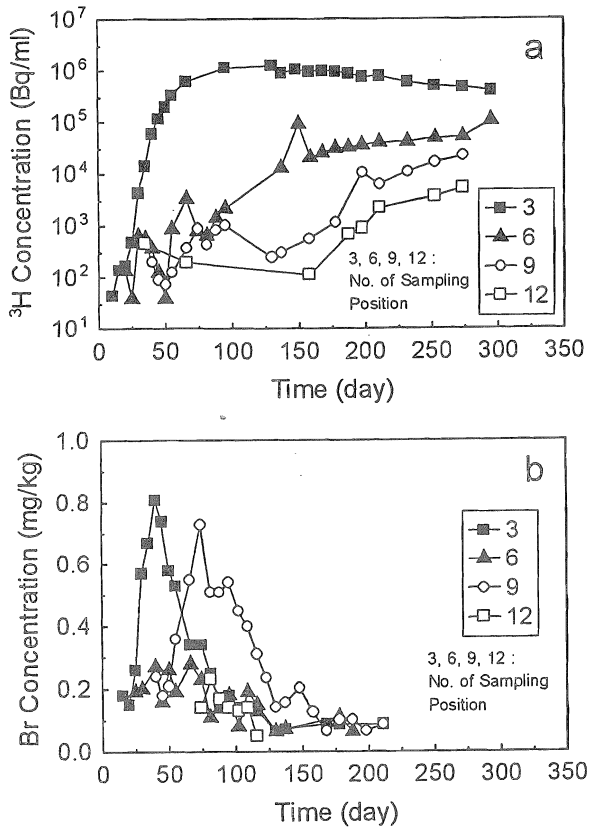

Fig. 12 Change of H-3 and Br concentration in groundwater sampled in the experimental pits.(3, 6, 9, 12 : No. of sampling position)

assembly into the wall of the shaft (fig. 10), and passing in-situ groundwater through the assembly. Flow rate of the groundwater is controlled by a attached valve on the end of the outflow port of the column. To accelerate tracer migration, flow rate is set to several times higher than the rate under the natural condition. Effluent solution out of the assembly is collected and measured to evaluate the concentration of tracers in it. Change of concentration of H-3 and Br in the effluent is to be a good indication of water flow heterogeneity. In the case of the assembly #4 and #5, irregular changes of H-3 and Br concentration, that is thought to indicate by-pass groundwater flow in the columns, were found, thus the columns have been replaced and restarted a year after.

2.2.3 Pit tests

Two of three experiment vaults are provided for experimental pits for testing three dimensional migration behavior of radionuclide and stable element tracers under natural groundwater flow condition. The dimension of the both pits is the same with 2 m long and 1.5 m in width. In the each pit, one injection pipe and twelve sampling pipes are inserted into the aquifer layer up to the depth of about 2.6 m below the floor of the URF. This depth is corresponding to about 0.6 m underneath the groundwater table.

The sampling pipe has three different suction ports apart 3 ~ 8 cm each other to take groundwater sample at three different depths with minimum disturbance in the aquifer layer. The sampling pipes are arranged in a sector form, vertex of which is so as to be positioned at the injection pipe, to take groundwater samples on the downstream and to obtain three dimensional distribution of H-3 and Br concentration as shown in the fig. 11. Groundwater has been periodically collected from the aquifer layer using a sampling device capable to suck groundwater by applying a vacuum pressure. Each volume of a groundwater sample is up to 10 ml.

Interim result of the change of H-3 and Br concentration in sampled groundwater is shown in the fig. 12a and fig. 12b. Figure 12a shows the change of H-3 concentration at the position of 30 cm downstream from the injection pipe. The concentration peak has been measured after 120 days from the starting. On the other hand, peak of Br concentration has been observed much faster period of 44 days than that of H-3 as shown in the fig. 12b at the the same distance as the pit for radionuclide tracer. This is considered at the moment to be caused by the difference of hydraulic gradient of groundwater table and/or hydraulic conductivity in the two pits. Groundwater table distribution in the URL shows that the gradient of groundwater table in the both vault are virtually the same, hence water conductivity of the aquifer layer in the pits, the other essential parameter to calculate groundwater flow rate, will be measured after completion of the tests.

3 Simulation migration test

Simulation migration test is conducted to obtain

Table 2 Test conditions of simulation migration test using aerated layer.

Column No.	Test medium	Dimension of column (cm)	Tracer layer (cm)	Tracer	Tracer amount	Sprinkling	Duration
1	Aerated soil	ID = 28 L = 120	Dia=28 T = 0.5	^{237}Np ^{238}Pu ^{90}Sr	4.7MBq 0.85MBq 15MBq	Distilled water ~5mm/d	3 years
2		ID = 28 L = 120	Dia=28 T = 0.5	^{237}Np ^{238}Pu ^{90}Sr	4.7MBq 0.85MBq 15MBq		2 years
3		ID = 28 L = 120	Dia=28 T = 0.7	Ce Nd Sr	5.7 g 2.8 g 16.5 g		2 years
4		ID = 15 L = 120	Dia=15 T = 0.7	Ce Nd Sr	1.6 g 0.8 g 14.7 g		1 year
A	Aerated soil	ID=15 L = 95	Effect of wick rope				-
B		ID = 15 L = 95					-
C		ID = 28 L = 120	Measure water flow velocity				-

ID : Inside Diameter L : Length Dia : Diameter T : Thickness

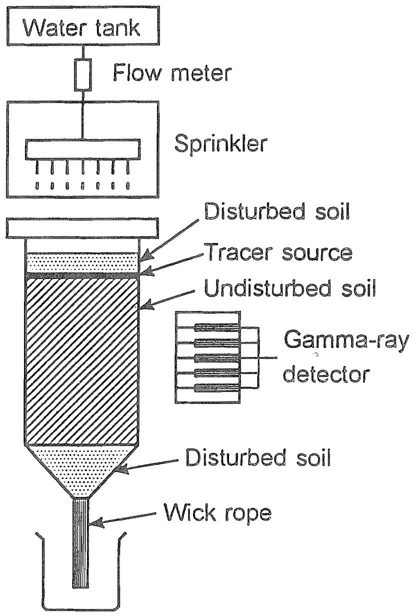

Fig. 13 Schematic illustration of apparatus for simulation migration test using aerated layer.

supporting data for the field test by using undisturbed samples taken from the same location as the field test is being conducted. Because water flow rate in the test is set to several times higher than the field test condition, the longer distance of migration can be expected to obtain distinct distribution and to yield reliable value for migration parameters.

3.1 Migration test using aerated layer

The migration test is conducted in laboratory of the CIRP using undisturbed soil samples taken from the field testing site where the field migration test is conducted. Schematic illustration of testing apparatus is shown in the fig. 13. Four columns (No. 1, 2, 3, 4) were provided to perform the test under different conditions shown in the Table 2. Two are tested using radioactive tracers and the other for stable element tracers. One of each column for radioactive and stable element tracers will be tested for 3 years and the others for 1.5 years. Test columns of No. A, B, C are used to study water movement using I-131 by detecting outside of columns undestructively, and the effect of a wick rope, tightly filled nyron fiber in a plastic tube with diameter of 2 cm, to reduce water content at the lower part of columns. Preliminary result shows that the relative radiation count of I-131 is successfully measured by gamma detectors with a collimator, and the vertical flow rate is found to be retarded at the lower position of the culumn with increasing water content. As to the effect of the wick rope, water content in the columns is reduced by connecting the wick rope to the outflow port of the column from 0.35 ~ 0.40 to 0.20 ~ 0.25 at the middle part and from saturated to 0.30 at the lower part. This distribution of the water content is close to that in the field tracer test in aerated zone under artificial rainfall condition.

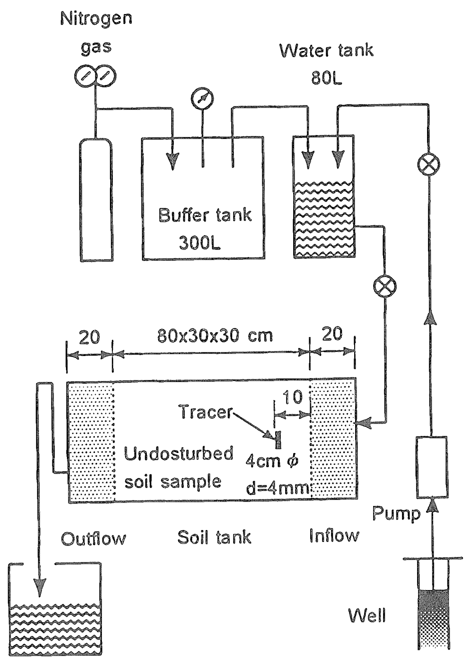

Fig. 14 Schematic illustration of apparatus for simulation migration test using aquifer layer.

Table 3 Test conditions of simulation migration test using aquifer layer.

Tank No.	Test period	Tracer amount
1	1.5 years	^{3}H 52MBq ^{90}Sr 6.03 MBq, ^{237}Np 0.489 MBq, ^{238}Pu 0.397 MBq (for one tank)
2	3 years	
3	1.5 years	Nd 0.186g, Ce 0.365g, Sr 1.090g (for one tank)
4	3 years	

3.2 Migration test using aquifer layer

The migration test is carried out in the URF using undisturbed soil sample from the aquifer layer. In addition to the undisturbed soil sample, the test uses in-situ groundwater sampled in the URF observation well and the water directly introduced into the soil sample without contact to atmosphere to simulate chemical condition in the field test being conducted. Test condition is shown in the table 3. The four test tanks have the same structure, length: 120 cm, inside section: 30×30 cm as shown in the fig. 14. Inflow and outflow sections at the both ends of the tank are filled with gravel (1 ~ 2 cm in diameter) and quartz sand (80 ~ 120 mesh). Tracer source with 4 cm in diameter and thickness of 4 mm is placed on the center axis at a distance of 10 cm from the inflow section. Effluent water out of the outflow tank has been periodically measured on H-3 or Br concentration to monitor water flow condition. The soil tank is decomposed after a determined period to obtain precise distribution of tracer concentration.

4 Laboratory test

For analysis on the field migration test and the simulation migration test, distribution coefficient (*Kd*) and diffusion coefficient for aerated and aquifer layer soil, bentonite, and mortar are measured in laboratory. Batch experiment is employed to obtain *Kd* value under various initial concentrations and contact time. Initial pH and liquid/solid ratio are set to 8.5 and 20 for the aerated and aquifer layer soil taking account of natural condition.

5 Site characterization

5.1 Groundwater level

Groundwater level on the CIRP's field testing site and in the URF are measured to determine the distribution of groundwater table and to obtain information on groundwater flow direction and rate. Twelve wells are available in and around the field test site and eight observation wells are used in the URF. Interval of the measurement is once a month for the well on the site by manual operation, and once a day for the well in the URF by an automatic acquisition monitor.

5.2 Meteorological observation

To measure infiltration amount into ground surface where the aerated tracer test is conducted, the amount of precipitation and evaporation is observed on the field test site. Automatic rainfall gauge, wind velocity meter, psychrometer, net radiation meter and soil heat flux meter are installed and being measured in the field testing site. Information from these instruments is compiled to calculate temporal change of evaporation amount by the Penman method and energy balance method.

6 Migration analysis

Migration analysis will be performed to discuss what degree the numerical calculation code can simulate the distribution of tracer migration in the tests. Conventional models of water flow and tracer migration, which consist of a combination of advection-diffusion equation and instantaneous equilibrium model of chemical reaction, are used in the calculation together with input data of parameters collected by various ways in the research program. If the conventional models do not have high validity, new models and parameters should be proposed for the improvement.

As the first step of the migration analysis, items of input data necessary for the migration analysis have been being summarized, and then values of each item are collected to input to the models to calculate. Distribution of tracers at the time when the field test is completed will be estimated using the obtained data. Estimated distribution will be compared with the actual distribution of tracers after the completion of the field test in 2000.

7 Concluding remarks

A six-year research program has been conducted between JAERI and CIRP to establish a safety evaluation methodology for shallow land disposal of low level radioactive waste;

1. Field tests of tracer migration in natural aerated and aquifer layers are being conducted in the CIRP's testing site,
2. Materials for natural barrier (loess soil) and engineered barrier (cement, bentonite, mortar) are tested in the aerated field test under natural and artificial rainfall conditions in nine pits on the ground surface,
3. Result at a half year artificial rainfall shows negligible migration of Np-237 and Pu-238 in loess soil compared to water movement calculated using H-3 tracer,
4. Two types of aquifer field test (assembly test and pit tests) are being conducted in the URF which allows direct access to the natural aquifer layer in the site,
5. The assembly test is in-situ migration test using undisturbed soil column at the lower part of experimental

shaft underneath the groundwater table,

6. The pit tests are performed in natural aquifer layer under natural groundwater flow condition to obtain data on three dimensional migration of both radioactive and stable element tracers,
7. No tracer besides H-3 and Br has been detected in the sampled groundwater in the sampling pipe located 30 cm downstream,
8. Migration tests using several 10 cm scale undisturbed aerated and aquifer soil sample are conducted under various conditions simulated the field test,
9. Laboratory experiment and site characterization for the evaluation of Kd, groundwater level, and infiltration amount are successively conducted for supporting migration analysis,
10. The migration analysis will be performed to evaluate validity of conventional models using obtained data from all the tests in the research program.

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