THE STUDY ON THE MIGRATION OF RADIONUCLIDES IN THE SHALLOW LAND

Shushen Li Zhiming Wang Zhentang Li Yingjie Zhao
Zede Guo Liangtian Guo Yingxia Shi
(China Institute for Radiation Protection)

Hiromichi Ogawa Toshikatsu Maeda Junko Matsumoto
Masayuki Mukai Tadao Tanaka
(Japan Atomic Energy Research Institute)

ABSTRACT

From 1995 through 2001 a cooperative study project on the migration of radionuclides in shallow land was carried out by CIRP and JAERI, which covers field test, laboratory simulation test , other laboratory studies and related model development. The radionuclides studied involve ^{90}Sr, ^{237}Np, ^{238}Pu. For comparison the nonradioactive elements Sr, Nd and Ce were also studied.

The field test was performed both in aerated zone and aquifer zone of loess. In the aerated zone the nuclide migration in engineering materials were also studied. The study in the aerated zone was carried out in 9 pits with the size of 2m×2m under natural conditions or artificial sprinkling conditions. The study in the aquifer was carried out in a new built Underground Research Facility with the area of 142m^2. The test results show that the order of adsorption activity of the nuclide on the loess is ^{238}Pu>^{237}Np>^{90}Sr and Nd, Ce>Sr.

During the 3 years period of test the migration of ^{238}Pu and Nd, Ce was not observable in both aerated zone and aquifer zone, the nuclide of ^{237}Np migrated a small distance, and the nuclide of ^{90}Sr had a relative large migration. The migration of the nuclides in engineering materials was not detected, which include cement, degraded cement, cement mortar, Chinese bentonite and Japanese bentonite.

INTRODUCTION

Wide varieties of nuclear applications have generated a wide range of radioactive wastes, 95% of which are, in terms of volume, classified to low-and intermediate-level radioactive wastes (LILWs). Shallow land disposal is an approach mainly for radioactive wastes bearing short-lived radionuclides, and also for the low level radioactive waste containing acceptable levels of long-lived nuclides.

The CJ2 Project was conducted during August 1995 to August 2001 by a joint co-operative effort by China Institute for Radiation Protection (CIRP) and Japan Atomic Energy Research Institute (JAERI) in collaboration with other 5 Chinese scientific institutions. It was a continued, deepened and advanced practice of the CJ1 Project, which was carried out during time period of January 1988 to January 1993. The CJ2 Project's focuses are on studying the nuclide migration behavior in loess aerated zone , loess aquifer and in aerated man-made barriers (cement block, degraded cement block and bentonite); environmental factors governing nuclide migration; and chemical speciation of long-lived radionuclides in given conditions. A number of innovative efforts have been made for the purpose of providing an effective set of approaches and methods, including parameters, models and codes, for safety assessment of shallow land disposal of LLWs involving predication of nuclear pollutant migration in shallow land and radiation environmental impact assessment. The results of the present project are also applicable to the safety assessment of middle-depth disposal of transuranic nuclides.

CONTENTS OF CJ2 PROJECT

The primary fieldwork of the CJ2 Project was conducted in loess aerated zone and groundwater aquifer at the CIRP's Field Test Site located in the semi-arid region in North China. Laboratory study and simulation were performed separately. In order to determine the appropriate scale of field nuclide migration test to be conducted, such as time length, nuclide input amount, sprinkling intensity, both the nuclide simulation experiment and numerical modeling calculation were first carried out in laboratory. With ^{3}H and Br^{-} as tracers, water velocities in aerated zone and aquifer were respectively determined; field site characterization survey and moisture transport pattern study were carried out, numerical modeling study was conducted using the data available from the relevant research work. The CJ2 Project included 13 Sub-projects and 59 topics. The summarization is given as follows.

- Development of safety assessment methodology for waste disposal
- Site characterization survey
- Study of moisture transport pattern at CIRP field test site
- Design and construction of URF
- Tracer source preparation and tracer analysis methods
- laboratory study on nuclide sorption and speciation
- In situ tracer tests in aquifer
- In situ tracer tests in aerated zone
- In situ test of nuclide migration in engineering barrier
- Simulated nuclide migration tests in aquifer
- Simulated nuclide migration tests in aerated zone
- Assessment model and procedure development and validation
- Quality assurance system and safety management

Field tests and laboratory simulation experiment were conducted in a synchronized way. That includes: (a) field tracer tests (radionuclides and stable elements) and laboratory simulation experiments for aerated zone nuclide migration, (b) field tracer tests and field simulation experiment for aquifer nuclide migration, (c) mixed ^{3}H and Br^{-} migration tests, and (d) field tests and laboratory column experiments.

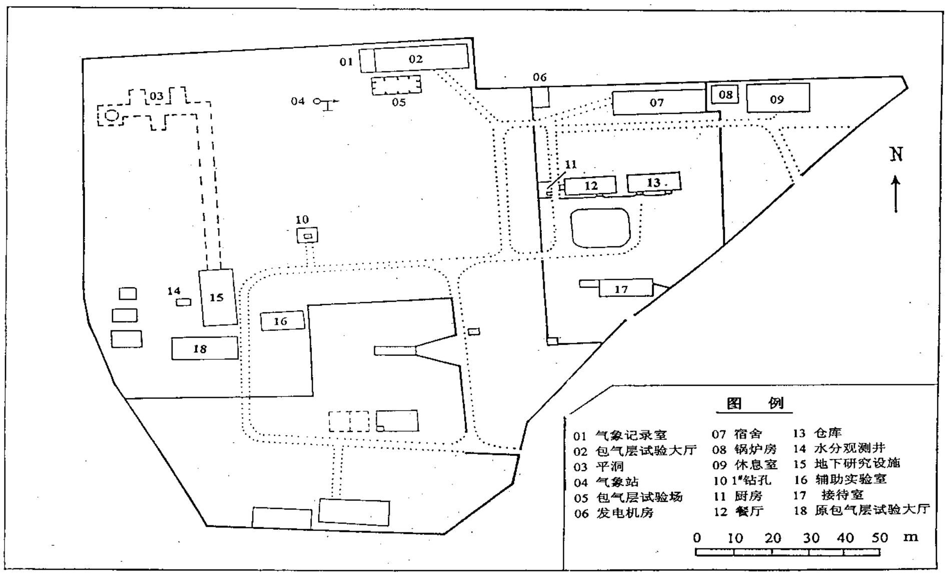
DESCRIPTION OF TESTS

In this paper the introduction stress is placed on the field tracer migration test in loess aquifer and in loess aerated zone, conducted from Jun. 1997 through Aug. 2000. In these tests two kinds of tracers were used, radioactive nuclides ^{3}H, ^{90}Sr, ^{237}Np, ^{238}Pu and stable elements Br^{-}, Sr, Nd and Ce.

Field Tracer Migration Test in Loess Aquifer

Field tracer migration test in aquifer is an important part of the methodology study on safety assessment of radioactive waste disposal. The test mainly consists of 3 parts: (a) the tracer migration test in the assemblies (Type I); (b) the radioactive tracer migration test in aquifer with a frame (Type II); (c) the stable element migration test in aquifer (Type III). Type II and Type III were directly conducted under undisturbed aqueous medium, and Type I was conducted in the assemblies filled by undisturbed aqueous medium sampled from aquifer.

This kind of test was conducted in the Underground Research Facility (URF) located at the CIRP's field test site. Fig. 1 shows the layout of the CIRP's field test site.



Note: 01 meteorological recording room, 02 aerated zone test hall, 03 gallery of URF, 04 meteorological observation station, 05aerated zone test site, 10 1# drilling hole, 14 observation well of moisture movement 15 the inlet of URF

Fig.1 The layout of the CIRP's field test site.

The URF was built at the 26m deep horizon and is 2m above aquifer level. It has a gallery measuring 29m long × 3m wide × 4m high and 3 vaults at wall side, each with the dimension of 5m long × 3m wide ×3m high. There is an experimental shaft measuring □ 2.5 m × 6 m deep (4 m within aquifer) built in the gallery. An inclined shaft measuring 68 m long × 2.8 m wide × 2.8 m high with 29 degree slope, is an exit to the ground surface. During the construction of URF, a great deal of technical measures were taken to ensure no change in groundwater flow field, no influence upon groundwater quality and no damage to stratum stress.

- **Tracer Migration Test in Assembly (Type I)**

In this test eight assemblies were inserted into aquifer zone through wall of experimental shaft of the URF as shown in Fig 2. The assembly is a loess column with a tracer layer . The details of the assemblies are listed in Table I.

Table I. Description of the assemblies in aquifer

No.	Size(cm)	Tracer	Tracer source		Note
			Shape	Size(cm)	
$2^{\#}$	Φ10.8×100	Radionuclides	Plane	Φ10.8, h 0.5	Undisturbed loess
$3^{\#}$	Φ10.8×100	Radionuclides	Plane	Φ10.8, h 0.5	Undisturbed loess□ humic acid:1g/100g loess
$6^{\#}$	Φ24×100	Radionuclides	spot	Φ1.4, h 2.4	Undisturbed loess□ humic acid:1g/100g loess
$7^{\#}$	Φ10.8×100	Stable elements	Plane	Φ10.8, h 0.5	Undisturbed loess
$8^{\#}$	Φ10.8×100	Radionuclides	Plane	Φ10.8, h 0.5	Undisturbed loess□ humic acid:1g/100g loess
$9^{\#}$	Φ10.8×100	Radionuclides	Spot	Φ1.4, h 2.4	Undisturbed loess
$4A^{\#}$	Φ24×100	Stable elements	Spot	Φ1.5, h 0.4	disturbed loess
$5A^{\#}$	Φ24×100	Radionuclides	Spot	Φ1.5, h 2.4	disturbed loess

During the test, one end of the assembly contacts closely with the loess aquifer. The groundwater passing through the assembly flows out from the its other end. The effluent passing through the assemblies was collected into sample bottle for measurement.

- **Tracer Migration Test in Undisturbed Aqueous Medium (Type II)**

The frame was designed to be a stainless steel hexahedron structure, with three sides being solid and the other three sides empty. The left and right sides are made of stainless steel plate with holes. The downstream side is built with double layers of stainless steel with holes drilled. Between the two layers, a piece of 100-mesh stainless steel net filled with the mixed anion and cation exchange resins is affixed. The frame measures 80 cm long × 40 cm wide × 180 cm high of which 120 cm high part was inserted into aquifer as shown in Fig.3. It was inserted into ground by means of an about 100 t pressing machine.

The purpose of the frame test is to acquire the three dimension data on their migration.

Field Tracer Test for Nuclide Migration in Aerated Zone

- **Field Tracer Test for Nuclide Migration in loessial Aerated Zone**

The test in aerated zones was carried out at the CIRP's Field Test Site. The tests were conducted under both artificial sprinkling and natural conditions. As shown in Fig. 4.

The test pits 1, 7, 11 under artificial sprinkling condition were located in a specially designed hall with sprinkling intensity 15 mm/d by two sets of artificial sprinkling device. The test pits 2, 8 under natural condition were on an open field in the front of the hall. The soil sample columns of the test under artificial sprinkling conditions were taken every 6 months, and the soil sample columns of the test under natural conditions were taken every year.

In addition to obtaining the information about tracer migration, there is still a need to understand the conditions under which nuclide migrates. For this purpose, the moisture transport pattern study was conducted in a synchronized way indoors and outdoors. For the purpose of studying the influence of quartz tracer layer on unsaturated water flow permeability, a comparative test of water infiltration was specially designed and performed on both laboratory simulation and field test scales. Apart from this, a comparative test of nuclide migration from both quartz sand tracer layer and soil tracer layer were carried out.

- **Field tracer test for nuclide migration in engineering barrier materials placed in aerated zone**

Wide ranges of study of nuclide migration in 24 bentonite sample blocks (Chinese and Japanese), 12 cement sample blocks and 12 degraded cement sample blocks were conducted in pits 3, 5, 9 under artificial sprinkling conditions and in pits 4,6 under natural conditions as shown in Fig. 4. The study of nuclide migration in loess with cement mortar as tracer layer was conducted in pit 3,9 and 4,6. The sampling cycles are the same as the tests in loess.

Simulated Test Study

The simulated test study includes following two parts.

(1) the study of migrations of ^{90}Sr, ^{237}Np and ^{238}Pu and Sr, Ce and Nd in aquifer loess tanks with the water velocity of 6.13cm/d

(2) the study of migrations of ^{90}Sr, ^{237}Np and ^{238}Pu and Sr, Ce and Nd in aerated loess columns with the water velocity of 5.5mm/d

Laboratory Study

the laboratory study includes following three parts.

(1) the adsorption of ^{90}Sr, ^{237}Np and ^{238}Pu and Sr, Ce and Nd on loess and engineering barrier materials

(2) the defusion of ^{90}Sr, ^{237}Np and ^{238}Pu on engineering barrier materials

(3)the speciation of ^{237}Np and ^{238}Pu in different conditions

Model Validation and Analyses

Through scenario analysis and experiment/test study, the safety assessment was made for the disposal of LILWs containing transuranic elements. The validation of model of nuclide source term release, the validation of model of nuclide migration in geological media and computer code application were completed separately by CIRP and JAERI.

CONCLUSIONS AND SUGGESTIONS

Conclusions

(1) The results of nuclide tracer migration tests in loess-aerated zone have shown that :
Under natural rainfall condition, no significant movement of ^{90}Sr, ^{237}Np, ^{238}Pu and Sr, Nd, Ce has been found during the three-year test period.

Under sprinkling condition, any movement could be hardly found of ^{37}Np, ^{238}Pu and Ce, Nd and an about 15 cm downward migration of ^{90}Sr has been found during 1078-day test period. Less significant migration of ^{90}Sr and Sr in loessial medium with quartz sand as tracer carrier than with loess as tracer carrier has been found during the same period. As calculated, the downward migration of mass center reached about 2.7 cm. Loess in the field test site has very strong capacity of sorbing ^{237}Np, ^{238}Pu. The study indicates that the sorption of ^{237}Np on loess is dominated by calcium carbonate, followed by cation exchange capacity.

(2) In spite of a only 7 mm thick layer of quartz sand, a double-layer medium could be formed by quartz sand layer combined with loess, and had very strong shielding effect on unsaturated water flow, hence caused detouring water to be formed. An additional inter-comparison test for 470days with quartz sand and loess carrier, respectively, demonstrated once again the above conclusion. Sr migrated downward 10 cm in the test with loess as tracer carrier but only 2.7 cm in the test with quartz sand as tracer carrier during 470 days.

(3) In aquifer test, through a 976-day test period an about 8 cm migration of ^{90}Sr and a less than 2 cm migration of ^{237}Np as well as little migration of ^{238}Pu in experimental assemblies were found. For nuclide migration test in aquifer, the ^{90}Sr migration distances during the 3 years period along X, Y and Z directions reached about 2-3 cm separately, with little movement of ^{237}Np and ^{239}Pu.

(4) The Laboratory simulation experiments for loess-aerated zone and aquifer all present the results somewhat higher than those obtained in field nuclide migration tests. However, they are roughly consistent. In the laboratory simulation for loess-aerated zone, ^{90}Sr and ^{237}Np migrated 7.6 cm and 3 cm respectively, while ^{238}Pu, Nd and Ce has little migration during 1073-day test period. In the laboratory simulation experiments for aquifer, ^{90}Sr and ^{237}Np migrated 16 cm and 3.9 cm respectively during the 527 days period, while ^{238}Pu, Nd and Ce have little movement.

(5) The Engineering barrier materials, including cement, degraded cement, bentonite and mortar, have very strong adsorption capacity to ^{237}Np, ^{238}Pu, ^{90}Sr and Sr, Nd and Ce.
Under natural rainfall condition, no significant movement of ^{237}Np, ^{238}Pu, ^{90}Sr and Sr, Nd, Ce has been found in three kinds of sample block, and mortar-loess during the three-year test period. Some slight movement of nuclides in various sample blocks would be entirely attributable to molecular diffusion.
Under artificial rainfall condition, no movement of ^{238}Pu and slightly downward movement of ^{237}Np has been found in bentonite sample block during the three-year test period. Any significant downward movement of ^{237}Np, ^{238}Pu and Nd, Ce in cement sample block, degraded cement sample block and mortar loess could be hardly found during the three years period.

(6) The results in the laboratory experiment have shown that small significant effect of colloid on the migration velocity of nuclides in loess has been found. In batch test with increasing addition of humic acid in loess, the adsorption capacity of Np and Pu on loess decreased. While in assembly test the addition of humic acid make migration velocity of ^{90}Sr slower than in absence of humic acid.

(7) The test results have shown that Br^- migrates at somewhat faster velocity than ^3H.

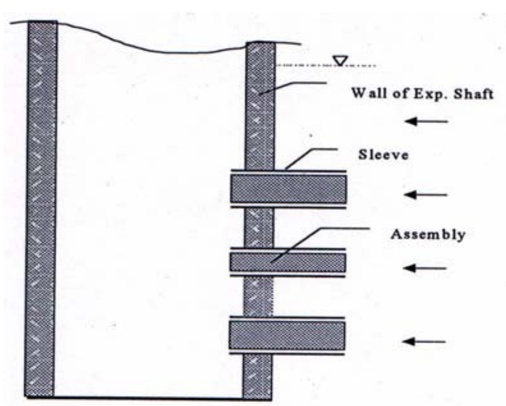
Through five years study a comprehensive and complete set of technologies and methodologies for safety assessment of near surface disposal of L(I)LWs have been developed, with large amounts of parameters, models and computer codes obtained. A wealth of experimental data have been obtained, especially the data of field nuclide migration, which in turn were used for verification and correction of the initial models and for validation of the developed non-equilibrium adsorption model.

Suggestions

(1) The continuing efforts should be devoted to the further study of the shielding effect of fine quartz sand on unsaturated water flow.

(2) It was found in the study of moisture transport that unmoving water flow has influence on nuclide migration to some extent. The effect on nuclide migration of immovable water in loess-aerated zone and aquifer should be further put under the study in the future.

Part 2



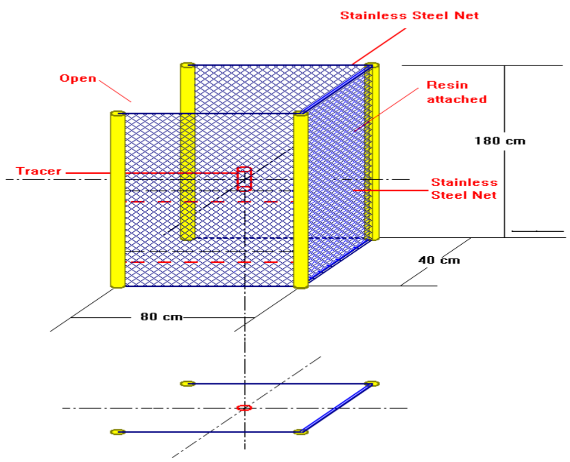


Fig. 2 Scheme of Experimental Shaft

Fig.3 Aquifer Test Frame

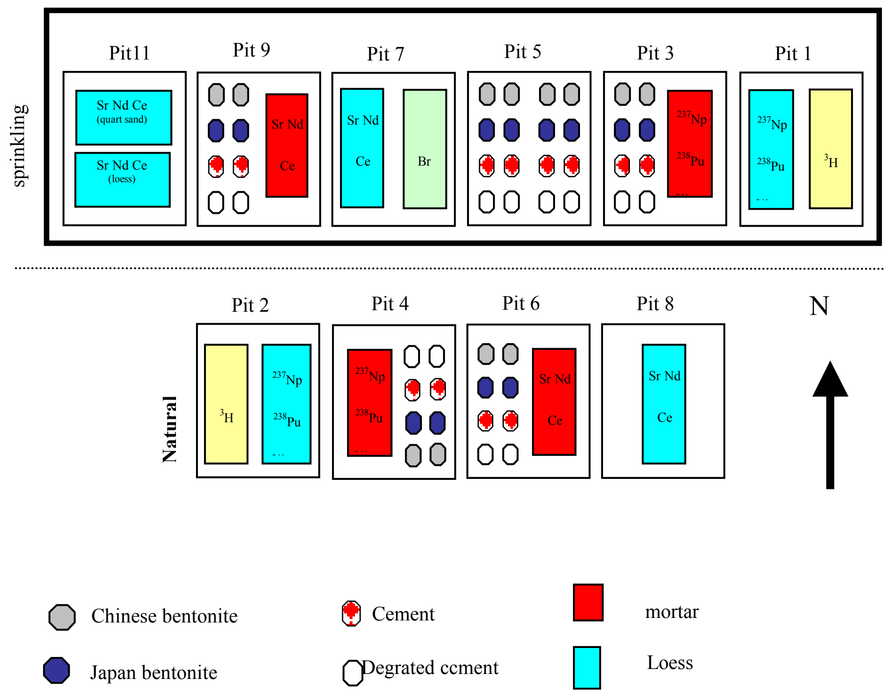


Fig. 4 Layout of the Arrangement of Test Pits in Aerated Zone