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Study of Test Methods for Radionuclide Migration in Aerated Zone

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Abstract Aerated zone is an important natural barrier against transport of radionuclides released from disposal facilities of LLRW. This paper introduces study methods for radionuclide migration in aerated zone, including determination of water movement, laboratory simulation test, and field tracing test. For one purpose, results obtained with different methods are compared. These methods have been used in a five-year cooperative research project between CIRP and JAERI for an establishment of methodology for safety assessment on shallow land disposal of LLRW.

Introduction

A cooperative research project has been conducted since 1988 between China Institute for Radiation Protection (CIRP) and Japan Atomic Energy Research Institute (JAERI) for an establishment of methodology for safety assessment on shallow land disposal of low level radioactive wastes[1]. The overall objective of safety assessment is the quantitative prediction of the performance of the whole disposal system over the time for which the wastes remain hazardous. For this, all factors affecting the performance of a disposal system and radionuclide transport should be considered. Emphasis of the cooperative project, however, has been put upon the reveal of properties of radionuclide migration in aerated zone, i.e. the unsaturated zone from ground surface to water table. CIRP's field test site was selected for implement of most contents of the project.

The main objectives of the project were:

1. To improve the field and laboratory methods for study of radionuclide migration in aerated zone, and compare the results obtained from field test and laboratory test.
2. To obtain the data used to guide safety assessment on disposal facilities which may be built on the places having the natural condition similar to the field test site.
3. To develop the models of radionuclide migration in unsaturated porous medium, and validate the models with field and simulation tests.

Following research contents have been conducted:

1. Investigation of site characteristics, including items such as regional

hydrogeological condition and geological structure, aquifer flow field, groundwater age, near-surface meteorological and pollution-meteorological conditions, radioactive background, health and diet of population, and socioeconomic situation.

2. Field study of water movement characteristics in loess aerated zone. More efforts were focused to observation of water movement, study of water infiltration , determination of unsaturated hydraulic conductivity with field and laboratory methods, measurement and calculation of evaporation and water budget balance.

3. Laboratory measurement of physicochemical properties of soil and groundwater, study of dynamic characteristics of soil-water, measurement of distribution coefficients of radionuclides. Estimation of leaching property of cement-solidified form under the simulated disposal environment.

4. Field tracing test of radionuclide migration in aerated zone.

5. Laboratory simulation test of radionuclide migration.

6. Research and development of models of safety assessment of shallow land disposal of LLRW.

Natural Conditions at Test Site

The test site is with an elevation of 954m. Investigation of regional geological and hydrogeological condition concludes that stratum in this region is mainly loess of the pleistocene series. Water table is 28m deep. The bedrock of about 170m in depth is water-bearing purplish-red sandstone of the riassic period.

According to the soil texture classification standardized by International Soil Science Society (ISSS), the designation of soil from ground surface to 9m depth is subsandy soil with granule-supported structure, so it has larger pore size and hydraulic conductivity. Soil in 9-18m and 21-28m is subclayey soil with granule-embedded structure, pore size becomes smaller and more cementing material exists among particles. Soil in 18-21m is defined to be red clay. Annual mean precipitation is 450mm. Annual mean temperature is 9.3°C, and the depth of the frozen earth is 0.81m.

Measurement and Calculation of Evaporation

The field test site was maintained bare surface and almost no runoff existed during the study, so that the infiltrating water was controlled by the precipitation and evaporation. The instruments used to measure data for calculation of evaporation rate included net pyrradiometer, heat flow meter, aspirated radiation shield wet and dry thermometer, rainfall gauge, ultrasonic anemometer-thermometer and others. All data were collected once per ten minutes and input to a personal computer. Four analysis methods (Energy balance method, Turbulence flux method, Iteration method and Penman's formula) were used for calculation of evaporation. The result shows that the annual evaporation at the test site is approximately equal to the precipitation, and ranges from 450 to 550mm per year[2].

Observation of Water Movement in Aerated Zone

Water movement is the major mechanism affecting migration and fate of radionuclides in geosphere. Observation of water movement under natural conditions was conducted for two years at the test site. A shaft with a depth of 9m was built, 93 tensiometers were inserted into the two side walls of the shaft for measuring soil suction potential at different time and depths. Moisture content and its change from surface to 27m depth were measured with a neutron moisture gauge. An one-dimensional seepage test was carried out on an area of 6mx6m on one side of the shaft. The instantaneous profile method of internal drainage redistribution was applied to determine unsaturated hydraulic conductivities. Fitting formulas were set up for soil at different depths[3,4,5].

Dynamic Characteristics of Soil-Water

Total of 46 samples were taken from surface to water table for measuring water retention curves in drying process. Several empirical equations were tested for fitting the measured data. Of them, Van Genuchten's equation produced the best fitting consistency. From the fitting parameters and dynamic theory of soil-water, we have calculated the distribution of equivalent pore diameters, the specific water capacity, $C(\theta)$, the hydraulic diffusivity, $D(\theta)$, and the unsaturated hydraulic conductivity, $K(\theta)$, for soil of each layers[6,7]. Unlike saturated conductivity, unsaturated conductivity is a function of water content (or matrix suction potential) in soil. For the given soil, change of water content (20%, e.g.) may induce unsaturated conductivity to change up to the order of magnitude of 2-4. The calculated unsaturated hydraulic conductivities were compared with the results measured directly by steady-state soil column method in laboratory, and with the results obtained from field observation of water movement mentioned above. Satisfactory consistency exists among these results for the given soil. Therefore, unsaturated conductivity can be predicted from easily obtained data of saturated conductivity and water retention curves, and be used to solve the equations of flow field and radionuclide migration in aerated zone.

Distribution Coefficients

Distribution coefficients of radionuclides, Kd, were measured in laboratory by batch method with groundwater and soil taken from the test site. Distribution coefficients of ^{60}Co, ^{85}Sr and ^{134}Cs are 5000, 80, 6000ml/g, respectively[8,9].

Laboratory Simulation Test

Laboratory simulation tests of radionuclide migration with two large soil columns were conducted[10,11]. The first column was made by filling soil taken from the test site with a size of a diameter of 280mm and length of 1200mm. Tracer ^{85}Sr was placed on the top of the column. Distilled water was sprinkled for 60 or 30 minutes per day. During 359 days, total sprinkling period and water volume were respectively 203.5 hours and 132.52

liters. A gamma detector with lead shield and collimating window was moved outside the column to determine directly activity. 18 times of direct measurements were carried out. After stopping sprinkling, soil samples were taken from the column and measured for distribution of ^{85}Sr. The second column was with the same size as the first one. It was an integral, undisturbed soil column taken from the test site. In the process of direct measurement of activity, the second column was rotating round its own axis while the detector was moving up and down outside the column. ^{60}Co, ^{85}Sr and ^{134}Cs were used as tracers.

Field Tracing Test

The field test of radionuclide migration was conducted in six pits with each size of 200cmx200cm at CIRP's field test site[12,13]. Pits A, B and C were used for test under natural condition, and pits D,E and F under artificial rainfall condition in a house. Pits A and D were 30cm deep, others were 100cm deep. ^{3}H, ^{60}Co, ^{85}Sr and ^{134}Cs were selected as tracers.

Mixture of soil with tracing radionuclides were distributed on the bottoms of pits A,B,D and E with each size of 150cmx150cm and thickness of about 7mm. Soil cores were taken from these four pits periodically and analyzed for distribution of radionuclides.

A PVC tube was vertically inserted at the center in pit C. The tube with a inside diameter of 240mm reached depth of 3m from ground surface. Mixture of soil with tracing radionuclides was distributed uniformly on the bottom with a diameter of 55mm and thickness of 6mm. Distance between boundaries of the tube and mixture was 100mm. A gamma detector with lead shield and collimating window was moved upward and downward to determine directly activities of radionuclides.

Pit F had the same design as pit C, used under artificial rainfall condition.
The artificial rainfall was controlled by 2 sets of sprinklers. For unsaturated condition in test pits, sprinkling intensity was set at 5mm/hr, and sprinkling continued 3 hours per day. Total amount of 47 soil cores were taken from pits A, B, D and E. 14 and 12 times of direct measurements were performed in pits C and F respectively. After finishing all direct measuring, pits C and F were open-cut to take soil samples for analysis in laboratory. By this way, the result of direct measurements can be checked.

Conclusion and Discussion

Results of the field tracing test, simulation test and measurement of distribution coefficients indicate that soil at the test site appears a strong ability to adsorb nuclides. After two years of field tracing test, ^{60}Co and ^{134}Cs were almost kept at the original position under both natural and artificial conditions because of their high value of Kd, and ^{85}Sr moved downward about 13 cm under artificial rainfall condition. Under the natural condition, considerable portion of ^{3}H and ^{85}Sr moved upward as the effect of evaporation.

Retardation factor, Rd, of ^{85}Sr is 375, calculated from Kd =80ml/g measured in laboratory. However, Rd of ^{85}Sr estimated with the fitting data of the field test is 132--265. This comparison suggests that Kd of ^{85}Sr measured in laboratory could be used to estimate movement of ^{85}Sr under field environment, but for conservative reason the measured value in laboratory should be reduced by a factor of 2-3 in prediction of migration of Sr in the specific field site. The Rd of ^{85}Sr estimated on the basis of laboratory simulation test is about 60-90.

Under natural condition, the travel time of ^{85}Sr through the aerated zone is estimated as more than ten thousands years, and much longer for ^{60}Co and ^{134}Cs. Result shows that a small portion of radionuclides move faster than their peaks, and two peaks of ^{85}Sr concentration appear under artificial condition. This phenomenon is important in safety assessment of radioactive wastes disposal. Reason for this is complicate. Preliminary study of the effect of environmental factors on distribution coefficients of ^{60}Co, ^{85}Sr and ^{134}Cs shows that pH and humic acid have strong effect on them. Maybe two chemical forms of ^{85}Sr exist during migration, therefore two distribution coefficients exist. Tiny portion of Co may form humic acid complex which is not strongly adsorbed on soil particles. To confirm this, adsorption mechanism of these nuclides is being studied.

The direct monitoring method has, in general, such limits that it can be applied only to high and medium energy gamma emitting nuclides with enough activities. However, as the nuclide migration can be monitored continuously with this method, nuclide migration due to rapid water penetration induced by such as a heavy rain, may be observed. Experiences obtained in this test will contribute to development of more realistic test technology at such deeper soil layer as 5-10m.

Great attention was paid to evaluate water budget balance, and to explore water movement characteristics in aerated zone. Two-year observation of the evaporation rate, and the distribution and change of water content provided data for modeling flow field in aerated zone. The four analysis methods of evaporation can generally produce perfect results in either moist or arid area. It should be noted, however, that a larger relative error may be exist in calculation of infiltration rate by precipitation rate minus evaporation rate in a arid area if evaporation and precipitation are very approximate to each other. In order to overcome this problem, direct observation of water content is recommenced with equipments described above in this paper or others such as lysimeters.

The first important hydraulic parameter in describing unsaturated flow field is unsaturated hydraulic conductivity. To reach it three methods have been adopted: the field one-dimensional seepage test, the measurement by steady-state soil column method in laboratory, and prediction from water retention curves measured in laboratory. Satisfactory consistency exists among the results from the three methods. This conclusion means that all these methods are effective, and the last method has a significant advantage over other two methods of lower cost and suitability for wider range of water content.

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