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ARTICLE

The migration experiment of Sr-90 in variable saturated soils and numerical simulation used Hydrus-3d

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There is a scientific support that migration data of Sr-90 in the nuclear waste landfill soil will provide for safety estimation and accident response measures in landfill.In order to make a more accurate prediction of the migration of Sr-90, it is critical to get the migration parameters such as longitudinal (or transverse) dispersivity and partition coefficient. This article try to use a method for estimating the migration parameters of Sr-90 in variable saturated soils combining experiment and numerical simulation, then explores the method with the migration of Sr-90 in silty loam in Shanxi province as an example. The migration experiment of Sr-90 was carried by column method , the water flow in is 375 mL/d, the pH value was about 7 at room temperature. The results show that the migration distance of central flow direction for Sr-90 is 3.9 cm and the peak concentration is 1.04×10^4 Bq/cm^3. According to the concentration distribution of Sr-90 in the soil column combined with three-dimensional numerical model of nuclide migration (Hydrus-3d), the partition coefficient in the silty loam is 79 ml/g while the longitudinal dispersivity is 0.7 cm and the transverse dispersivity is 0.8 cm by fitting. This method takes the porosity and water velocity of solid into consideration that static experiments fail to cover and obtains the longitudinal and transverse dispersivity at the same time, which making the results more tally with the actual situation.

Keywords: ***Sr-90; saturated - unsaturated soils; solute migration; dispersivity; partition coefficient***

1. Introduction

As a special kind of solute, the migration of Sr-90 in soil has not only the typical characteristics of other normal solutes, including convection, dispersion, adsorption, decay, biodegradation etc., but also has particularity--- the delayed effect caused by adsorption function receives more consideration. The accurate description of the migration of Sr-90 in soil depends on two key parameters: the hydrodynamic dispersion and the partition coefficient that reflects adsorptive characters. At present, the calculation of partition coefficient mainly uses the static adsorptive experiment by which the Sr-90 is put into a solution whose solid-to-liquid ratio is known and the ratio of these two is the partition coefficient after the concentration of solid and liquid levels out. However, the static method ignores the influence of porosity and flow velocity in the solid and liquild. The hydrodynamic dispersion is usually calculated by soil column method, but the longitudinal dispersivity can only be gotten based on the penetration curve of tracer and the longitudinal and transverse dispersion can barely gotten by using the three-dimensional model [1-7]. In order to get more reasonable partition coefficient of Sr-90 and calculate the hydrodynamic dispersion at the same time, this experiment employs the Sr-90 as the tracer. First, to establish three-dimensional soil column physical experiment and then the partition coefficient and hydrodynamic dispersion of Sr-90 are gotten by building the three-dimensional saturated-unstaurated zone numerical migration model with Hydrus-3d software based on the longitudinal and transverse concentration distribution of Sr-90 in the soil column.

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2. The materials and methods of the migration experiment

2.1. Materials

The soil sample is from Yuci, Shanxi province and the texture is silty. The percentage of sand , silt and clay are 25.5%, 64%, 10.5% respectively with bulk density 1.35 cm^3/g. Among the mineral composition, the content of quartz is 30~35%, mica 10%, plagioclase 15~20%, potassium feldspar 8%, chlorite 8%, hornblende 3%, calcite 15~20% and kaolinite 3%.

Tracer: Sr-90 in $SrCl_2$, 1.535×10^6 Bq, 1 ml solution mixed with 160 g quartz sand

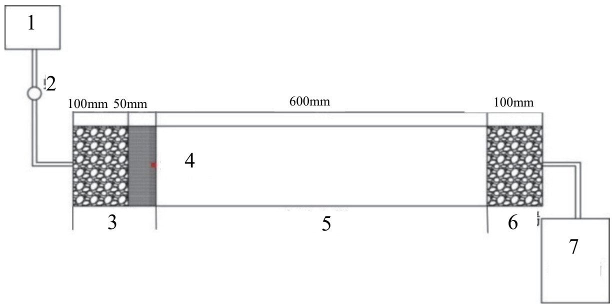
Measuring instrument: HPGe γ Spectrometer (OTREC company), the range of energy response 40 KeV-10 MeV, energy resolution less than 1.8 KeV

(1.332 MeV).

Sr-90 was measured with bremsstrahlung spectra.

2.2. Experimental facility

The experimental device consists of 3 parts: ①the water supply facility, including water box and peristaltic pump used to control the flow; ②the soil column facility; ③the collection of effluent facility, as shown in **Figure 1**.



1. Water box ; 2. Peristaltic pump; 3. Water entry; 4 Tracer; 5. Soil sample 6. water exit; 7.waste liquid collection barrel

Figure 1. Soil column experiment device.

The material of soil column is PMMA(polymethyl methacrylate) and the thickness is 1.0 cm. The size of experiment box is 850 mm×150 mm×150 mm. The experiment box has 4 parts: water entry, tracer, soil sample and water exit. The length of water entry is 150mm and filled with 100mm coarse sand and 50mm quartz sand (80 mesh) in turn.The tracer(Sr-90 in quartz sand) had been put in the soil which in the junction of water entry. The location seen in Figure 1 , No. 4 . The length of experiment soil sample taken from silt soil in Yuci is 600 mm. The length of water exit filled with coarse sand is 100mm. The water entry is connected to peristaltic pump and the flow is 375 mL/d (according to the real flow velocity). Distilled water is used in this experiment. The water exit is connected to waste liquid collection barrel and the volume of flow liquid is measured daily. The pH value is consistent with the pH value of the experimental soil environment, which is about 7 . The experiment had been carried out at room temperature.

2.3. Disintegration and Measurement

The experiment lasts for 260 days and water supply is ensured during this period. In order to get the longitudinal dispersivity, the tracer point (the junction of water entry and experimental soil sample) is set as zero and the direction of water flow as positive. The soil column is put into slices with each 3 mm in thickness and 7.8 cm long. As to the transverse dispersivity, The cutting ring put every slice into 4 soil sample circles with radius 1cm, 3 cm, 5 cm and 7 cm separately. There are 7 regions with two-layer (except the 1cm radius one) (**Figure 2**) and 182 soil samples. The samples are put into the polyethylene box and then dried, weighed, and finally the concentration of Sr-90 is measured.

2.4. The measurement of soil hydraulic parameters

Soil hydraulic parameters include soil water characteristic curve and saturated hydraulic conductivity. The measure of soil water characteristic curve uses the sand funnel method and mechanical strain method. The sand funnel method is used when the absorption force of soil is less while the mechanical strain method is used when the soil has great absorption. The absorption value with different volume water content is measured. The saturated water content θ_s is 0.445 cm^3/cm^3 and the residual water content θ_r is 0.046 cm^3/cm^3 by using the Van Genuchten(1980)model. The reciprocal value of air admission α is 0.00464 and n is 3.20. In addition, the saturated hydraulic conductivity of silt soil is 2.26×10^{-5} cm/s by constant head method.

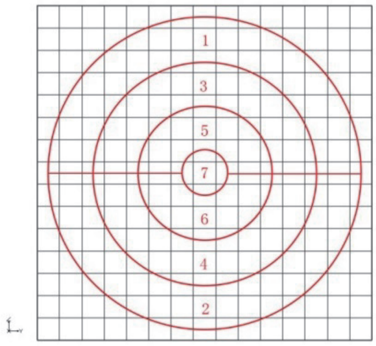


Figure 2. Sampling schematic.

3. Results of the migration experiment

The longitudinal concentration distribution curves of Sr-90 in each region are obtained from this experiment. The peak concentration is shown in **Table 1**. The concentration distribution curves of Sr-90 in each region show the unimodality in normal distribution and the concentration peaks of the curves only move down by 2.7~3.9 cm, which means silty loam has strong absorption to Sr-90. Its migration principle can be described through equilibrium adsorption theory [8-11], i.e. Sr-90 in the silty loam can easily get to adsorption and desorption equilibrium and the concentration of Sr-90 in the solid is the product of partition coefficient and liquid concentration at this moment.

4. Parameter solution

4.1. Method of parameter solution

In order to solve the migration parameters, including hydrodynamic dispersivity and partition coefficient, the three-dimensional numerical simulation method must be used. The method is described as follows: the measured concentration distribution curve of Sr-90 is obtained in each region according to the measured concentration distribution along the longitudinal direction in these 7 regions and the concentration value are regarded as validation data.

The three-dimensional variable saturated soil solute transport numerical model based on equilibrium absorption mode is applied in terms of the results of the experiment lasting for 260 days. The concentration distribution curve of Sr-90 is obtained by plotting along the longitudinal direction in 7 regions. The analysis is made by comparing the measured distribution curve and calculated distribution curve. According to the level of fitting, the partition coefficient K_d, the longitudinal dispersivity D_L and transverse dispersivity D_H in the silt loam are worked out.

Table 1. Peak concentration distribution of ^{90}Sr.

Regions	Peak migration distance (cm)	Peak concentration (Bq·cm^{-3})
1	2.7	1.12×10^3
2	3.3	1.86×10^3
3	3.6	2.00×10^3
4	3.6	4.04×10^3
5	3.6	8.03×10^3
6	3.6	9.80×10^3
7	3.9	1.04×10^4

4.2. Verfication of flow

Before establishing the numerical migration model of saturated-unsaturated flow, it is necessary to construct the flow model and get the measured data used for verification of flow. The tracer is set as initial point and the piezometer tubes are laid along flow direction every 15cm which are used to measure the distribution of water head along axial direction (**Figure 3**). Two piezometer tubes are laid around 2# up and down (5# and 6#) and the interval is 6 cm which can measure the distribution of water head along radial direction.

4.3. Numerical simulation model

The variable saturated soils three-dimensional flow model is established by Hydrus-3d after acquiring the distribution of water head in the soil column. The soil column is put into 3 mm rectangle cell along the flow direction in the model space and 10 mm rectangle cell in the y and z direction. The size of the cell is 3 mm×10 mm×10 mm and subdivided into 15 layers,15lines and 283 rows. Thus there are 72704 nodes and 127350 grids. The accuracy of established flow model is verified according to the measured head water value (**Table 2**). **Figure 4** shows the distribution of water head calculated in the experiment box.

4.4. Results of numerical simulation

The measured water head value and calculated value are basically consistent by comparison and the established model reveals the real condition of the flow in the box. The value of hydraulic gradient J is 1.10 which is calculated with the difference of the two water head and Δx. The osmotic coefficient of the soil column is 1.76×10^{-5} cm/s by using the Darcy Formula and the result is basically coincident with measured value of water head.

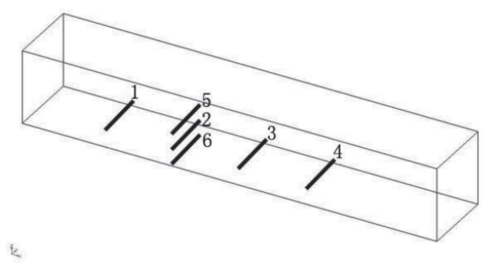


Figure 3. Piezometer tubes distribution.

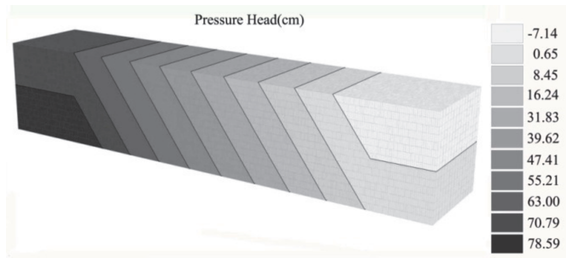


Figure 4. Calculation of water head distribution.

Table 2. Results of measured values and calculated values.

Piezometer tubes position	NO.	1	2	3	4	5	6
Coordinate	x (cm)	0	15	30	45	15	15
	z (cm)	8	8	8	8	2	14
Measured water head/(cm)		61.23	47.85	44.40	13.67	52.00	44.12
Calculated water head /(cm)		70.56	53.06	35.21	17.36	59.06	47.06

4.5. parameter solution

It is considered that the concentration of Sr-90 in solid is the product of partition coefficient and liquid concentration on the basis of equilibrium adsorption theory after the establish and verification of the flow model. The three-dimensional model of saturated-unsaturated flow migration is set up by Richards formula based on equilibrium adsorption mode according to the concentration distribution of Sr-90 in the axial and radial direction. The mathematical expression as Eq. (1).

$$\begin{cases} \theta\dfrac{\partial C}{\partial t}=\dfrac{\partial}{x_i}\left(\theta D_{ij}\dfrac{\partial C}{\partial x_j}\right)-\dfrac{\partial}{\partial x_i}(q_i C)-\rho_b\dfrac{\partial S^k}{\partial C} \\ \dfrac{\partial S^k}{\partial t}=K_d\dfrac{\partial c}{\partial t} \end{cases} \quad (1)$$

In this expression, θ is volume moisture content, cm^3/cm^3 ; t is the migration time of solute, d; q_i is Darcy flow rate, cm/d; D_{ij} is dispersion coefficient, cm^2/d; ρ is volume dry density, g/cm^3; C is concentration of dissolved phase, Bq/cm^3; S^k is concentration of adsorbed phase, Bq/cm^3; K_d is partition coefficient under the equilibrium adsorption, cm^3/g.

The concentration of Sr-90 in the whole box is obtained through the model calculation. The model in the horizontal direction is divided into 15 layers from bottom up and the concentration of Sr-90 in 7 layers (2,4,6,8,10,12,14) are selected (see **Figure 5**). The calculated results are consistent with the measured ones. The maximum value is the 8th layer(the center one) and the value decreases away from center along up and down. While the concentration in bottom half is greater than that of upper half at the symmetrical part. Sr-90 has

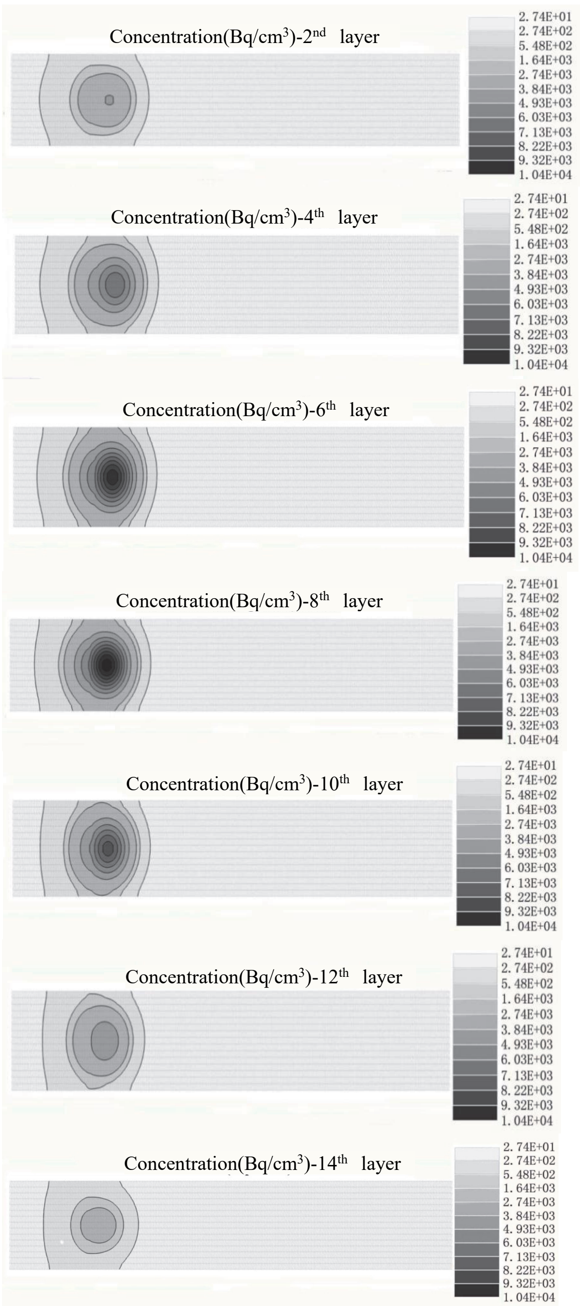


Figure 5. Calculated concentration of hydrus-3d.

already migrated from center to top and bottom during the 260 days, which reveals the transverse dispersivity is the key of the radial migration of Sr-90.

The curve of calculated concentration distribution is obtained by plotting the concentration of Sr-90 in each region (1-7) along longitudinal direction. A comparison is made between the measured distribution curve and the calculated curve by numerical model.

The parameter fitting results are shown in **Figure 6**. It shows the good agreement of numerical and the experimental results. The partition coefficient of Sr-90 in the silt loam at Yuci, Shanxi is 79 ml/g, the results take the porosity of solid and flow velocity into fully consideration with the comparison of the static adsorption experiment, thus tallying with the actual condition. The longitudinal dispersivity D_L is 0.7 cm and the transverse dispercity is 0.8cm. These two values are consistent which indicates they are isotropic.

The silt loam of this experiment is from field testing ground in Yuci of CIRP. In the 1990s, Jinsheng Wang [12] et al. made the in-situ experiment of Sr-85 migration in the silt loam at this field and acquired the partition coefficient 71.18 ml/g, longitudinal dispersivity D_L is 0.915 cm and the transverse dispercity is 0.55 cm. Sr-85 and Sr-90 are isotopes and they have same chemical property. The calculated results are consistent with the data of the field experiment, so this method can be used to obtain the key migration parameters of Sr-90 in soil.

5. Conclusion

This paper analyzed the establishment of the saturated-unsaturated flow physical experiment model and the migration principle of Sr-90 in the silt loam through the three-dimensional soil column experiment combined with numerical simulation method as well. The results indicate that the migration principle accords with the equilibrium adsorption and the key migration parameters (partition coefficient, hydrodynamic dispersivity) were obtained according to the concentration migration curve. This method breaks through the limitations of the traditional static adsorption experiment which ignores the porosity of solid and the water velocity, thus make the results more tally with actual condition. In addition, the longitudinal dispersivity and transverse dispersivity obtained at the same time fill the gap of only getting the longitudinal dispersivity before, which offers a solving method to accurately predict the migration of Sr-90 in soil at nuclear waste landfill.

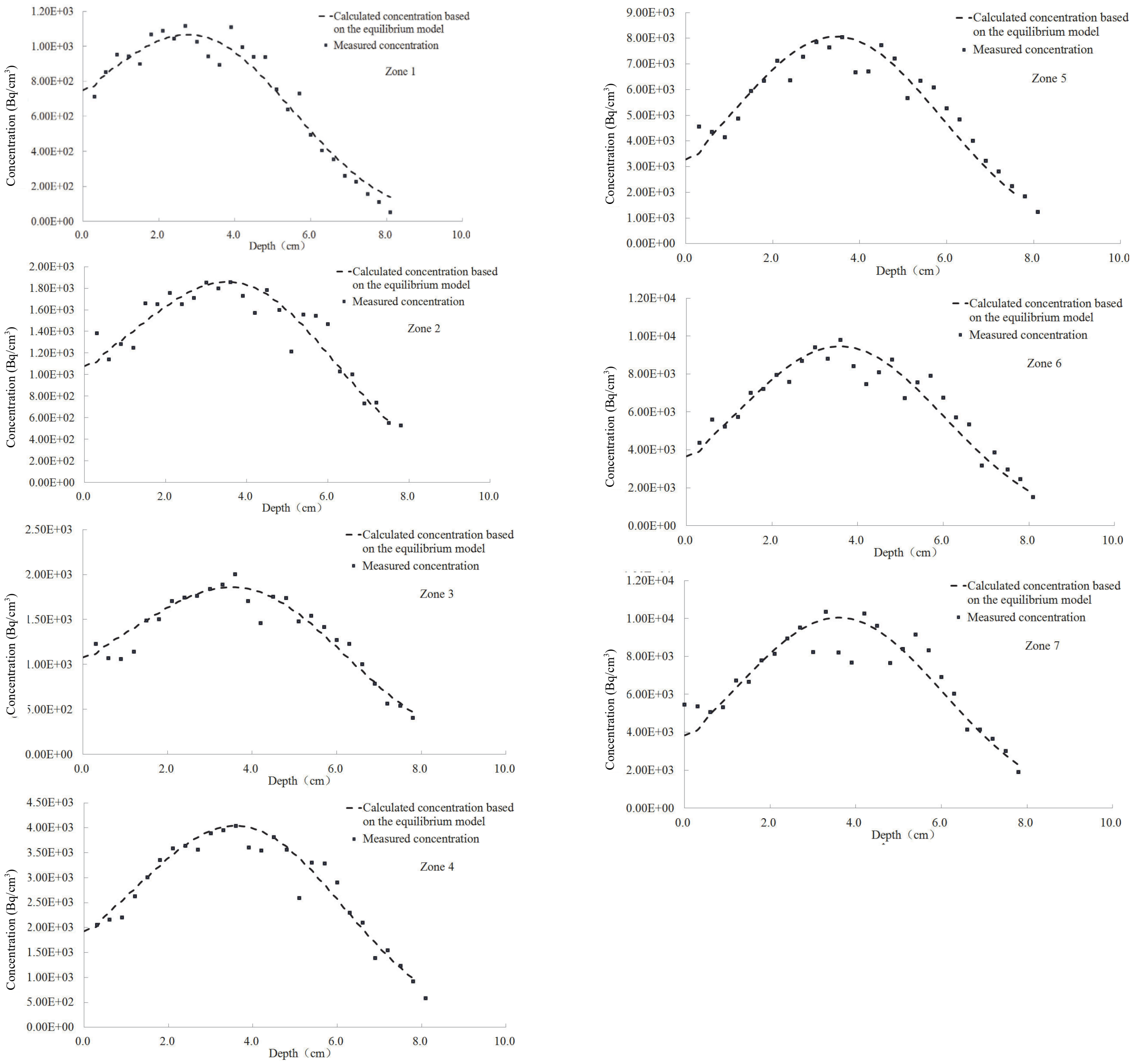


Figure 6. Fitting of measured and calculated value.

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