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EXPERIMENTAL ANALYSIS OF SALT DIFFUSION IN COMPACTED CLAYS BY THROUGH DIFFUSION AND HALF-CELL TECHNIQUE

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Abstract: The estimation of the model parameters namely effective diffusion coefficient and retardation factor of a potential landfill liner material was presented in this paper using the experimentally measured salt concentration data. Experimental data of concentration variation of time and spatial distance in compacted bentonite were obtained using two diffusion measurement techniques viz., through diffusion and half-cell technique respectively. The bentonite was subjected to the same concentration gradient and compacted density in both the experimental methods to compare the results and understand underlying mechanism in the diffusion tests. The measured data from the laboratory diffusion techniques were analysed using a GUI (Graphical User Interface) based Dot-net application CONTRADIS. The CONTRADIS was used to estimate the model parameters by the inverse analysis. The application uses the solution of the forward analysis and stochastic algorithm for the inverse analysis. The retardation factor obtained theoretically was validated using laboratory batch sorption tests.

Keywords: Through-diffusion, Half-cell, Clay, CONTRADIS

1 INTRODUCTION

The barrier materials used in the landfill liners are mostly clayey soil having very high plasticity and hydraulic conductivity as low as 10^{-9} cm/sec. In such a case the solute transport will be mostly governed by diffusion and the flow due to advection will be negligible [1, 2, 3]. In most of the municipal solid waste landfills or highly toxic and hazardous waste landfills, flow due to diffusion is considered to be the significant transport mechanism [4]. As such for the effective design of landfill liners, knowledge of the parameter governing the diffusive mechanism which is described by diffusion coefficient, becomes essential. Also flow through the plastic compacted clays is influenced by the sorption characteristics of the soils. The sorption potential is best described by a parameter known as retardation factor [5]. Hence proper estimation of both the model parameters which are the effective diffusion coefficient and retardation factors becomes important. Diffusion coefficients are obtained experimentally from laboratory diffusion tests and the retardation factors are obtained from the equilibrium batch tests [3, 5]. However estimation and comparison of the model parameters by different available methods are scarcely conducted.

In this paper two different laboratory techniques namely through-diffusion and half-cell technique were used to estimate the model parameters. The tests were performed on duplicate specimens at a particular density and under same concentration gradient to understand the migration rate of a particular type of ion, to have a comparative analysis of the model parameters, in which not much literature is available.

The paper also describes the development of a dot-net application based software package CONTRADIS that was built for the purpose of analyzing experimental results obtained from through-diffusion and half-cell technique. This software is capable of using the contaminant transport data from experiments and predicting the diffusion and linear sorption parameters of the soils.

2 GOVERNING MECHANISM

The governing equation describing the transport of solute through saturated soil is formulated utilizing Fick's second law which is given as [5, 6, 7]:

$$\frac{\partial c}{\partial t} = \frac{D^*}{R_d}\frac{\partial^2 c}{\partial x^2}$$

where R_d is the retardation factor and D^* is the effective diffusion coefficient.

3 MATERIALS AND METHODS

Bentonite soil rich in montmorillonite mineral having liquid limit of 393% was used in the present study. The specific surface area snd the cation exchange capacity of the soil are 495 m^2/g and 68 meq/100g respectively.

The diffusion cell, made of Poly methyl methacrylate glass tubes having diameter of 2.5 cm and thickness of 1 cm was used for the diffusion testing.

3.1 Through- Diffusion Technique:

Bentonite soil compacted a particular density of 1.5 g/cc was palced in the diffusion cell and saturated for a period of 21 days. The clay plug was placed in contact with two reservoirs namely source and collector reservoir as depicted in Figure 1. At the end of the saturation period diffusion testing was started by placing NaCl solution of 0.2 M in the source reservoir and and pure distilled water in collector reservoir. Due to this concentration gradient one dimensional diffusive mass flux would occur from source to collector reservoir. Both the reservoir concentrations were analysed with time to obtain the plot of relative concentration *(c/c_0)* of sodium ions in both the reservoirs with time. c_0 is the initial concentration which is 0.2 M or 12000 ppm.

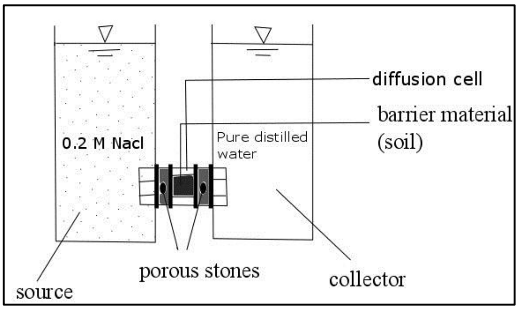

Fig 1 Through diffusion set-up

3.2 Half-Cell Technique

Two half cells of diameter 3.9 cm and 4 cm length were used in this technique. In both the half cells soil is compacted at 1.5 g/cc where one half cell was saturated with 0.2 M NaCl and the other half cell is saturated with pure distilled water. After saturation both the cells were brought in contact with each other and diffusion process took place from the tagged half-cell with NaCl to the untagged hal-cell (Figure 2). At the end of the diffusion testing period both the half cells were dismantled and the soil in the cells were sliced at 5 mm thicknes and analysed for concentration variation along the length of the half cells.

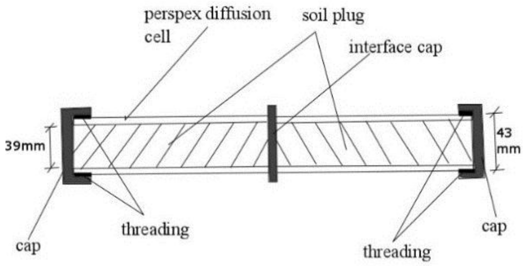

Fig 2 Half-cell set-up

Based on various initial and boundary conditions analytical solutions are developed for both the methods [7, 8].

3.3 Batch Test

Batch study was carried out in the present study to determine one of the model parameter experimentally which is the retardation factor and compare with the theoretical result. The soil to solution ratio maintained in the test is 1:20. Batch equilibrium test was performed on air-dried soil to determine the sorption characteristics of the clay soil with respect to sodium ions. 10 grams of the soil is mixed with 200 ml of NaCl of different initial concentrations. Six different initial concentrations are maintained in six different soil samples having weight of 10 grams. The six different initial concentrations that were analysed are 2, 4, 6, 8, 10 and 12 g/l. The soil solution is mixed thoroughly in an orbital shaker for 48 hours as equilibrium time is assumed to be 48 hours. After thorough mixing the slurries are taken in a centrifuge tube and centrifuged for 2000 rpm for 10 minutes. The supernatant from each tube is analysed for the equilibrium concentration of the sodium ion by flame photometer. The results were plotted to determine the adsorption isotherm. The sorbed concentration of sodium ions in the soil was plotted against the equilibrium concentration of the ion in the solution to get the sorption isotherm which is given as:

$$q = \frac{(C_0 - C_e) \times V}{M_s} \qquad (1)$$

where V is the volume of the solution and M_s is the mass of the soil sample.

4 CONTRADIS

The name CONTRADIS stands for "CONtaminant TRAnsport due to DIffusion in Soils". CONTRADIS is a software package that was built for the purpose of analyzing the experimental readings obtained from various methods that have been used by the previous studies in the field of contaminant transport in soils.

This application based software package was developed to overcome the shortcomings of the existing software POLLUTE which is a commercially available software and is based on semi-analytical solutions. However the present application CONTRADIS can perform inverse analysis and estimate the model parameters using stochastic optimization algorithm [6].

The CONTRADIS utilizes the analytical solutions [7, 8] of both the above described experimental methods and can generate theoretical concentration profile by performing inverse analysis. This software package was verified on the synthetic data obtained from the calculated concentration value as an input data. With the input parameter of D^* and R_d a theoretical concentration profile is obtained and the actual estimate of the model parameters are taken to be the one for which the experimental profile fits well the theoretical profile with minimum RMSE.

5 RESULTS AND DISCUSSIONS

5.1 Results from through-diffusion techniqe

The concentration profile was obtained for a period of 60 days for both the reservoirs. It was observed that the concentration in the source reservoir decreased with time and concentration in the collector reservoir increased with time. The experimental data was used in CONTRADIS for the inverse analysis which compares it with the theoretical obtained data by minimizing the RMSE error. The fitting of the theoretical and the experimental concentration profiles as obtained from the software by inverse analysis is given in Fig. 3. The effective diffusion coefficient so obtained is $7.7 \times 10^{-6} cm^2/sec$ and retardation factor is 10.

5.2 Results from Half-Cell Technique

After dismantling the half cells at the end of the diffusion testing period the soil plugs were sliced and analysded for the sodium ion concentration after mixing the soil with 1 M ammonium acetate and mixing it for a period of 16 hours. Ammonium acetate replaces all the adsorbed and the free exchangeable ions present in the soil layer. The mixture was then centrifuged and the supernatant so obtained was analysed for total sodium ion concentration for all the slices. The mobile ion or the free ion concentration was obtained by substracting the total ion concentration from the adsorbed concentration for the same dried mass in the particular slice. A total of 16 soil slices were analysed. The concentration profile along the length of the two half cell obtained from the experimental half cell technique shows that the profile reaches to the ends of the half cell which is finite porous medium case. Hence the theory of finite porous medium case is used in CONTRADIS and the experimental data was analysed in the sofrware package for getting the model parameters by inverse analysis. The theoretical data was fitted with the experimental data with minimum root mean square error. The best fit curve showing the theoretical concentration data and experimental data is shown in Figure 4. The effective diffusion coefficient so obtained is $1.51 \times 10^{-5} cm^2/sec$ and retardation factor is 1.5.

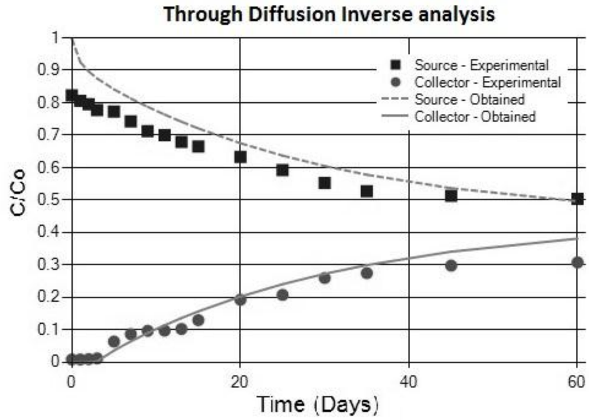

Fig 3 CONTRADIS conc. profile for through-diffusion test

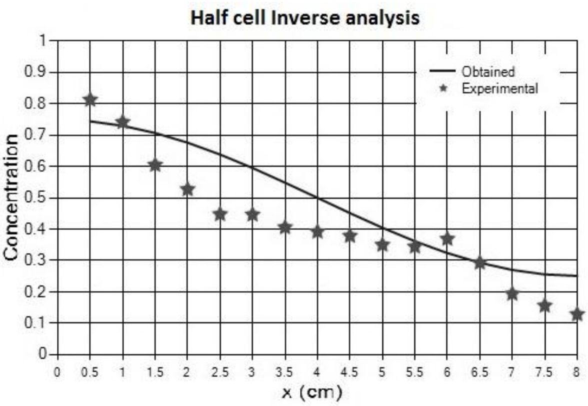

Fig 4 CONTRADIS conc. profile for half-cell test

5.3 Results from Batch Test

Batch test results were utilised to obtain both linear and non-linear sorption isotherms. Figure 5 and figure 6 shows that the linear sorption isotherm does not fit the experimental data as accurately as non linear Freudlich model does. This is beacuse of the fact that at higher concentration the sorption isotherms are highly non-linear. However for simpliciy of solving the governing differential equation 1, the sorption isotherm is considered linear. Beacuse of this assumption we can see that in Figure 3 and Figure 4 experiental data and the theoretical data could not be as well-fitted as is expected. The equations describing the process to determine the retardation factor from linear and Freudlich model can be found elsewhere [3, 5]. The retardation factor from the linear model obtained experimentally is 4.8 and the retardation factor obtained from the linearized Freudlich model is 6.1.

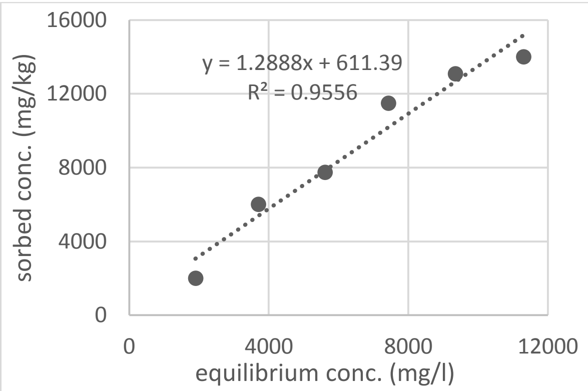

Fig 5 Linear isotherm model

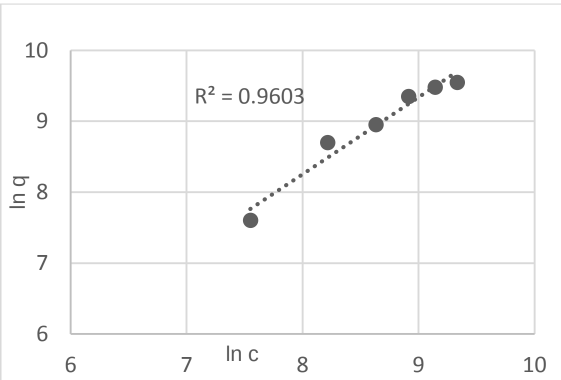

Fig 6 Freudlich isotherm model

6 SUMMARY AND CONCLUSIONS

Measured salt Concentration profiles with time and length of the soil plugs were obtained for two different laboratory techniques viz., through diffusion and half-cell technique respectively. A new software package named CONTRADIS was developed which utlizes the analytical solutions of the governing differntial eqaution, for both these techniques to determine the model parameters by inverse analysis using optimization algirithm. In order to validate one of the model parameter namely retardation factor, laboratory batch equilibrium test was conducted. The test results shows that the model parameters obtained by through diffusion technique were more relaible as the retardation factor obtained experimentally nearly matched the theoretically obtained value. The results of the half-cell technique might not be considered as realistic owing to experimental error like improper contact between the half cells. However more tests need to be conducted on various contaminating species using both these techniques to understand the relaibility of the model parameters which would be useful in the design of landfill liners.

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