A METHODOLOGY TO EVALUATE THE DIFFUSION COEFFICIENT OF RADIONUCLIDES THROUGH ROCK MASS IN A SHORT EXPERIMENTAL DURATION

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ABSTRACT

Geological isolation of nuclear waste requires laboratory and field investigations to evaluate migration parameters such as diffusion and dispersion coefficient of radionuclides in order to assess the site suitability for locating a high level radioactive waste repository. However, migration of radionuclides through the rock mass away from the radioactive waste repository is an extremely slow process and the laboratory experiments to evaluate the diffusion coeffficient (D_i) of radionuclides are time consuming.

The present study deals with the development of a methodology adopting a centrifuge modeling technique to evaluate the diffusion coefficient of radionuclides through the fractured and the intact rock mass over a very short experimental duration. This is achieved by simulating the diffusion of Iodide, Cesium and Strontium ions through sliced rock samples in a specially designed diffusion cells under a high gravitational field in a Geotechnical Centrifuge.

Results indicate that the diffusion time of these ions through the rock mass in the centrifuge is reduced by N^2 times compared with conventional laboratory diffusion tests, where the N is the applied 'g' (acceleration due to gravity). D_i of I, Cs and Sr through the fractured and intact rock samples is found to range from 0.60 to $8.3 \times 10^{-9} m^2/s$ and 1.2 to $4.9 \times 10^{-14} m^2/s$, respectively.

Evaluating migration parameters rapidly helps to simulate the in-situ conditions needed to study the long term effect of radionuclide migration in geo-environment.

INTRODUCTION

Isolation of nuclear waste in deep geological formations is considered worldwide as the suitable option to protect man and the environment for extended periods. In a deep geological repository migration of waste components away from the disposal site mainly depends on the physical condition of the rock mass, the type of the minerals present in it and their reactivity with the various radionuclides (sorbing or non-sorbing) released from the waste [1]. Mechanisms which control the contaminant migration through the rock mass are advection and/or diffusion [2]. To understand these mechanisms conventional laboratory experiments are conducted [3,4] and the results are used for the development and validation of theoretical models to predict the long-term behaviour of radionuclides. The interaction between various geometrical factors (such as

fundamental properties/characteristics of the porous medium, fractured porous medium) and several other physico-chemical processes (such as retardation, matrix diffusion, particle transport, dispersion) result in a distribution of radionuclide between the solid and the liquid phase in the geo-environment. Other factors are related to the nature of the aqueous phase and the dissolved radionuclides [5]. This necessitates a thorough understanding of the waste-geo-environment interaction for selection of a suitable disposal site.

The laboratory experiments are quite time consuming and cost intensive. At the same time, these experiments suffer from the limitations associated with the incorporation of material complexity, difficulty associated with reproduction of the boundary conditions, which control the governing mechanism(s) and differences in the time scales for completion of the processes [6]. At the same time, theoretical models developed based on laboratory and field experiments lack validation due to mismatch of time scale and uncertainties associated with long-term predictions to demonstrate the safety of deep disposal options [7]. In this context, several researchers [8,9,10] have recently demonstrated the usefulness of Centrifuge modeling techniques to simulate the naturally slow processes of contaminant migration in soil mass in a short duration of time. These researchers have also derived the scaling factors for the modeling purpose. Villar and Merrifield [11,12] have simulated the fate and behavior of disposed radioactive waste, through sands, under the conditions prevailing in a deep geological repository. These studies help in development of an improved conceptual understanding of contaminant transport mechanisms. However, the centrifuge modeling technique has not been extended to study and simulate contaminant migration through the rock mass. Realizing the fact that the contaminant migration through the rock mass is an extremely slow process, an attempt has been made in this paper to demonstrate the feasibility of the centrifuge modeling technique to model the contaminant diffusion through intact and fractured rock mass in the absence of other migration mechanisms like advection, dispersion etc.

With this in view, a methodology has been developed to evaluate the diffusion coefficient of various waste components over a very short experimental duration. This has been achieved by simulating the diffusion of Cesium, Strontium and Iodide ions through intact and fractured rock masses in specially designed diffusion cells in a geotechnical centrifuge. Diffusion coefficients of these ions thus estimated are validated vis-à-vis those reported by the earlier researchers.

CENTRIFUGE MODELING

Centrifuge modeling is emerging as a useful technique to study and model various geo-environmental problems [6]. The technique is here extended to study diffusion of contaminants through the rock mass. In a centrifuge, the rock sample (model) experiences the same magnitude and distribution of self-weight stresses as those of its prototype. The main difference between the model and its prototype is that the linear dimensions of the prototype are scaled down by a factor of N, at centrifugal acceleration N times greater than g, the acceleration due to Earth's gravity. However, the model in the centrifuge has a free unstressed upper surface and within the sample the magnitude of stress increases with depth at a rate related to the sample density and strength of the acceleration field, N.g.

During operation of the geotechnical centrifuge, g along the length of the model is different due to the linear variation of the acceleration, $(\omega^2.r)$, where ω is the angular velocity and r is the (effective) radial distance of an element in the model, from the axis of rotation, and can be obtained by using the following equation [13]:

$$r = r_t + \frac{h_m}{3}$$

(Eq. 1)

where r_t is the radial distance to the top of the model and h_m is the length of the model. In the present study, a small geotechnical centrifuge, with the details presented in Table I, has been employed. However, to overcome various shortcomings associated with the small centrifuge and a small size model, a big centrifuge must be used. This would enable to some extent the incorporation of inherent material heterogeneity as well as anisotropy. At the same time, the transport mechanisms occurring in the model under controlled boundary and initial conditions may also be used to verify and improve the capabilities and efficiency of various mathematical and analytical models used for predicting the transport mechanisms involved in the geo-environment. However to predict the prototype behaviour correctly, from the observed model behaviour, similar conditions must be established for the model and the prototype [8,10].

Table I Centrifuge details

Type	Swinging buckets on both sides of the arm
Arm radius	200 mm
Max. outer radius	315 mm
RPM range	250-1000
Max. acceleration	300 g
Capacity	0.72 g tons
Spin-up time	20 s
Spin-down time	80 s

Scaling laws for contaminant transport through porous media.

Arulanandan et al. [8], Hensley and Schofield [9] have derived scaling laws that govern the relationship between the centrifuge model and its prototype for contaminant migration through soils. For the sake of completeness, the summary of the scaling relationships is presented in Table II.

Table II Scaling factors for centrifuge modeling

Parameter	Model scale
Length	$1/N$
Pore size	1
Porosity	1
Stress	1
Strain	1
Mass	$1/N^3$
Mass density	1
Time (diffusion)	$1/N^2$

Scaling of the diffusion process and time

For a problem where advection, diffusion, dispersion and adsorption occur, the physical properties that define the concentration, C, of a contaminant may be given by:

C= f (μ, D_i, s, v_s, σ, ρ, g, l, d, t, bulk soil properties)   (Eq. 2)

where C is the concentration of the contaminant, μ is the dynamic viscosity of the fluid; D_i is the coefficient of molecular diffusion, s is the mass of adsorbed contaminant per unit-volume, v_s is the interstitial flow velocity, σ is the surface tension for fluid/particle interface, ρ is the density of the fluid, g is the acceleration due to the gravity, l is the characteristic macroscopic length (the sample thickness), d is the characteristic microscopic length (particle size), and t is the time.

The Coefficient of diffusion for an ion in the porous media is a function of both the medium and the free diffusion coefficient of the ion in the solution. For the same radionuclide and an identical rock mass, subjected to similar stress in the model and its prototype, the condition $(D_i)_m = (D_i)_p$, where subscripts $_m$ and $_p$ indicate model and prototype, respectively, should result. To maintain the ratio $(D_i.t/l^2)$ invariant, the condition $t_p=N^2.t_m$ must be met.

Scaling of linear dimensions

If the sample used for the model and its prototype is identical and the model is subjected to higher acceleration, in a spinning centrifuge, the vertical stress at a depth h_m, in the model, will be identical to that in the corresponding prototype at depth h_p.

As such, for the rock sample of density, ρ, the vertical stress, $σ_m$, at a depth, h_m, in the model can be represented as:

$σ_m = ρ.N.g.h_m$ (Eq. 3)

Similarly, for the same sample, the vertical stress $σ_p$, at a depth, h_p, in the prototype would be:

$σ_p = ρ.g.h_p$ (Eq. 4)

As such, for $σ_m$ to be same as $σ_p$:

$h_m = h_p.N^{-1}$ (Eq. 5)

As per Eq. 5 the scale factor (model to prototype) for the linear dimensions is $1/N$. The equation states that stress similarity is achieved at homologous points in the model and its prototype by accelerating a model of scale $1/N$ to N times Earth's gravity. Since the model is a linear scale representation of the prototype, the same scale factor may be imposed to displacements also. It therefore follows that strain scales to a factor of 1. As such, the sample stress-strain properties of the model are identical to that of the prototype.

EXPERIMENTAL INVESTIGATIONS

Rock cores used in the present study have been collected from the deep boreholes drilled in the Charnockite rock formation of Kalpakkam, India. The mineralogical and chemical composition, and physical and mechanical properties of the rock samples are presented in Table III [14] and Table IV [15], respectively.

Table III Mineralogical and chemical composition of the rock samples

Mineral	Modal percent	Oxide	% by weight
Quartz	35.8	SiO_2	63.06
K-feldspar	20.8	Al_2O_3	6.59
Plagioclase	16.4	Fe_2O_3	0.79
Biotite	7.5	FeO	5.55
Apatite	2.2	MnO	0.05
Opaques	4.1	CaO	3.16
Hypersthene	---	MgO	2.96
Garnet	10.0	Na_2O	3.66
Pyroxene	3.2	K_2O	2.51
		TiO_2	0.75
		P_2O_5	0.46
		LOI	0.46

Table IV Physical and mechanical properties of the rock samples

Parameter	Value
Total porosity (%)	0.32
Water absorption (%)	0.22
Young's modulus (GPa)	95
Poisson's ratio	0.22
Uni-axial Compressive Strength (MPa)	161
Bulk density (g/cc)	2.58

These cores are sliced into the required thickness for conducting prototype (1-g tests) and centrifuge (*N*-g tests) diffusion tests. The slicing is done using high-speed rock cutting machine. These slices are polished, with the help of rock polishing machine, and their surface is cleaned using an ultrasonic surface cleaner. These rock slices were soaked in water for about four months before conducting experiments. The thicknesses of the samples chosen for the diffusion through intact and fractured rock slices are 3mm and 60mm respectively. Using a rock core splitter, a fracture along the rock core axis has been created in the 60mm thick samples and the fracture aperture is determined using a microscope after fixing it in the experimental set up. To ensure no leakage of the source solution in the measuring compartment, the sides of the rock sample have been sealed using silicone adhesive. The fracture aperture obtained for the samples is in the

range of 72 to 226 microns and hence the average aperture can be assumed to be equal to 148. For the sake of generality, three identical samples have been tested. As such, 1-g tests are conducted for the sake of comparing the diffusion coefficient of ions obtained under normal laboratory conditions with that obtained from centrifuge tests. However, the diffusion time and the sample length are not comparable between 1-g tests of the present study and N-g tests due to scaling of diffusion time and sample length during centrifugation.

Type-I Diffusion cell

The diffusion cell fabricated for testing the intact rock samples, at 1-g and *N*-g, is depicted in Fig. 1. The cell is made of a Perspex (transparent acrylic material) cylinder of size 110mm in length with its inner and outer diameters equal to 54mm and 60mm, respectively. The cylinder is divided into three compartments by two intact rock samples of size 54.5mm in diameter and thickness, L, 3mm. The gap between the Perspex cylinder and the rock samples are sealed with silicone adhesive to avoid any leakage between the compartments. The rock samples are positioned in such a way that a large volume (approximately 200cc) is available, between the two rock samples, for filling the source solution with a certain concentration of the contaminant (C_0). The two end compartments are filled with the ultrapure water with conductivity <0.5μS/cm (approximately 50cc). Each of these compartments is provided with a 6mm hole at the top with rubber seal for sampling purposes and measuring the diffused ion concentration. The two ends of the diffusion cell are sealed with the Perspex plates of size 100mm×100mm×10mm. The advantage of this type of partitioning is that two rock samples can be tested at a time. Also, sampling to measure the change in concentration of the solution is simplified.
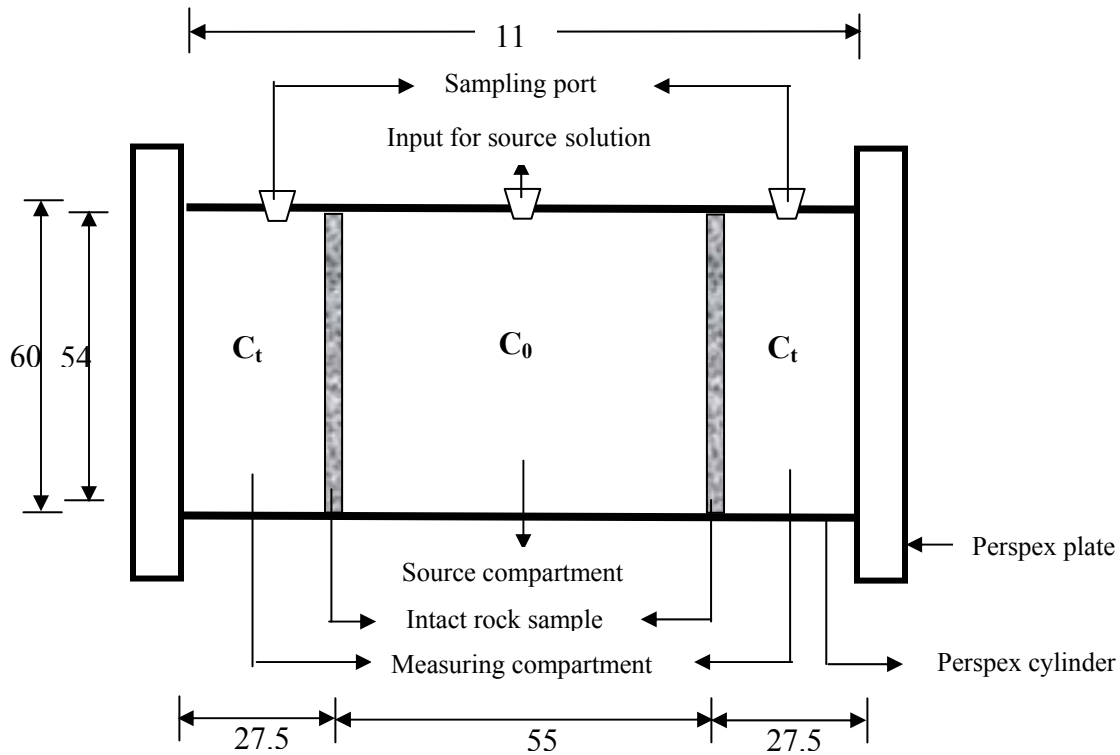
Type-II Diffusion cell

The diffusion cell fabricated for testing fractured rock samples, at 1-g and *N*-g, is shown in Fig. 2. As shown in the figure, the diffusion cell consists of a 160mm long graduated Perspex cylinder with inner and outer diameters of 54 and 60mm, respectively. The rock sample divides this cell into a source compartment (250 cc) and a measuring compartment (~50cc). A Perspex base plate (100mm×100mm×10mm) is provided so that the cell can be placed on a horizontal surface. A fracture is created in the rock sample of thickness, L, 60mm, with the help of a rock core splitter, along its axis. The average width of the fracture, as measured by a microscope, is 0.148mm. Later, this rock sample is tight fitted in the diffusion cell and sealed (using silicone adhesive).

Calibration of the Centrifuge

Calibration of the centrifuge is carried out with respect to the diffusion cells designed for the study. To obtain the effective radius of rotation, r, Eq. 1 is modified as:

$$r = (r_0 - t_b - t_m) \tag{Eq. 6}$$

where r_0 is the maximum outer arm radius, t_b is the thickness of base plate (= 10 mm) and t_m is the height from the top of the base plate to the middle of the rock sample.

Fig. 1 Type-I diffusion cell

The t_m for the Type-I set up is 50.25mm whereas, for the Type-II set up it is 58.5mm. For the accelerated environment:

$$\omega = \sqrt{\frac{N.g}{r}}$$
(Eq. 7)

Substituting $\omega = 2\pi.n/60$, where n is the speed of centrifuge in rpm; Eq. 7 can be rewritten as:

$$n = \frac{60}{2\pi}\sqrt{\frac{N.g}{r}}$$
(Eq. 8)

Eq. 8 can be used to calculate appropriate centrifuge speed (rpm) at a required level of centrifugal acceleration, N. The speed at various acceleration levels, for these diffusion cells, is presented in Table V.

Table V Calibration of the Centrifuge.

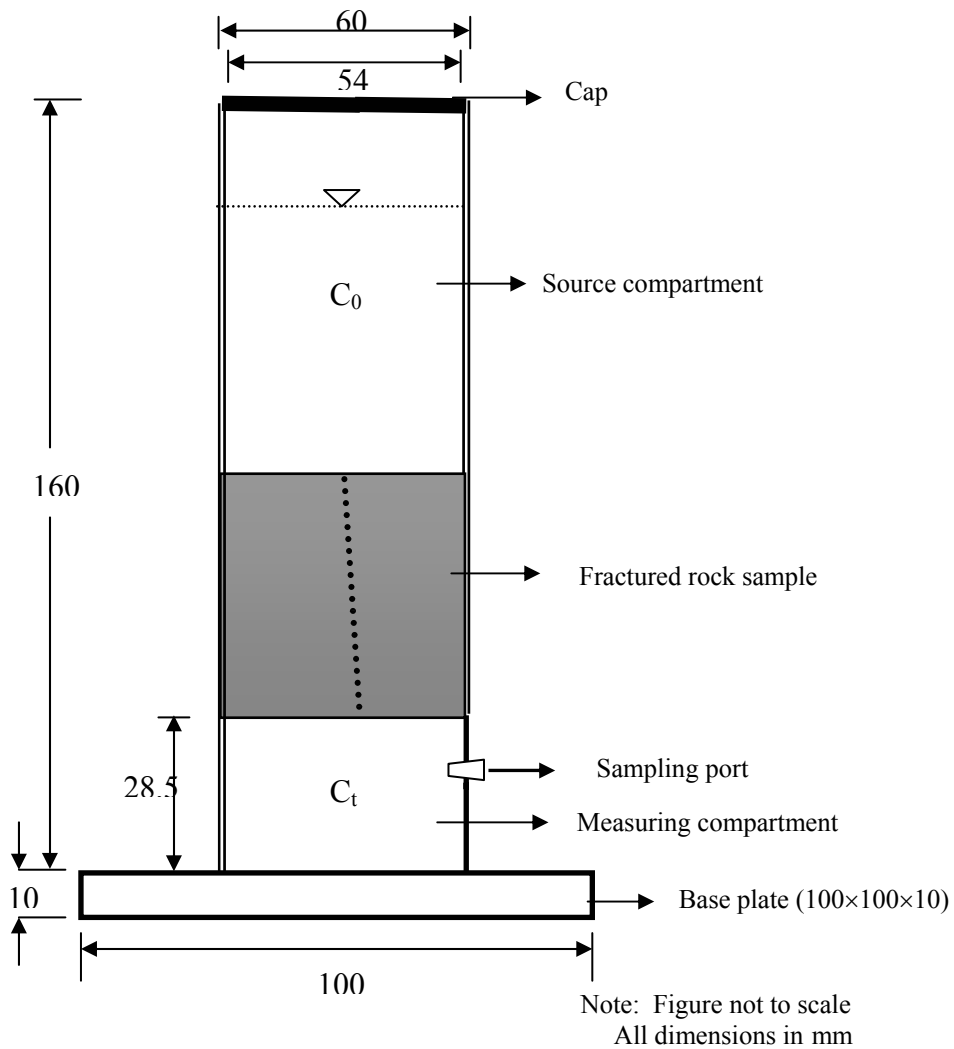| N-g	rpm	
	Type-I Diffusion cell	Type-II Diffusion cell
33	340	346
50	419	425
75	513	521
100	592	602

Fig. 2 Type-II diffusion cell

Nature of source solutions

Two sorbing ions, Cesium as CsI and Strontium as $SrCl_2$, and one non-sorbing ion, Iodide as CsI, have been used as the model contaminants in the present study. Diffusion of these ions has been studied in their non-radioactive form, due to the lack of facilities associated with handling of radioactive solutions. As such, the diffusion behaviour of all these ions would be similar in their radioactive and non-radioactive form. However, the advantage of using radionuclides for such studies is that the diffused concentration can be measured to the order of 10^{-9} moles/l. For the sake of comparison, diffusion of Cs^{137} and Sr^{85+89} radionuclides has also been studied by conducting 1-g tests.

Detection of diffused ion concentration

The concentration of diffused ions in the measuring compartment is to be measured as a function of time. To achieve this, 2ml of the solution is drawn during the experimentation, at regular time intervals, from the measuring compartment and is tested for diffused ion concentration. The concentration of I ion is determined with the help of a UV spectrophotometer and the concentration of Cs and Sr ions are determined with the help of a Inductively Coupled Plasma-Mass Spectrophotometer, ICP-MS [16]. Later, the solution drawn is replaced with equal volume of ultrapure water. For the sake of simplicity the overall change in concentration of the measuring compartment has been ignored. Radionuclide concentrations are measured in terms of its radioactivity, measured as cps (counts per second) with the help of a Gamma spectrophotometer with Sodium iodide-Thallium, NaI-Tl, crystal attached to it.

Computation of diffusion coefficient (D_i)

The concentration of the solution in the measuring compartment, C_t, is monitored with varying time and centrifugation efforts. This concentration when normalized with the source concentration (C_0) and plotted as (C_t/C_0) versus t yields the diffusion curve for the contaminant, as described in the following:

For the test setup used in the present study, the 1-D diffusion equation can be written as:

$$D_i \frac{\partial^2 C(x,t)}{\partial x^2} = \alpha. \frac{\partial C(x,t)}{\partial t}$$

(Eq. 9)

where D_i is the diffusion coefficient and α is the rock capacity factor. With the help of suitable initial and boundary conditions, Eq. 9 can be solved analytically, as discussed below:

Fractured rock mass

For the fractured rock mass the following initial and boundary conditions can be used [17].

$C(x,t)\big|_{t=0} = 0$ for $x > 0$ ....(a)

$C(x,t)\big|_{x=0} = C_0$ for $t \geq 0$ ....(b)   (Eq. 10)

$\dfrac{\partial C(x,t)}{\partial t}\bigg|_{x=L} = 0$ for $t > 0$ ....(c)

If the concentration of a radionuclide at x=L is such that C(L, t)<<C₀, and t is large then the solution of Eq. 9 can be represented as:

$$\frac{C_t}{C_0} = \left(\frac{D_i.A}{L.V} \times t - \frac{\alpha.A.L}{6V} \right)$$ (Eq. 11)

where A and L are the area and thickness of the rock slice, V is the volume of the solution in the measuring compartment and C₀ is the concentration of the source solution. Eq. 11 represents a straight line with slope, s:

$$s = \left(\frac{D_i.A}{L.V} \right) \qquad \text{or} \qquad D_i = \frac{s.L.V}{A}$$ (Eq. 12)

Intact rock mass

Eq. 9 can be solved analytically [18] by imposing the following modified boundary conditions for the intact rock mass.

$C = 0$ for $x > 0$ and $t = 0$ ....(a)

$C = C_0$ for $x = L$ and $t \geq 0$ ....(b)   (Eq. 13)

$\dfrac{\partial C}{\partial x} = 0$ at $x = 0$ for $t \geq 0$ ....(c)

$$\ln \left[\frac{\pi}{4} \left(1 - \frac{C_t V}{C_0 A L} \right) \right] = - \frac{D_i \pi^2 t}{4L^2}$$ (Eq. 14)

$$- \frac{\pi^2 D_i}{4L^2} t = \ln(G)$$ (Eq. 15)

where $G = \dfrac{\pi}{4}\left(1 - \dfrac{C_t V}{C_0 A L} \right)$, V is the volume of the solution in the measuring compartment, A is

the area of the rock sample, and C₀ and C_t are initial and instantaneous radionuclide concentrations, respectively. Eq. 15 indicates that from the slope, s, of ln (G) versus t relationship, the diffusion coefficient, D_i, can be determined using the following relationship.

$$D_i = s \times \left(-\frac{4L^2}{\pi^2} \right) \qquad \text{(Eq. 16)}$$

For the fractured rock samples, the slope of the linear portions of the diffusion curves has been used for determining D_i. However, for the intact rock samples, slope of the ln (G) versus time graphs plotted using Eq. 16 with modified boundary conditions have been used for determining D_i.

RESULTS AND DISCUSSION

To demonstrate the utility of the geotechnical centrifuge for modeling diffusion of various ions through intact and fractured rock samples over a short duration, results have been analysed by time modeling for different N-g tests. For the sake of completeness, only the diffusion curves plotted for I, Cs and Sr ions through fractured rock sample are depicted in Fig. 3. It is generally noticed that the diffusion process is comparatively linear for fractured rock samples at N-g where as it is ion specific for the intact rock samples. In the case of intact rock samples the diffusion trend becomes linear after passing through a transition phase. Similar trends are observed for the radionuclides Cs^{137} and Sr^{85+89}. This transition phase depends on the rock capacity factor, α and is an indication that the rock sample has attained sorption saturation [19]. For fractured rock samples no such transition phase is observed. This may be attributed to the fact that the samples have attained sorption saturation within the very short time of the experiment. D_i values computed for the ions are presented in the Table VI and Table VII for the fractured and the intact rock samples respectively.
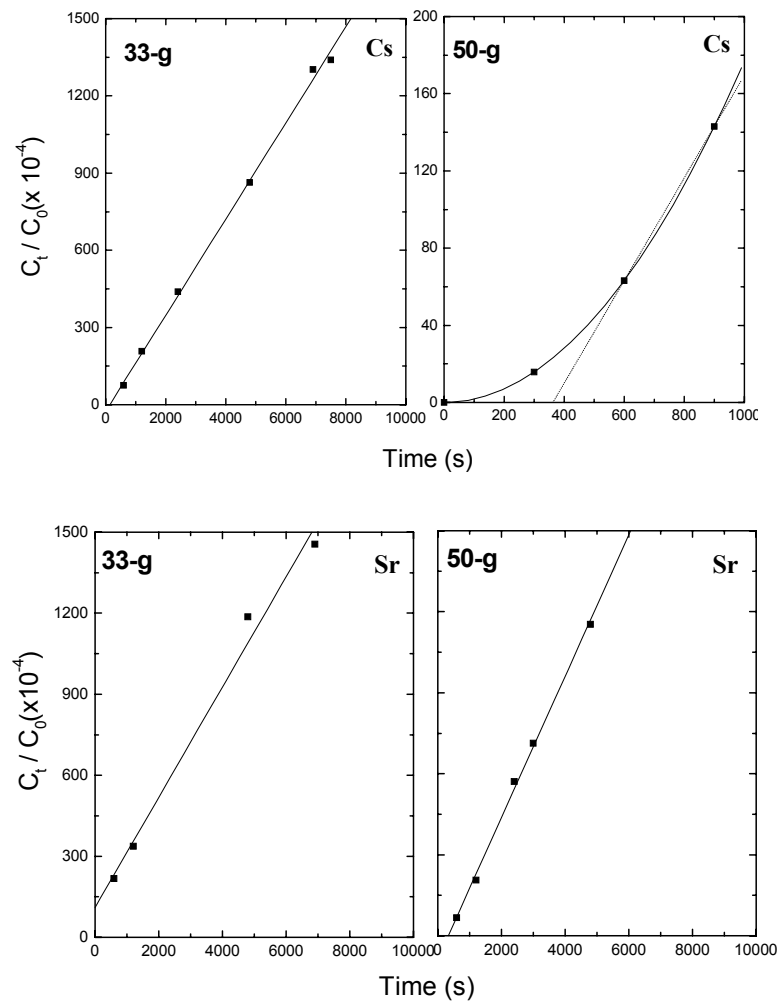
Fig. 3 Diffusion curves for I, Cs and Sr ions through
fractured rock sample at *N-g*.

Table VI D_i for Various Ions through
Fractured Rock Sample at N-g.

Ion	D_i ($\times 10^{-9}$ m^2/s)		
	1-g	N-g	
		33	50
I	6.710	8.381	1.565
Cs	0.834	0.603	0.862
Sr	0.856	0.639	0.847

Table VII D_i for Various Ions through Intact Rock Sample at N-g

Ion	D_i ($\times 10^{-14}$ m^2/s)				Reported values [20,21] for granites (1-g)
	N-g				
	1	50	75	100	
I	3.726	2.033	2.635	4.892	2 to 20
Cs	1.298	2.343	1.784	2.629	3.2
Sr	2.511	3.357	1.592	3.308	2.7
Cs^{137}	1.235	-	-	-	3.2
Sr^{85+89}	1.527	-	-	-	2.7

-      test could not be conducted

It can be noticed from the D_i data that the values obtained for the centrifuge tests are in agreement with those obtained for the normal laboratory diffusion experiments (1-g tests). This indicates the validity of the centrifuge modeling technique to evaluate D_i of various ions over a very short experimental duration.

Scale factor for diffusion time

In order to obtain the scale factor, y, for the diffusion time, t, the following analysis has been conducted. Scaling of the diffusion number, π_4, as defined by Arulanandan et al. [8] leads to similarity of the diffusion processes in the model and its prototype. The diffusion number, π_4, can be expressed as:

$$\pi_4 = (D_i \cdot t / L^2)$$ (Eq. 17)

where D_i is the diffusion coefficient, t is the diffusion time and L is the sample thickness. Eq. 17 indicates that for the same contaminant and the same rock sample, the following condition $D_{im} = D_{ip}$ must be satisfied. As such, to maintain similarity of the diffusion number, the condition $t_m = N^{-y} t_p$ must be valid.where t_p and t_m correspond to the diffusion time for the prototype and its model, respectively. The above condition can be rewritten as follows to evaluate y.

$$y = -\frac{\ln(t_m/t_p)}{\ln(N)}$$ (Eq. 18)

Eq. 18 indicates that y is the slope of the log-log relationship between diffusion time, t, and N. The negative sign associated with y indicates that as N increases the diffusion time decreases. To obtain value of y for the fractured and intact rock samples, the following methodology has been adopted. The time required to achieve $C_t/C_0 = 20 \times 10^{-4}$, for I ion, and 800×10^{-4}, for the other ions, has been obtained for the fractured rock sample, as presented in Table VIII. Similarly, the time required to achieve $C_t/C_0 = 20 \times 10^{-4}$ for different ions for the intact rock samples are presented in Table IX.

Table VIII Diffusion time for Various Ions at N-g through Fractured Rock Sample.

C_t/C_0 ($\times 10^{-4}$)	Ion	t (s)		
		$N=1$	33	50
20	I	5.02×10^6	8117	3842
800	Cs	0.20×10^6	1755	691
800	Sr	0.22×10^6	1378	365

Table IX Diffusion Time for Various Ions at N-g through Intact Rock Sample.

Ion	t ($\times 10^5$ s)			
	$N=1$	50	75	100
I	964	4.6	3.55	1.9
Cs	725	1.12	0.5	0.36
Sr	373	1.0	0.6	0.3

Data presented in Tables VIII & IX when plotted as depicted in Fig. 4, yield values of scale factor, y. It can be noted that irrespective of rock mass condition, the value of scale factor, y, is close to 2, which indicates the validity of the scale factor for the diffusion time. This demonstrates the utility of the centrifuge modeling technique for modeling diffusion of the sorbing ions through the fractured and intact rock masses.

CONCLUSIONS

The present work is an attempt to simulate and model transport of non-sorbing (Iodide) and sorbing (Cesium and Strontium) ions through intact and fractured rock masses. Utility of the small geotechnical centrifuge, as a research tool, to model migration of various non-sorbing and sorbing ions through the intact and the fractured rock mass has been established. Diffusion cells (Type-I and Type-II) designed to simulate the migration of ions through the intact and fractured rock mass are found to be quite useful for N-g experiments. The diffusion coefficient for the model, D_{im}, and for the prototype D_{ip} experiments match very well and these values are in agreement with literature values for similar ions through Granite. The scale factor for the diffusion time is found to be 2 for non-sorbing and sorbing ions through the fractured and intact rock mass. The study demonstrates the usefulness of the geotechnical centrifuge for modeling radionuclide migration through the rock mass in a short duration. It has been noted that for the sorbing ions the time taken to achieve $C_t/C_0 = 20 \times 10^{-4}$, corresponding to 1-g and 100-g, would be

of the order of 10^8s (=1157days) and 10^5 s ($\cong$1day), respectively indicating that conventional laboratory experiments are impractical. In such situations, centrifuge modeling is the only viable alternative.
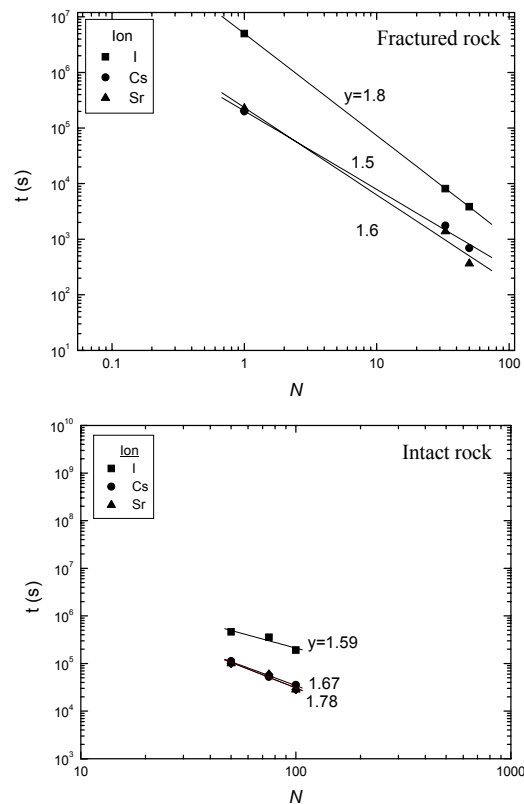

Fig. 4. Scale factor for diffusion time
for various ions through fractured
and intact rock sample

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