

Transport of chloride through saturated soil column: An experimental study

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Abstract. The groundwater is a very important part of the environment and must be protected for the benefit of the present and future generation. The contamination of soil and groundwater by chemicals has become an increasing concern in the recent past. These chemicals enter the groundwater system by a wide variety of mechanisms, including accidental spills, land disposal of domestic and industrial wastes and application of agricultural fertilizers. Once introduced into an aquifer, these contaminants will be transported by flowing groundwater and may degrade water quality at nearby wells and streams. For improving the management and protection of groundwater resources, it is important to first understand the various processes that control the transport of contaminants in groundwater. Predictions of the fate of groundwater contaminants can be made to assess the effect of these chemicals on local water resources and to evaluate the effectiveness of remedial actions. In this study, an attempt has been made to investigate the behaviour of solute transport through porous media using laboratory experiments. Sodium chloride was used as a conservative chemical in the experiment. During the experiment, pulse boundary condition and continuous boundary conditions were used. Experimental results have been presented for conservative solute transport in the sand. The pattern of the break through curve remains almost same in all the cases of varying flow rate and initial concentration of conservative chemical.

Keywords: contaminant transport; porous media; saturated; solute column experiment; pulse type and continuous type

1. Introduction

The groundwater is a valuable natural resource. It is generally less susceptible to contamination and pollution when compared to surface water bodies. Also, the natural impurities in rainwater, which replenishes groundwater systems, get removed while infiltrating through soil strata. But, In India, where groundwater is used intensively for irrigation and industrial purposes, a variety of land and water-based human activities are causing pollution of this precious resource. The groundwater must be protected for the benefit of the present and future generation. The source contaminating groundwater is normally detected much later, since groundwater moves at a very slow rate in aquifers. Most of the groundwater contaminants are reactive in nature and they

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infiltrate through the vadoze zone, reach the water-table; and continue to migrate in the direction of groundwater flow and pollute the soil media and groundwater. Therefore, it is essential to understand the transport process of contaminants through the subsurface porous media.

1.1 Sources of subsurface contamination

The waste from residential, commercial, industrial, agricultural activities and nuclear waste (Zheng and Bennett 2002, Schwartz and Zhang 2003) can seriously affect groundwater quality. Leaching of natural chemical deposits can result in increased concentration of chlorides, sulphates, nitrates, Iron and other inorganic chemicals. Naturally occurring substances in rocks like iron, manganese, chlorides, fluorides, sulphates or radionuclides are dissolved in groundwater.

The leachate from landfills or industrial waste disposal sites frequently contains toxic substances. Some care is necessary in choosing the location of these landfills to prevent significant amount of toxic substances from being transported into the groundwater system. The movement of pollutants through relatively impermeable soil is quite slow; however, it is conceivable that significant pollution might occur in the long term and so it is important that such disposal sites be designed to prevent the possible contamination of groundwater system in both the short term and long term.

1.2 Solute transport through soils

The solute transport in soil and groundwater is affected by a large number of physical, chemical and microbial processes and media properties. Advection of contaminants is the movement caused along with the flowing groundwater at the seepage velocity in porous media. Diffusion is the process by which a contaminant in water will move from an area of greater concentration towards an area where it is less concentrated. The solute is seen to spread out from the flow path. This spreading or mixing phenomenon is called dispersion. Adsorption of a solute onto an aquifer material (e.g., clay) results in a reduction of concentration in the aqueous phase and a retardation of the velocity of contaminant migration. The degree of retardation experienced by a particular organic contaminant will depend on the fraction of organic carbon of the aquifer materials. The higher the fraction of organic carbon the more sites there are available for adsorption. An advection dispersion transport equation is generally used for estimating the level of contamination in groundwater.

2. Review of literature

As the present study reports an experimental investigation into the behaviour of solute transport through porous media, some of the works reported by earlier researchers have been reviewed briefly in this section.

Klotz *et al.* (1980) conducted a large number of laboratory and field experiments which dealt with the relationship between longitudinal and transversal dispersion coefficients in loose soil and the mean flow velocity. Barone *et al.* (1992) described a laboratory diffusion-test for estimation of the diffusion coefficient (D) and the adsorption coefficient (K_d) for several volatile organic species in a clayey soil. Rowe and Badv (1996) conducted a series of chloride diffusion tests on a clayey silt, silt and sand- both for single layer and two layer systems and suggested that the existing solute transport theory can adequately predict the chloride migration through landfill liners at near

saturated conditions. Wang *et al.* (1998) conducted the two types of column and well simulation tests to obtain the properties necessary to model the movement of contaminants in porous media.

Rosqvist and Destouni (2000) modelled lithium transport through an undisturbed solid waste sample and a pilot-scale experimental landfill by use of probabilistic Lagrangian approach. Rowe *et al.* (2000) performed the several inorganic diffusion tests on *Geosynthetic Clay Liners* (GCL) and bentonite specimens; and stated that 'there is a linear correlation between the diffusion coefficient and the final bentonite void ratio for both sodium and chloride'. William and Ning (2004) described a new lecture module and laboratory experiment for demonstrating chemical transport phenomena in soils. The Experimental studies provide a proper detailed knowledge and have been conducted to determine the parameters (Badv 2006). Starr *et al.* (2008) performed laboratory experiments and mathematical simulations were used to investigate the transport of a reactive solute through a sand layer located between two silt layers. The experimental results showed breakthrough curves that were delayed in the time of appearance, had a lower peak concentration, and were more highly dispersed as compared to nonreactive solute.

Patrick *et al.* (2011) presented an experimental and numerical investigation of contaminant transport in porous media. Simulations indicated that, a reduction in specimen height yielded earlier breakthrough and higher levels of contaminant mass outflow. Massimo *et al.* (2012) performed multi-tracer laboratory bench-scale experiments and pore-scale simulations in different homogeneous saturated porous media. The results show that a non-linear compound-dependent parameterization of transverse hydrodynamic dispersion is required to capture the observed lateral displacement over a wide range of seepage velocities.

Sharma *et al.* (2013) reported the experimental investigation of contaminant transport through saturated layered soil using the soil column experiment. The results of experimental breakthrough curves showed that the order in which the soil layers were stratified in water -saturated profile did not influence the effluent solute concentration distribution. Swami *et al.* (2013) performed laboratory experiments to investigate the behaviour of solute transport through stratified porous media. A numerical model was used to simulate the breakthrough curves of experimental data of chloride and fluoride. It was seen that the peak of breakthrough curves was higher for the non-reactive solute compared to that of the reactive solute. Yang and Weng (2013) reported a multi-phase model for predicting the effective chloride migration coefficient of ITZ in cement based materials.

Patil and Chore (2014) presented the review of the various experimental and studies with respect to the contaminant transport through porous media and underscored the necessity of more experimental work in order to understand the phenomenon of the transport of the contaminants, rather than resorting to the numerical studies. Paul *et al.* (2014) presented chloride diffusion study in different types of concretes using finite element method.

On the backdrop of the above referred literature, the laboratory experiments have been carried out to investigate the behaviour of solute transport through porous media. The sodium chloride was used as a conservative chemical in the experiments and its transport through saturated soil column was studied. The sand was used as a porous medium. The experiments provide valuable insight about the porous medium and the behaviour of chemicals.

3. Theory and governing equations

In order to predict the environmental consequences of groundwater contamination one should

know where the contaminant will interfere, when it will arrive and what are the potential concentrations. The contaminant is introduced in groundwater by: (i) the *advection* which is caused by flow of groundwater; (ii) the *dispersion* which is caused by mechanical mixing and molecular diffusion; and lastly (iii) the *retardation* which is caused by adsorption.

The mathematical relationship between these processes can be written as follows

$$\frac{\partial}{\partial x_i} \left[D_{ij} \frac{\partial C}{\partial x_j} \right] - \frac{\partial}{\partial x_i} (C v_i) - \frac{C' W'}{n} = R \frac{\partial C}{\partial t} \quad (1)$$

$$V_i = \frac{-K_{ij}}{n} \frac{\partial h}{\partial x_j} \quad (2)$$

$$R = \left[1 + \frac{\rho_b K_d}{n} \right] \quad (3)$$

Where

- C = contaminant transport
- v_i = seepage or average pore water velocity in the direction x_i
- D_{ij} = dispersion coefficient
- K_{ij} = hydraulic conductivity
- C' = solute concentration in the source or sink fluid
- W' = volume flow rate per unit volume of the source or sink
- n = effective porosity
- h = hydraulic head
- R = retardation factor
- x_i = Cartesian coordinate

The following discussion uses a simplified two-dimensional representation to describe the transport of contaminants in groundwater. In a homogeneous, isotropic medium having a unidirectional steady state flow with seepage velocity V

$$D_L \frac{\partial^2 C}{\partial x^2} + D_T \frac{\partial^2 C}{\partial y^2} - V \frac{\partial C}{\partial x} = R \frac{\partial C}{\partial t} \quad (4)$$

Where

- C = contaminant concentration
- V = seepage or average pore water velocity
- D_L = longitudinal dispersion coefficient
- D_T = transversal dispersion coefficient
- R = retardation factor

3.1 Advection

Advection and hydrodynamic dispersion are the physical properties that control the solute flux. Advection is governed by the Darcy's law as it is the transport of the solute with respect to flowing groundwater and hydrodynamic dispersion results from mechanical mixing and molecular diffusion.

Darcy's law states that the flow rate of water through soil from point 1 to point 2 is proportional to the head loss and inversely proportional to the length of flow path

$$Q = -K.A \frac{h_2 - h_1}{L} \quad (5)$$

Where

- Q = groundwater flow rate
- A = cross sectional area of flow
- h_2-h_1 = head loss between point 1 and point 2
- L = distance between point 1 and point 2
- K = hydraulic conductivity

The actual seepage or average pore water velocity can be calculated as

$$V = -\frac{Q}{n.A} = -\frac{K}{n} \frac{h_2 - h_1}{L} \quad (6)$$

Where n is effective porosity or percent of interconnected pore spaces that actually contributes to the flow. Eq. (2) is a conservative estimate of the migration velocity of the contaminant transport. When only advection is considered, a contaminant moves with the groundwater flow at the same rate as water, but in reality it is also affected by dispersion and retardation.

3.2 Dispersion

It is the result of two processes, molecular diffusion and mechanical mixing. The mechanical dispersion or mechanical mixing occurs when contaminated groundwater mixes with non-contaminated groundwater resulting in a dilution of the contaminate which is called dispersion. The molecular diffusion is the process where ionic or molecular constituents move in the direction of their concentration gradients. In this, the constituents move from regions of higher concentrations to regions of lower concentrations, the greater the difference greater the diffusion rate.

The molecular diffusion can be expressed by Fick's law as

$$F = -D_f \frac{dC}{dx} \quad (7)$$

Where

- F = mass flux per unit area per unit time
- D_f = diffusion coefficient
- C = contaminant concentration
- dC/dx = concentration gradient

Fick's law was derived for chemicals in unobstructed water solutions. When this law is applied to porous media, the diffusion coefficient should be smaller because the ions follow longer paths caused by the presence of solid particles in the solid matrix and because of adsorption on solids. This application yields an apparent diffusion coefficient D^* represented by

$$D^* = w.D_f \quad (8)$$

Where 'w' is empirical coefficient less than 1 which takes into account the effect of the solid phase of the porous media on the diffusion. Freeze and Cherry (1979) suggests that 'w' should range between 0.5 to 0.01 to account for tortuosity of the flow path.

Since these two processes cannot be separated in groundwater flow, the coefficient of hydrodynamic dispersion is taken into account.

$$D_L = a_L.V + D^* \quad (9)$$

$$D_T = a_T.V + D^* \quad (10)$$

Where

D_L = longitudinal mechanical mixing component of dispersion

D_T = transversal mechanical mixing component of dispersion

a^L = longitudinal dispersivity

a^T = transversal dispersivity

a rough approximation of a^L by Gelhar *et al.* (1992) is

$$a^L = 0.1 L$$

Where L is the length of the flow path.

For lengths less than 3500 m, given the $a^L = 0.0175 L^{1.46}$

For transverse dispersivity. The value of a^L is typically 1/10 to 1/100 of the longitudinal dispersivity a^L . These coefficients can be determined from breakthrough column tests in the laboratory or tracer tests in the field.

3.3 Advection-dispersion transport

The advection and dispersion is predominant in saturated or nearly saturated clay barriers. For one dimensional flow

$$D_x \frac{\partial^2 C}{\partial x^2} - \tilde{v}_x \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \quad (11)$$

This can also be expressed along a flowline by using L for x where L is the coordinate direction along the flowline. D_L is the longitudinal coefficient of hydrodynamic dispersion and \tilde{v}_L is the average linear velocity along the flowline.

The analytical solution to the Eq. (11) is given below

$$\begin{aligned} \text{Initial condition} \quad C(x, 0) &= 0 \quad x \geq 0 \\ \text{Boundary condition} \quad C(0, t) &= C_0 \quad t \geq 0 \\ \text{Boundary condition} \quad C(\infty, t) &= 0 \quad t \geq 0 \end{aligned} \quad (12)$$

$$C(x, t) = \frac{C_0}{2} \left[\operatorname{erfc} \left(\frac{x - vt}{2\sqrt{D_L t}} \right) + \exp \left(\frac{vx}{D_L} \right) \operatorname{erfc} \left(\frac{x + vt}{2\sqrt{D_L t}} \right) \right]$$

Where x = distance from injection point

Argument of $\exp\left(\frac{vx}{D_L}\right)$ is the Peclet number $P_e = \frac{\tilde{v}_L x}{D_L}$

This is a measure ratio of the rate of transport by advection to the rate of transport by diffusion. Large Peclet numbers ($P_e > 100$) indicate that advection dominates. When advection dominates, the second term on the right hand side becomes negligible. When advection dominates, the second term on the right hand side becomes negligible

3.4 Sorption

The sorption is the exchange of molecules and ions between solid phase and liquid phase, including adsorption and desorption. Adsorption is attachment of molecules and ions from the solute to the solid phase causing a decrease of concentration of the solute this is called Retardation. Desorption is the release of molecules and ions from the solid phase to the solute. The retardation coefficient can be calculated on the distribution or adsorption coefficients of the contaminant and the characteristics of the porous medium as

$$R = \left[1 + K_d \frac{\rho_d}{n} \right] \quad (13)$$

Where

K_d is distribution or adsorption coefficient described previously. The values ρ_d and n are the bulk density and porosity of the soil. The velocity of the contaminant in groundwater can be calculated as follows

$$V_c = \frac{V}{R} \quad (14)$$

Where V_c is the velocity of the contaminant movement in groundwater, V is the groundwater velocity, and R is the retardation factor. A high retardation factor i.e., high adsorption coefficient significantly retards the movement of the contaminant in groundwater.

4. Experimental set-up and procedure

The experiments can be used to obtain the properties necessary to model the movement of contaminant in porous media in a realistic situation. In nutshell, the experiments provide valuable insight about the porous medium, the behaviour of chemicals; and associated processes such as diffusion, dispersion, anion exchange and sorption during transport. The experimental set-up, used in the present investigation, consists of a column, made up of an acrylic pipe with thickness of 0.8 cm, 10 cm diameter and 80 cm length. The cross section area of pipe was 78.50 cm². The details of soil column experiment are shown in Fig. 1.

The tank was filled with granular medium using a funnel and during the filling, the dry soil was carefully packed in small increments into the column avoiding any soil particle – size segregation. The particle-size distribution curve of sand is shown in Fig. 2. The specific gravity of sand particle is 2.6, porosity 0.485, moisture content 10.3% and bulk density 1.507 gm/cm³.

Medium porosity fitted glass end plates were provided as a stable support of the soil column. The soil contains organic and inorganic substances. Therefore, before starting the transport

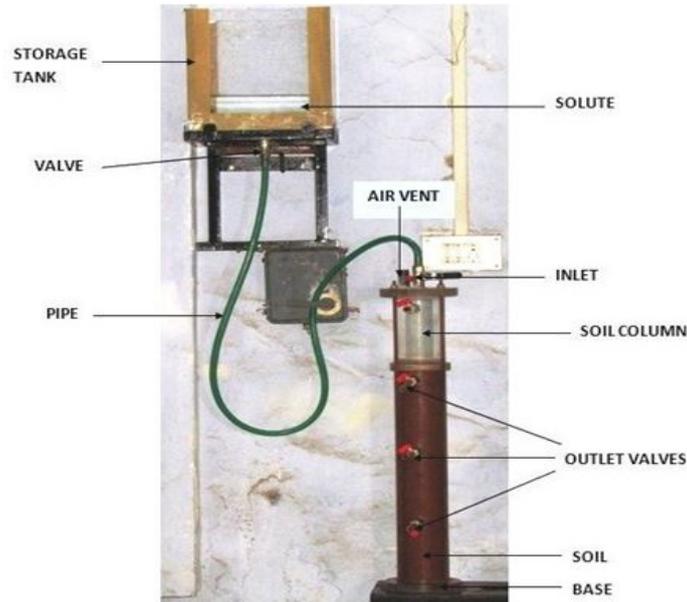


Fig. 1 Soil column with necessary appurtenances

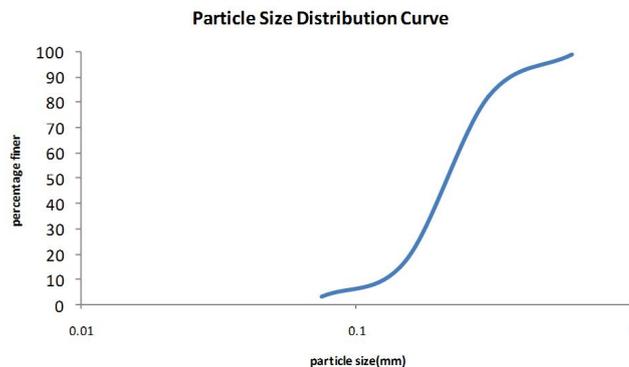


Fig. 2 Particle size distribution curve for a soil used in the present study

experiments, the soil column was washed out by deaired tap water for 2 hours. Utmost care was taken that the soil column always remains flowing. Tap water was tested prior performing the experiments to measure the concentrations of free chloride. After measuring the concentration of tap water, a storage tank was used to store the prepared solution of known concentration. The solution thus prepared was introduced into the soil column at a constant flux, keeping the head constant of input solution. First, the soil column was saturated by the tap water so that the steady-state water flow condition established. The water level was always kept above the grains to achieve a uniform packing and to avoid the entrapment of air bubbles.

Then sodium chloride solution (NaCl) with known concentration as tracer was injected into the column as the input for 30 minutes. Concentration of the tracer NaCl during the experiment was measured in the column with a constant time interval. The solute samples were collected at the

outlet of the column and concentration were measured by the titration procedure. The effluent solutions were collected in fractions of each 15 minutes into the collector. After measuring cylinder was used to collect a fraction of water sample for accurate measurement and to cross-check the volume collected in particular duration of time for velocity calculations. The velocity was calculated by dividing the volume collected in 1 minute by the effective cross-sectional area. All the collected samples were stored in air-tight bottles to protect from direct sunlight. After the sampling each sample was labelled and tested. For measuring chloride in the fraction collected. All the samples were titrated with potassium chromate as indicator and silver nitrate as a titrant. The effluent solute concentration was expressed as relative concentrations (C/C_0), where C and C_0 are the solute concentration in the effluent fraction and input pulse, respectively.

5. Results and discussion

The results for the pulse solute and constant solute boundary conditions have been presented in the subsequent paragraphs by means of breakthrough curves. These curves represent the plot between time and relative solute concentration at the outlet of soil column.

5.1 Pulse solute boundary condition

If the supplied solute source is for the predetermined time, it has a finite pulse source. During the experiment pulse type boundary condition was used. The initial concentration of chloride was increased to 332 mg/liter and the flow rate was kept 30 ml/minute. The break through curve with reference to the data observed for this case is shown in Fig. 3.

The relative concentration with respect to the afore-mentioned case is found to increase with time up to 60 minute. The corresponding relative concentration is observed to be 0.92 and thereafter, again the concentration is observed to decrease. The concentration is found to be constant beyond the time interval of 120 minutes with the average concentration being 0.06. In the next instance, the concentration was kept 178 mg/liter and the flow rate was maintained at 40 ml/minute. The corresponding break through curve is shown in Fig. 4.

The relative concentration with respect to the afore-mentioned case is found to increase with

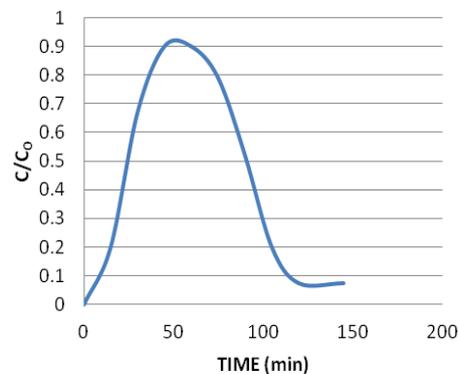


Fig. 3 Break through curve for initial chloride concentration 332 mg/lit with flow rate of 30 ml/minute

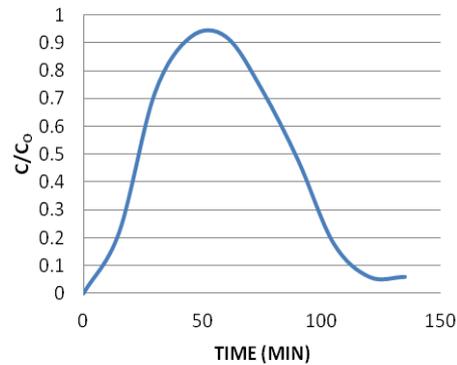


Fig. 4 Break through curve for initial chloride concentration 178 mg/lit with flow rate 40 ml/minute

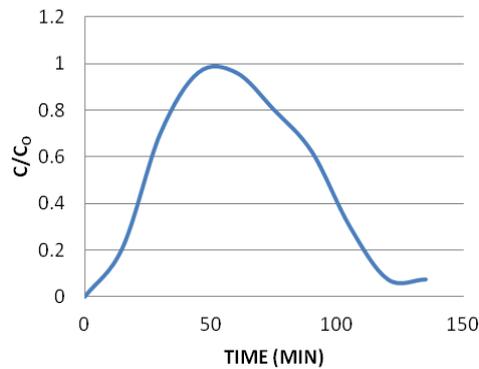


Fig. 5 Break through curve for initial chloride concentration 500 mg/lit with flow rate 60 ml/minute

time up to 60 minute. The corresponding relative concentration is observed to be 0.9 and thereafter, again the concentration is observed to decrease. The concentration is found to remain constant beyond the time interval of 120 minutes and the average concentration is found to be 0.07. Further, both, the initial concentration and the flow rate, were increased. The concentration was kept 500 mg/ litre and the flow rate was kept 60 ml/minute. The break through curve for this case is shown in Fig. 5.

The relative concentration with respect to the afore-mentioned case is found to increase with time up to 60 minute. The corresponding relative concentration is observed to be 0.96 and thereafter, again the concentration is observed to decrease. The concentration is found to remain constant beyond the time interval of 120 minutes. The average constant concentration is found to be 0.07. The observed solute break through data reveals relatively regular distributions at most of the measuring points. With increase in time the relative concentration increases up to certain limits beyond which the relative concentration decreases and becomes constant after certain time.

5.2 Continuous solute boundary condition

If the supplied solute source at the inlet has a constant concentration and it remains throughout the experiment, it is known as the continuous solute source. During the experiment continuous

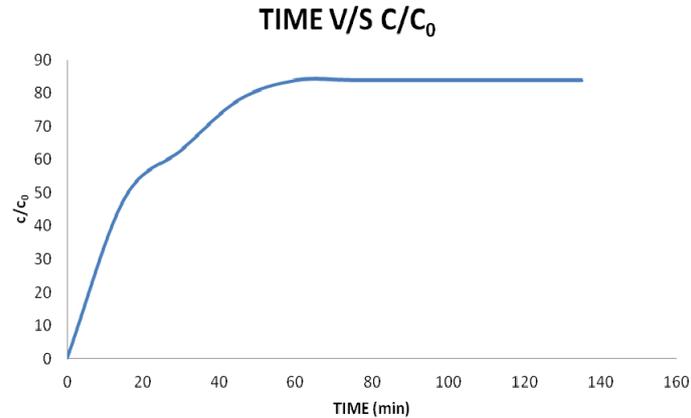


Fig. 6 Variation of initial chloride concentration 150 mg/lit with flow rate 20 ml/minute

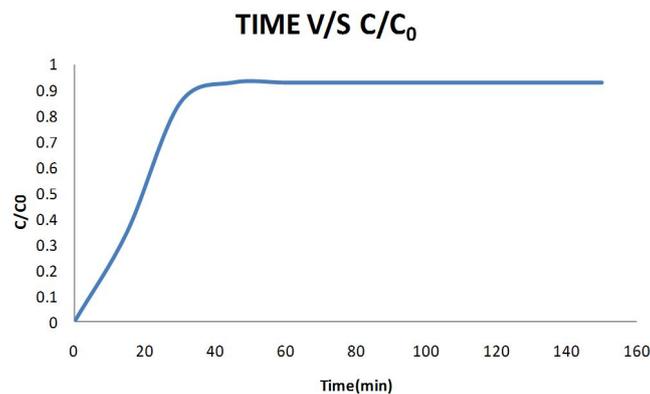


Fig. 7 Variation of initial chloride concentration 600 mg/lit with flow rate 20 ml/minute

solute boundary condition was used. The graphical representation for the initial chloride concentration of 150 mg/lit with flow rate of 20 ml/ minute is shown in Fig. 6.

The relative concentration with respect to the afore-mentioned case is found to increase with time up to 60 minutes. The corresponding relative concentration is observed to be 0.86. The concentration is found to be constant beyond the time interval of 60 minutes. The graphical representation for the initial chloride concentration of 600 mg/lit with flow rate of 20 ml/ minute is shown in Fig. 7. The relative concentration with respect to the afore-mentioned case is found to increase with time up to 45 minutes. The corresponding relative concentration is observed to be 0.93. The concentration is found to be constant beyond the time interval of 45 minutes.

6. Conclusions

From the results presented, following broad conclusions are deduced:

- In pulse type boundary condition, the pattern of the break through curve remains almost

same in all cases of varying initial concentration of the solute and flow rate.

- The maximum concentration in all the cases with respect to pulse type boundary condition is found to attain after 60 minute.
- The maximum value of the relative concentration is observed to be in the range of 0.9-0.96.
- The relative concentration becomes almost stable after 120 minutes in almost all the cases.
- The average value of the relative concentration observed after the afore-mentioned period is almost same and is found in the range of 0.01-0.07.
- In continuous type boundary condition the concentration is found to be constant beyond the time interval of 45 to 60 minutes.

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