

SOLUTE TRANSPORT IN PACKED SAND COLUMNS BY COLLOIDAL GAS APHRONS: EXPERIMENTAL EVIDENCE AND MODELING APPROACH*

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ABSTRACT

Application of microgas dispersions called colloidal gas aphanes (CGAs) in decontaminating soils is an emerging innovative technology. This study was conducted to analyze the solute transport in columns with CGA suspensions and compare that with the aqueous solution. The breakthrough curves for both solutions demonstrated early breakthrough and tailing. The mobile-immobile model (MIM) was able to describe the solute transport by both aqueous solution and CGAs in the packed sand columns. Three column lengths were used in this study. For aqueous solutions the dispersivity increased with the length of the column; however, with CGA suspension, the dispersivity decreased with column length. This indicated that due to the blocked large pores, the flow occurred through narrow channels resulting in diminishing the variation in interstitial velocity.

INTRODUCTION

Accidental spills, improper disposal practices, and leakage from underground storage tanks result in contamination of the subsurface environment. Because of the expenses involved in subsurface remediation by conventional methods, like incineration and landfill disposal, *in-situ* procedures such as “pump heat” technology have been adopted for many sites. Approximately 68 percent of Superfund Records Decisions (RODs) selected groundwater “pump and treat” as a means to

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achieve aquifer restoration in 1990 [1]. Depending upon the nature of contamination and geologic conditions, this treatment can take several years. Even highly soluble contaminants get trapped in the finer pore structure and are difficult to remove due to the preferential flow in high permeability zones. In-situ remediation can be considerably enhanced by using a variety of surfactants [2-4]. Flushing the contaminated soil or aquifer with surfactant solution yields better removal efficiencies because of the increased solubilization of contaminants partitions more of them to the mobile phase. However, even this modification of the "pump and treat" technology does not overcome the problem of preferential flow, making the process less efficient [5-6].

One of the promising techniques that overcomes the problem of preferential flow is the application of microbubbles or colloidal gas aphyrons (CGA) to soil. Application of the CGAs to the soil blocks the high permeability layers or streaks and solubilizes the contaminants. The name "colloidal gas aphyron" (CGA) was originally proposed by Sebba for microgas dispersions [7]. CGAs consist of approximately 66 percent gas by volume and therefore form a low density liquid. These microbubbles do not coalesce easily and differ remarkably from conventional soap bubbles (Figure 1) in their stability and flow through properties [8]. The most striking feature of a CGA suspension is its stability which allows microbubbles to be pumped from the point of generation to the application site. CGAs have been shown to be useful in a variety of environmental applications. Our studies have shown the enhanced effectiveness of CGA flotation process over conventional sparged air or solvent sublation in separation of the organics from aqueous waste streams [9]. Based on the results reported in the literature and our ongoing research on the application of CGAs to mobilize organics from soil columns [10-11], it appears that this technology may be very effective in decontamination of soils. CGAs have also been found to enhance transportation of microorganisms and can increase the available oxygen in the soil so as to enhance biodegradation of organics in the soil [12]. The potential usefulness of CGA technology for in-situ remediation of hazardous waste site suggests that additional information should be obtained on the transport of CGA in soil matrix.

Study of solute transport in porous media is essential to our understanding and application of various subsurface remediation technologies. In view of this, the current study was undertaken 1) to investigate mixing of solutes in packed soil columns when CGA suspensions are injected, 2) to study the possible mechanism of solute transport in columns using CGAs as a flushing solution, and 3) to study suitability of mobile immobile model (MIM) to describe the flow of CGAs in porous media.

THEORY

Due to hydrodynamic dispersion, contaminants migrating through subsurface spread in longitudinal and transverse directions of flow. The longitudinal

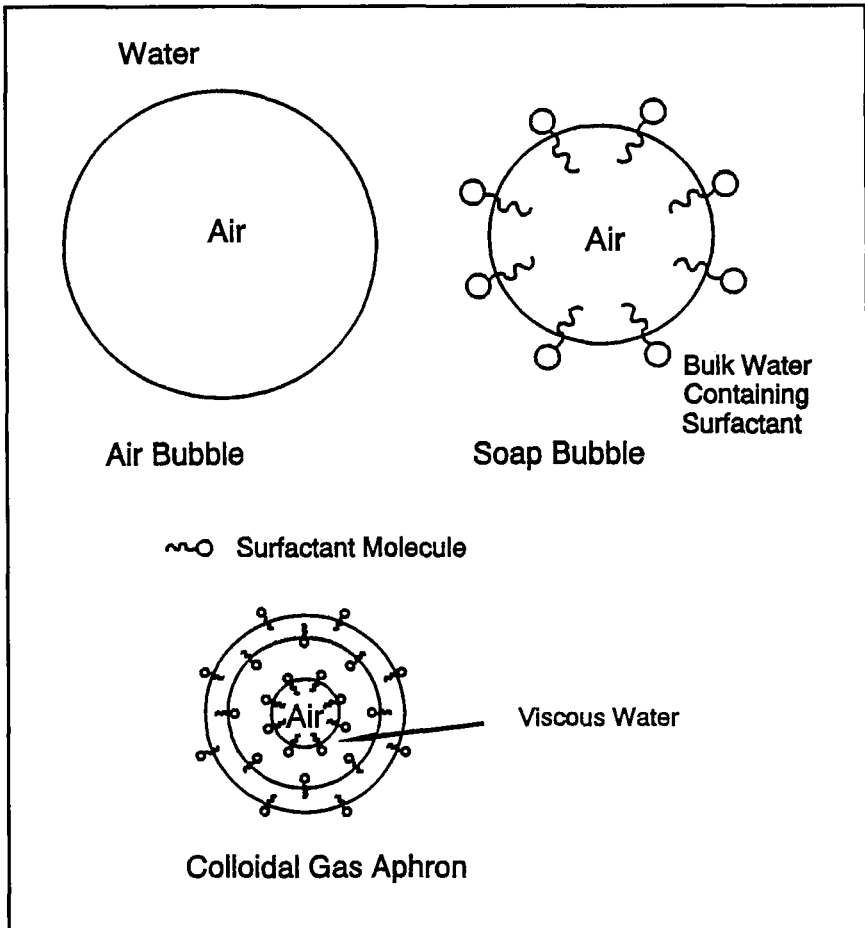


Figure 1. Structure of an air bubble, soap bubble, and a CGA.

dispersion occurs primarily due to velocity variations at the pore levels, which in turn occur due to differing pore sizes. Transverse spreading results from the molecular diffusion that occurs due to concentration gradients. Advection, mechanical dispersion, and molecular diffusion are the most important phenomena that affect the concentration distribution of a solute as it moves through a porous medium. Other factors such as adsorption, precipitation, degradation, and anion exclusion may influence the transport process of solute in the soil; however, they were considered negligible in this study.

One dimensional transport of a nonreactive solute in saturated porous media is generally described with the convective dispersive equation (CDE)

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \quad (1)$$

where C denotes solute concentration ($M L^{-3}$), t time (T), x distance (L), D the dispersion coefficient ($L^2 T^{-1}$), and v the pore water velocity ($L T^{-1}$). Dispersion coefficient D is sometimes described as a linear function of pore water velocity i.e.

$$D = \lambda v \quad (2)$$

where λ , also known as dispersivity (L) is a constant.

One of the limitations of this model stems from the fact that in practice larger pores dominate the flow and mixing of solute in the transport process, resulting in stagnant water bodies in the matrix. The stagnation of water usually results from dead end pore spaces, intra-aggregate water, and thin films that surround the particles within the porous media. Also, CDE fails to take into account the apparent large variation in the microscopic pore-water velocities that occur especially at high flowrates. To address these problems resulting from apparent heterogeneities, Coats and Smith added a capacitance term to the convective dispersive equation [13]. The equation therefore is given,

$$\beta \frac{\partial C_m}{\partial t} + (1 - \beta) \frac{\partial C_{im}}{\partial t} = D \frac{\partial^2 C_m}{\partial x^2} - v \frac{\partial C_m}{\partial x} \quad (3)$$

where C_{im} represents the solute concentration in the stagnant zone, D_m the dispersion coefficient of the flowing fraction, and β the flowing fraction. Subscripts "m" and "im" denote the mobile and immobile phases of water contents in porous media respectively. The mass transfer between the flowing and stationary phase is proposed to be a first order reaction,

$$(1 - \beta) \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \quad (4)$$

where α represents the rate constant (T^{-1}). The flowing fraction (β) is expressed

$$\text{as } \beta = \frac{\theta_m}{\theta} \quad (5)$$

where θ denotes the volumetric water content ($L^3 L^{-3}$) that comprises mobile and immobile water phase and is denoted by subscript "m." The dispersion coefficient D for the overall mobile immobile region can be defined in terms of the dispersion coefficient of the flowing fraction:

$$D = D_m \beta \quad (6)$$

This modified approach assumes that the liquid phase in the porous media remains partitioned in mobile and immobile regions. Convective-dispersive transport is restricted to mobile water phase while transfer of solutes from the

immobile phase is assumed to be diffusion limited. Substituting equations (5) and (6) in equations (3) and (4) the following equations are obtained.

$$\theta_m \frac{\partial C_m}{\partial t} + \theta_{im} \frac{\partial C_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 C_m}{\partial x^2} - q \frac{\partial C_m}{\partial x} \quad (7)$$

$$\theta_{im} \frac{\partial C_{im}}{\partial t} = \alpha (C_m - C_{im}) \quad (8)$$

where q represents the liquid flux (LT^{-1}). Analytical solutions for equations (6) and (7) have been discussed for variety of initial and boundary conditions by De Smedt and Wierenga [14]. Parker and van Genuchten [15] have provided an analytical solution to the mobile immobile model (MIM) for the following initial and boundary conditions:

$$-D \frac{\partial C}{\partial x} + vC = vC_0(t) \quad x = 0, \quad t > 0 \quad (9a)$$

$$\frac{\partial C}{\partial x} = 0 \quad x \rightarrow \infty, \quad t > 0 \quad (9b)$$

$$C = C_i(x) \quad x > 0, \quad t = 0 \quad (9c)$$

where the subscripts 0 and i denote the inlet and initial concentration respectively. The equation (9a) arises from an assumption that at the injection plane, a boundary layer of infinitesimal thickness develops over which the system parameters change from those of a perfectly mixed inlet reservoir to those of the bulk porous media [15]. Furthermore, for the ion not native to soils, the initial concentration $C_i(x)$ within the soil profile is assumed to be zero. For the present study a pulse of solute with concentration C_p , was introduced for a time t_p . The inlet boundary condition (9a) can therefore be modified as

$$-\frac{D}{v} \frac{\partial C(t)}{\partial x} + C(t) = C_p \quad x = 0, \quad 0 < t < t_p \quad (10a)$$

$$-\frac{D}{v} \frac{\partial C(t)}{\partial x} + C(t) = 0 \quad x = 0, \quad t_p < t < \infty \quad (10b)$$

where C_p and t_p denote the concentration and the duration of the pulse respectively.

In case of unsaturated flow, early breakthrough and tailing of the chemicals not adsorbed or excluded by the media, have been shown to be caused by the immobile fraction of water in the unsaturated soils [16, 17]. The MIM model therefore has not only been used to describe solute transport through saturated

porous media, but several researchers have used the model to describe breakthrough curves from unsaturated media as well [18].

Transport of CGAs Through Porous Media

CGAs differ in their structure from ordinary soap bubbles since their encapsulating film has an inner as well as an outer surface and these surfaces have surfactant monolayers adsorbed on them (Figure 1). The encapsulation of a microbubble in a double layer of surfactant molecules, as observed with CGAs, retards its coalescence, thereby increasing its stability. "Stability" of microbubble dispersion can be defined as the length of time during which the number of bubbles and their size distribution remains constant. CGA dispersions, if left undisturbed, will eventually cream and convert to ordinary foam, leaving a clear aqueous layer below.

In a soil washing/flushing operation the contaminant is initially removed by displacing the trapped globules and later it is transported in its solubilized form. Preliminary work with CGAs in our laboratory has indicated that solubilization is the dominant mechanism of contaminant removal [11]. CGAs offer a way of lowering the interfacial tension between organics and water while at the same time providing the viscous forces needed for an efficient areal sweep.

The concept behind using CGA lies in the fact that the gas phase will block the larger pores with lamella. As a result, the suspension is forced through smaller pores where the contaminant is trapped at residual saturation. This flushing medium would prove helpful in heterogeneous medium where high permeability regions will remain plugged, thereby forcing the CGAs to flow through the low permeability contaminated regions. Although the CGA suspension is a homogeneous fluid at the time of pumping, within the porous media separation of the air and solution phase may occur thus resulting in two-phase flow system. The mechanism by which CGAs are transported within porous media may be similar to that of foam. Fugate studied the flow of CGAs through a flume and noted that they tended to accumulate near the point of injection, forming a stationary front [19]. However, in another study carried out by Longe, two separate fronts of gas and liquid were observed [20]. The gas front followed the liquid front owing to its lower permeability. Solute transport by CGA suspension, however, was not considered in these two studies.

Opinions differ among researchers about the mechanism by which the foam or its components move through porous media. Mahmood and Brigham [21] have summarized possible mechanisms for flow of foam in a porous media as follows:

1. **Bubble Flow:** Homogeneous flow of gas and the surfactant solution
2. **Intermittent Flow:** Foam flows in such a way that liquid is transported by a continuous network of liquid membranes, while gas flows as a discontinuous phase through breaking and reforming the bubbles.

3. Trapped Gas Flow: Some gas in the foam is trapped in the porous media while the remainder flows as a free phase. The flowing fraction of the gas is generally small compared to the fraction of trapped gas.
4. Membrane Flow: Foam is generated as lamella at specific locations in the porous media.

It should be noted that in most of these studies, the porous media was first saturated using surfactant solution with concentrations ranging from 1 percent to 5 percent, and air was then injected into the porous media. Since CGAs are generated with much lower surfactant concentrations and pumped as a homogeneous liquid, their behavior in the porous media may not be identical to that of the foam. Also, because of the intricate nature of CGAs and the complex nature of the porous media, probably no single model can completely describe the transport of CGA in the soils. In practice it is more likely that in the porous media more than one mechanism is at work that varies spatially as well as temporally. Our visual observations during column flushing experiments with CGAs have further indicated this.

MATERIALS AND METHODS

Production and Characterization of CGAs

Based on methods suggested by Sebba [22], a unit was fabricated in our laboratory for the production of CGAs. It consisted of a motor fitted on top of a 3 liter cylindrical container. A flat disk, 50 mm in diameter, was mounted at the end of the shaft that was rotated at 8000 rpm. The level of the surfactant solution is initially adjusted to be approximately 15 mm above the disk. Strong waves are created on the liquid surface when the disk is rotated at very high speeds. The waves strike the baffles that are fixed to the lid of the container. On reentering the solution, the waves entrain air in the form of microbubble dispersion or colloidal gas aphanes. Size distribution of CGAs was determined using a particle size analyzer (Microtac 7995-10, Leeds and Northrup). A detailed description of the CGA apparatus and size distribution analysis can be found in our previous work [23].

In this study CGAs were generated using a nonionic surfactant Tergitol 15-S-12 (MW = 738) at its critical micellar concentration (CMC) 0.15 mM. Samples of the surfactant were obtained from Union Carbide Inc. Tergitol was selected because, being nonionic, it is less likely to be adsorbed on the negatively charged sand. Furthermore, CGAs generated with Tergitol were observed to be smaller and remained in suspension for longer time than both cationic and anionic surfactants [23]. Most of the microbubbles had diameters between 30 and 200 μm . The mean diameter of the microbubbles was found to vary between 65 and 80 μm and the standard deviation of bubble diameter varied between 29 and 35 μm .

Experimental Procedure

The sand used in this study was tested to be free from organic and inorganic impurities. Sieve analysis of the sand revealed a d_{60} of 0.85 mm and d_{60}/d_{10} ratio of 2.86. Glass columns (6.3 cm ID) with lengths of 2.5, 11.5 and 30 cm were packed by adding sand in 90 gm portions and were compared to achieve an approximate bulk density of 1.66 gm/cc. Stainless steel screens were placed on top and bottom of the packed sand. The experimental setup is shown in Figure 2. Two sets of experiments were carried out using Cl^- as a nonreactive tracer. The effluent was collected using fraction collector. In the first set of experiments, breakthrough curves were obtained with aqueous solution pumped with a Masterflex positive displacement pump (Cole Parmer Inc.) at a constant flow rate between 8.5 and 9.0 ml/min. For these experiments, the column was initially saturated with 0.05 M KNO_3 solution, and a pulse equivalent to one pore volume of 0.1 M NaCl was pumped in at the same rate. The column was then flushed with 0.05 M KNO_3 solution to obtain the Cl^- breakthrough curves.

For the second set of experimental runs, the column was initially saturated with 0.05 M KNO_3 , and a pulse of CGAs generated with 0.1 M NaCl and 0.15 mM Tergitol was pumped into the column. Gas comprises approximately 66 percent of CGA suspension by volume. Since the time of CGA tracer pulse equaled the aqueous pulse time, the actual volume of tracer pumped with CGA was less than one pore volume. The volumetric measurements of the effluent began as soon as the first bubble appeared at the effluent end. The column was then flushed with CGAs generated with 0.05 M KNO_3 and 0.15 mM Tergitol to obtain the

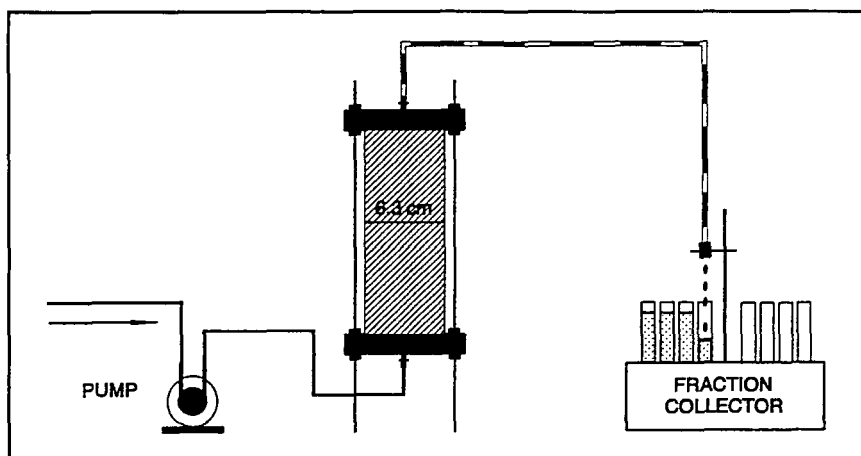


Figure 2. Schematic experimental setup.

breakthrough curves. After each CGA run the contents of the column were removed and its moisture contents were determined using gravimetric analysis.

RESULTS AND DISCUSSION

The experimental results on Cl^- breakthrough using water and CGA suspension for three different column lengths are shown in Figures 3 and 4 respectively. Experimental observations appear to be skewed, showing both early arrival and slow approach to the final concentration. It is not surprising that attempts to fit the CDE model to the experimental data were not successful since CDE predicts symmetrical peaks. Because of the inability of the CDE model to fit the data, an attempt was made to fit the Cl^- breakthrough generated by water and CGA suspension using MIM. The program CXTFIT developed by Parker and van Genuchten [15] allows one to fit the analytical solution of equations (7) and (8) for the dimensionless concentrations (C_i/C_p) as a function of number of pore volumes, T :

$$T = v \frac{t}{L} \quad (11)$$

where v represents the pore water velocity, L the length of the column, and t the time at the measurement. The curve fitting method minimizes the sums of squares of the residuals between the observed and the calculated concentrations. In this study, the observed data was fitted for parameters D_m , number of pore volumes in the tracer pulse T_p , β , and ω . The dimensionless parameter ω is used to calculate the reaction rate constant (α) and is defined as:

$$\omega = \alpha \frac{L}{q} \quad (12)$$

The predictions by MIM are shown by the solid lines in Figures 3 and 4. Close agreement between the experimental data and the predictions for different column lengths validates the applicability of MIM for predicting the solute mixing process in soil matrix using both water and CGA suspension.

Figure 3 shows the breakthrough curves of solute with CGAs for different column lengths. Although the shape of the breakthrough curves by the CGAs resembles the tracer displacement with aqueous solution (Figure 4), there seems to be a difference in the tailing portion of the curve. In the case of CGA runs, the tailing becomes more pronounced as the column length increases, indicating that the reaction between mobile and immobile fraction becomes a dominant feature of the flow in the longer column. The opposite occurs for the aqueous breakthrough where tailing diminishes with an increase in the column length (Figure 4).

CDE assumes that the effective dispersivity remains constant for any length of the column for a constant velocity, but in practice it increases with the distance

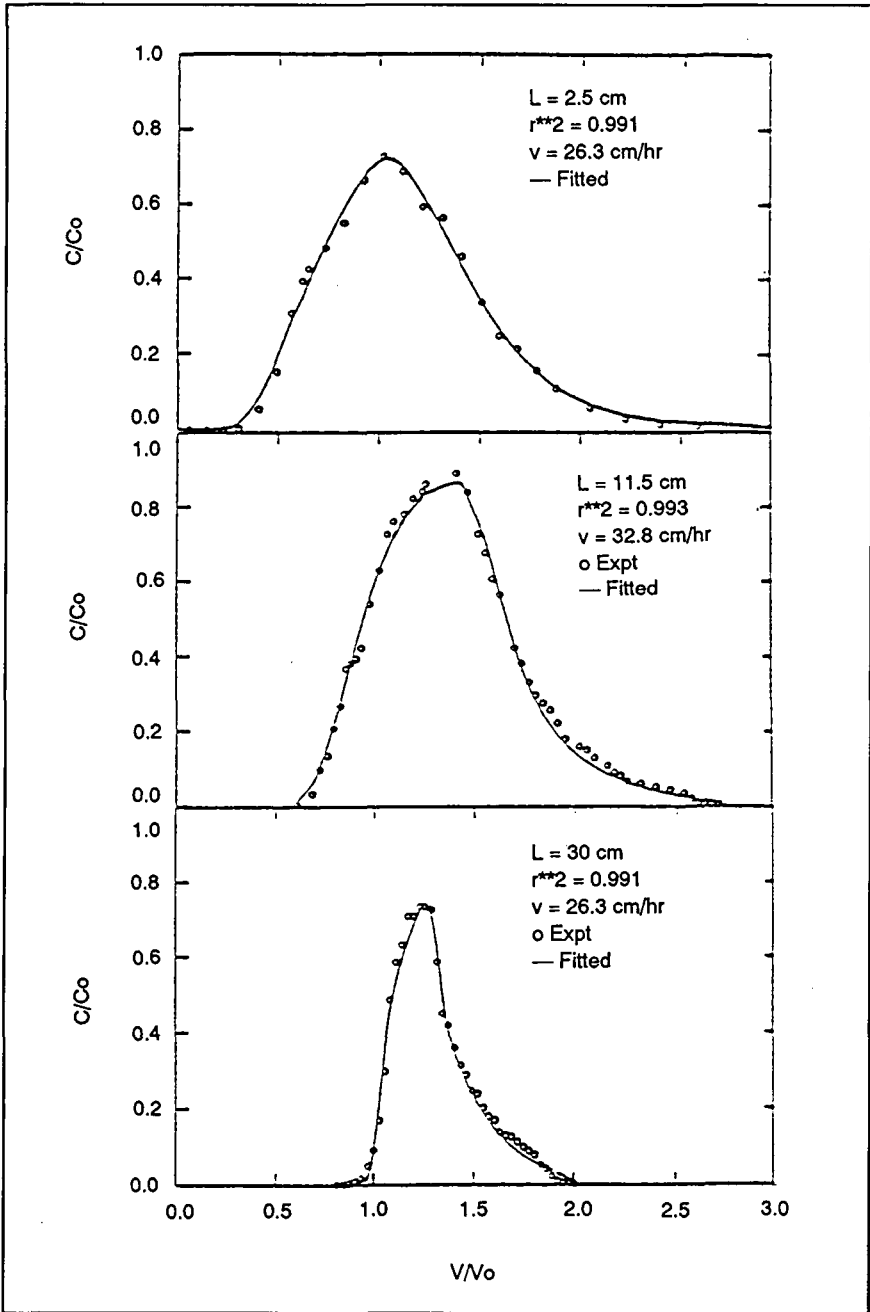


Figure 3. Solute transport by CGA suspension.

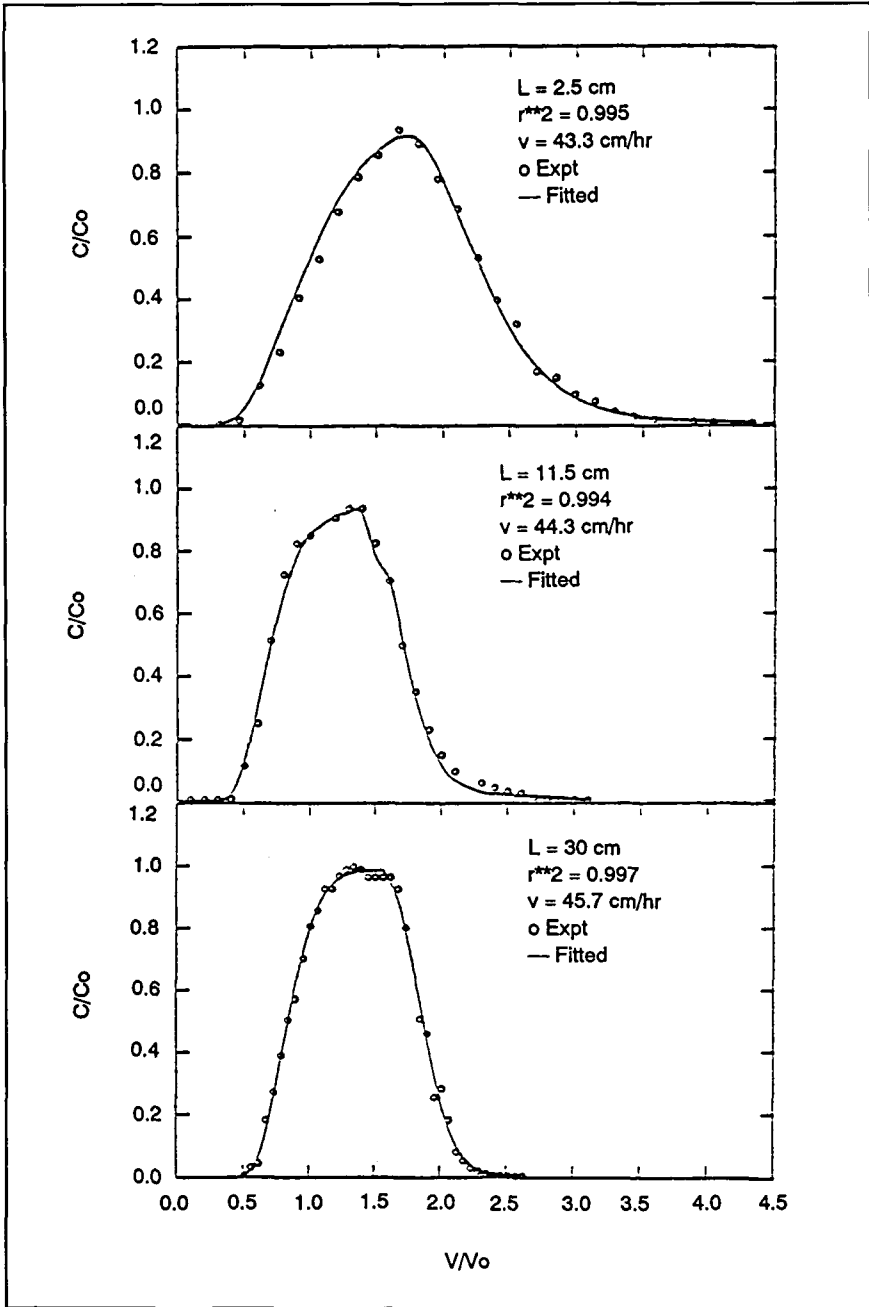


Figure 4. Solute transport by aqueous solution.

from the source. This phenomenon called “scale effect” is well reported in the literature [24-26]. It occurs due to local variation in the water velocity along the direction of motion, causing the solute to spread differently in the longitudinal direction. Since the local variation in the velocity increases with the distance, the dispersion would increase until the velocity differences normal to the direction of the flow are smoothed by transverse mixing [27]. Figure 5 shows the plot of fitted dispersivity versus column length for aqueous solutions and CGAs. For aqueous solutions, the dispersivity increases with the length of the column. These results are consistent with the hypothesis that dispersivity will approach a constant value if the residence time in the column is sufficient to smooth out solute variations by lateral mixing. For CGA runs, dispersivity decreased with an increase in length of the column between 2.5 cm and 30 cm (Figure 5). Although CGA suspension is homogeneous at the time of pumping, it should be noted that separation of air and aqueous phase occurs within the porous media. This situation is analogous to that of a stagnant CGA solution in a column where the bubbles cream to the top and coalesce. Because of the smaller flow through periods, the flow remains more or less homogeneous in the smaller columns. Therefore, separation of air and liquid phases may not be significant in the short columns. The extended residence time in the longer columns, however, will cause separation of phases. Our visual observations confirmed the evidence of phase separation in the longer columns. The effluent from the longer columns appeared in the form of a train of large bubbles separated by the liquid phase. In the case of long columns, flow of the liquid phase through the narrow channels simulated plug flow because the phase separation blocked the high permeability pores by the lamellae. Increase in the length of the column resulted in reduction of the interstitial velocity variation, which in turn reduced the dispersivity.

The pore velocity of CGA suspension was determined by dividing the liquid flux with the volumetric moisture contents of the column at the end of the run. For CGA suspension, the pumping rate was maintained the same as that of the aqueous solution. Liquid flow, however, remained restricted to narrow channels, which resulted in reduction of liquid flux with increasing length of the column. Figure 6 shows the decrease in pore water velocity with increasing column lengths. The time for the solute pulse of CGAs was kept the same as that of the aqueous solution. However, since entrapped air constitutes nearly 60 to 70 percent of CGA suspension, the tracer volume pumped for the CGA runs was less than that for the aqueous solution. In CGA runs, the fitted value T_p decreased with increasing length of the column because of the reduction in liquid flux (Table 1).

The best estimates of the parameters fitted to the curve for CGAs and the aqueous solution are given in Table 1. It can be seen that the fitted parameters α and β do not show a definite trend. Khan and Jury found similar results when they applied the mobile-immobile model to a nonreactive tracer by aqueous solutions in the columns [27]. The mobile water fraction (β) reduced from 0.99 to 0.7 for the aqueous solution. In case of aqueous solution, the rate constant α for the reaction

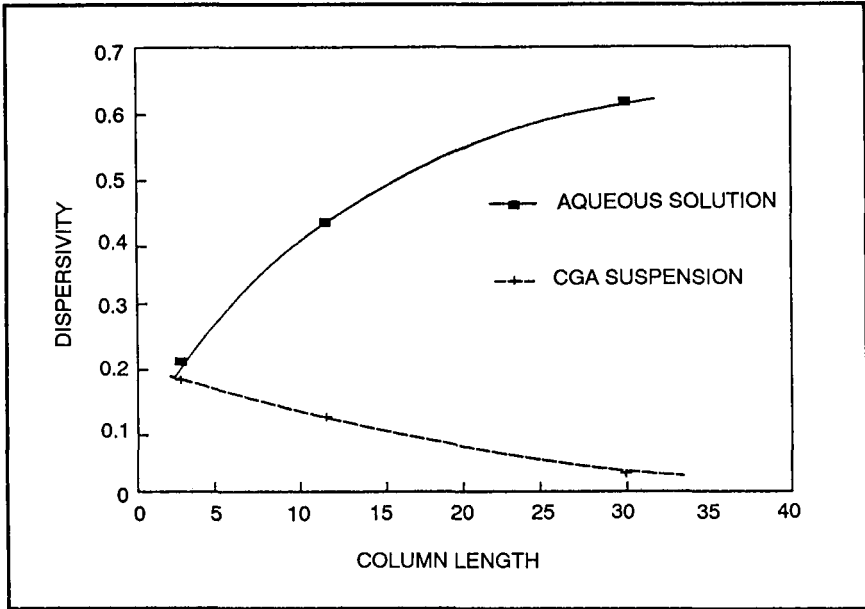


Figure 5. Effect of column length on the dispersivity.

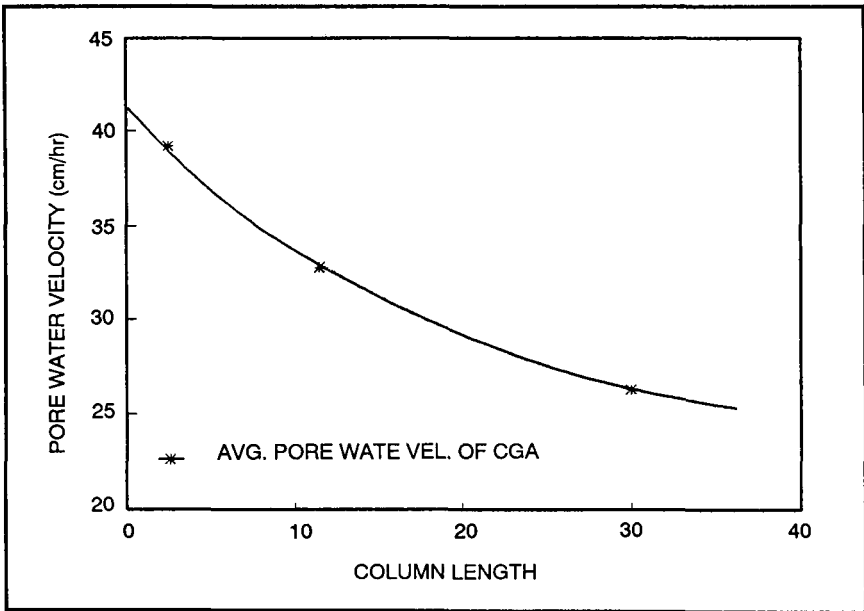


Figure 6. Variation in pore water velocity with column length.

Table 1. Fitted Parameters to Solute Transport by Aqueous Solution and CGA Suspension

Length (cm)	v (cm/Hr)	D_m (cm ² /Hr)	D (cm ² /Hr)	λ_m (cm)	λ (cm)	α (/Hr)	β	T_p
Solute Transport by Aqueous Solution								
2.5	43.3	8.98	8.89	0.21	0.21	0.07	0.99	1.13
11.5	44.3	18.89	13.03	0.43	0.29	0.12	0.69	1.04
30.0	45.7	28.45	23.90	0.62	0.52	0.01	0.84	1.04
Solute Transport by CGA Suspension								
2.5	39.2	9.36	7.21	0.24	0.18	0.23	0.77	0.70
11.5	32.8	4.84	4.07	0.15	0.12	0.24	0.84	0.73
30.0	26.3	0.83	0.71	0.03	0.03	0.12	0.86	0.29

between mobile and immobile phases of aqueous solution, varied between 0.01 and 0.12. Cool et al. report that such a variation in the apparent rate constant α indicates that either the reactions did not follow first order chemical kinetics or that the reaction was not diffusively controlled [28].

For CGA suspension, the α values were higher than for aqueous solution. This could be the result of the complex hydrodynamics of the CGA flow in porous media. When a CGA pulse with Cl^- ions is applied to the column, it may leave behind lamella with high concentration of the solute in the pores. The subsequent CGA suspension would come in contact with these films at random. This type of mixing precludes the gradient induced elution encountered in aqueous solution. In case of CGAs, the mobile fraction β varied within small range for all column lengths. The moisture contents of the columns at the end of the CGA runs were also found to be very similar irrespective of the column length. This significant observation suggests that although the flow was blocked in the large channels, the flowing fraction did not change, and equilibrium was established between the flowing and stationary portions of the porous media.

CONCLUSIONS

Solute transport in coarse sands by aqueous solution is better described by the mobile-immobile model (MIM) than the conventional CDE because the latter fails to predict the early breakthrough and tailing of the breakthrough curve. Furthermore, predictions using MIM model fit the experimental data very well. For experimental runs using aqueous solutions, the dispersivity increased with an increase in the length of the column, whereas the opposite was true for the runs using CGA solutions. The

plot of dispersivity versus column length for aqueous solution indicated that dispersivity will attain a constant value when using a column long enough to smooth out the variation in the interstitial velocities. The MIM model also predicted the breakthrough curves of the solute with CGAs. However, the dispersivity for CGA suspension decreased with the column length, indicating that the flow was channeled through narrow channels that reduced the interstitial velocity variation.

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