Viscous Fingering and Gravity Segregation through Porous Media: Experimental Findings

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ABSTRACT: During downward vertical flow of a viscous solution, the viscous fingering (VF) phenomenon affects miscible displacement of solutes through a soil profile. On the other hand, during horizontal flow, when the liquid residing in a horizontal bed of porous materials is displaced by another liquid of different density, the resulting hydrodynamic dispersion is modified by the formation of a tongue of denser liquid undershooting the less dense liquid, a phenomenon known as gravity segregation (GS). To explore VF and GS phenomena, the authors present laboratory experimental results on the vertical and horizontal transport of bulk solution and ions of different concentrations and/or densities through inert and reactive porous media. The study showed that, with miscible liquids, breakthrough starts later and ends earlier. The authors predicted the behavior of immiscible liquids by the nondimensional gravity segregation number $\beta$: that is, with increase in $\beta$, the segregation becomes extreme. The curve fitting technique CXTFIT 2.0 fitted the experimental breakthrough curves well, showing

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that the apparent coefficients of hydrodynamic dispersion vary much less with pore-water velocity in horizontal than in vertical flow, but retardation factors are not influenced by the orientation of flow. This work is relevant to the preferential flow of viscous liquids such as liquid fertilizers in agricultural fields, oil recovery processes, and the intrusion of saline water into the freshwater of coastal aquifers.

**KEYWORDS:** Viscous fingering; Gravity segregation; Miscible displacement

### 1. Introduction

Development of fingerlike intrusions, commonly termed as viscous fingering (VF), during the miscible displacements of two fluids affect the recovery efficiency in many oil recovery processes (McCloud and Maher 1995). During miscible displacements, one fluid mixes with and displaces another fluid (Kirkham and Powers 1972). For example, in laboratory chromatography columns, the incoming displacing fluid displaces the resident fluid. A practical example is the leaching of salts from a soil, because the added irrigation water mixes with and displaces the concentrated soil solution through the vadose zone. Movement of water containing dissolved fertilizer and herbicides into and through a soil profile is another example of miscible displacement. Miscible displacement studies were initially motivated for in-depth understanding of oil reservoir recovery issues (e.g., Tan and Homsy 1986; Chang and Slattery 1986; Yortsos 1987; Tan and Homsy 1988; Yortsos and Zeybek 1988; Zimmerman and Homsy 1992).

In laboratory columns, the phenomenon of VF is the flow instability resulting from a less viscous solvent displacing a more viscous solvent (Broyles et al. 1998) during vertical flow through porous media. Similarly, when another liquid of different density displaces the liquid residing in a horizontal bed of porous material, the resulting hydrodynamic dispersion is modified by the formation of a tongue of denser liquid undershooting the less dense liquid, a phenomenon known as gravity segregation (GS; Rose and Passioura 1971a). The phenomenon of VF and GS occur in shape of preferential flow of viscous liquids: that is, from oil spill or liquid fertigation in agricultural fields (Bowman and Rice 1986; Jaynes et al. 1988; Abbasi et al. 2003) and during the intrusion of saline groundwater into coastal aquifers (Dogan and Fares 2008), respectively.

It is important to achieve uniform sample concentration and flow velocity distributions across the head of a chromatography column for accurate analysis of solute plume movement. Shalliker et al. (Shalliker et al. 1999) studied the band profile of solute migrating along a high-performance liquid chromatographic (HPLC) column using an on-column visualization technique and reported that, except where the VF is prevalent, the design of the inlet header has little influence on the outcome of the viscous fingers. They argued that the flow heterogeneities do not arise only because of the design of the chromatography column, but the differences between the solute and the mobile phase viscosities can also affect the migration of the solute bands. Catchpoole et al. (Catchpoole et al. 2006) investigated the VF phenomenon within chromatography columns to quantify the VF phenomenon during small viscosity contrast between solute and mobile phase. They observed VF phenomenon by packing a glass column with particles that have the same refractive index as the mobile phase and injecting plugs of dye solutions that have viscosities different from that of the mobile phase. They reported the severe fingering effects if the viscosity
difference exceeds 0.17 cP. For such conditions, they suggested that the chromatographers should consider the consequences of the viscosity contrast between the sample and the mobile phase on chromatographic separations.

Studies on GS have been the interest and topic of research for petroleum engineers (e.g., Muskat 1937; Craig et al. 1957; Crane et al. 1963; Gardner et al. 1962), but their concern has been with the immiscible displacement of one fluid by another and this can be analyzed by relatively simple theories (e.g., Gardner et al. 1962). With miscible displacement, however, as occurs with aqueous solutions, the resulting interactions between hydrodynamic dispersion at the displacing front and gravity segregation are complicated. Can models predict VF and GS?

Philip (Philip 1991) argued that simulation modeling has largely supplanted laboratory experimentation and field observation in soil science and hydrology. He emphasized the need for more critical experimentation on important natural phenomena and less simulation modeling. The importance of such work is likely to grow with time, given the increasing extent of global salinity (Ghassemi et al. 1995), increasing human development of coastal regions worldwide, and extensive use of fertilizers in sandy and clay loams. The objective of this study was to explore VF and GS phenomena by conducting laboratory column experiments on the vertical and horizontal transport of ionic situations of different concentrations and densities through inert and reactive porous media.

2. Materials and methods

Solid glass spheres were used to construct inert media and the reactive material was sepiolite (from Vallecals, Spain) supplied as porous aggregates (Berk Mineral Products, Worksop, United Kingdom). This sepiolite has a cation exchange capacity of 0.26 mol c kg\(^{-1}\) and a total (internal plus external) surface area of 330 m\(^2\) g\(^{-1}\), giving a density of surface charge of 0.79 \(\mu\)mol c m\(^{-2}\) (Robertson 1957). The aggregates retain their geometry and physical structure as the concentration of the saturating solutions changes (Robertson 1957). Sieved 1.7–2.0-mm fractions of these materials were packed uniformly (Table 1) into two XK50 gel-filtration columns (Amersham Pharmacia Biotech, Little Chalfont, United Kingdom) with a diameter of 50 mm, mimicking sandy and clay loams. The pistons (flow adaptors) of these columns are designed to provide uniform flow at the entrance and exit of the bed of porous material and a negligible dead volume that is less than 1% of the total column volume. The columns were saturated under vacuum with deionized water and remained saturated with liquid throughout the experiments.

The solutions used were potassium bromide (KBr) of various concentrations. The electrical conductivity (EC) of the leachates was measured with a standard conductivity cell and a 4320 conductivity meter (Jenway Ltd., Felsted, United Kingdom). The concentrations of bromide (Br\(^{-}\)) and potassium (K\(^{+}\)) were measured using calibrated ion-specific electrodes. The electrodes and conductivity cell were calibrated frequently using four or five standard solutions over the range of concentration used in these displacement experiments.

In these experiments, KBr solution displaced deionized water resident in a column of porous material and vice versa. Pore-water velocities, ranging from 1.13 to 178 cm h\(^{-1}\), were used through the ballotini and between 0.98 and 64 cm h\(^{-1}\) through the sepiolite. The maximum velocity through the sepiolite was below the
limiting velocity (Passioura and Rose 1971) above which the convection–diffusion equation (CDE) fails to apply to an aggregated material. The flows were controlled at the lower velocities by a cartridge pump (Model 7553–85, Cole Parmer Instruments, Chicago, Illinois) and at the higher velocities by a standard peristaltic pump (type MHRE, Watson-Marlow Bredel Pumps, Falmouth, United Kingdom). Some experiments involved potassium permanganate (KMnO₄) solution displacing water; the purple color of the KMnO₄ enabled us to visualize the miscible segregation and record it by video and still cameras.

Column effluent was collected using a fraction collector (type FC203B, Gilson, Middleton, Wisconsin) modified to accept volumes of up to 80 cm³ per aliquot. Miscible or immiscible displacements were continued until breakthrough was completed or four pore volumes of liquid had been collected, whichever was earlier. All experiments and chemical analyses were done at a temperature of 20 ± 1°C.

For data analyses, we assumed that the simultaneous flow of water and solutes through a porous material during miscible displacement accompanied by VF and GS is described by CDE and the equation of van Genuchten and Wierenga (van Genuchten and Wierenga 1986),

$$ R \frac{\partial c}{\partial t} = K \frac{\partial^2 c}{\partial x^2} - v \frac{\partial c}{\partial x}. $$

Here, c is concentration of solute, t is time, v is pore-water velocity, x is linear distance, K is coefficient of hydrodynamic dispersion, and R is retardation factor.

Experimental breakthrough curves (BTCs) were produced for the electrical conductivity of the leachate and/or for the concentrations of the individual ions for each displacement. These breakthrough data were then analyzed using the conventional CDE model in CXTFIT 2.0 (Toride et al. 1995) for the time course of the flux concentration of effluent at the exit of the column of porous material (x = L) following an initial step change in concentration and therefore density at the entrance of the column (x = 0). These analytical solutions are case A-1 of van Genuchten and Wierenga (van Genuchten and Wierenga 1986). The value of the pore-water velocity, v = q/θ, was specified as that delivered by the calibrated pump to control flow, where q is the Darcy velocity. However, the optimized values of R and K are apparent or effective rather than real when describing horizontal displacements.

### 3. Results and discussion

Stages of miscible displacement influenced by VF and GS are shown in the photographs of 0.08-M potassium permanganate solution displacing distilled water during upward vertical flow (Figure 1), during downward vertical flow (Figure 2),

Table 1. Properties of the experimental columns: L is length (m), θₘ is porosity in micropores (m³ m⁻³), θₐ is porosity in aggregates (m³ m⁻³), and θₜ is total porosity (θₘ + θₐ).

<table>
<thead>
<tr>
<th>Material</th>
<th>L</th>
<th>θₘ</th>
<th>θₐ</th>
<th>θₜ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sepiolite</td>
<td>0.322</td>
<td>0.400</td>
<td>0.396</td>
<td>0.796</td>
</tr>
<tr>
<td>Ballotini</td>
<td>0.318</td>
<td>0.381</td>
<td>—</td>
<td>0.381</td>
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</table>
and in a horizontal column of ballotini when deionized water is displacing the denser resident liquid of KMnO₄ (Figure 3) and denser liquid of KMnO₄ is displacing the deionized water (Figure 4) were recorded. These photographs clearly demonstrate (i) the fingering phenomenon, (ii) the development of the tongue of denser KMnO₄ solution undershooting the less dense water, (iii) the extended plane interface between the liquids, and (iv) the progressive blurring of the interface by hydrodynamic dispersion as the displacements occur.

To investigate the effect of orientation of flow on the behavior of solutes, we made 11 displacements of KBr vertically through sepiolite with the same range of velocities and concentrations as in the horizontal displacements. Change of orientation from vertical to horizontal extends the length of the interface between resident and displacing liquids so that one might expect enhanced effective dispersion during horizontal flow compared to vertical flow.

The BTCs for vertical flow became steeper as the pore-water velocity ν fell and the Peclet number $P$ increased: Figure 5 compares representative observed and fitted BTCs for the vertical and horizontal displacements at the two extreme speeds of
displacement we used. At the lowest speed during horizontal flow, gravity segre-
gation dominates and results in a dispersed BTC because of greater mixing due to
the shallow gradient of the tilted interface between the two liquids; during vertical
displacement, the BTC is steep because of little hydrodynamic dispersion about the
stable interface between the liquids. The situation is reversed at the highest speed
because less dispersion occurs during horizontal flow than during vertical flow due to
less mixing by segregation about the steep gradient of the interface.

A total of 11 horizontal displacements were conducted with density differences
increasing from $0.81 \times 10^{-4}$ to $67.8 \times 10^{-4}$ g cm$^{-3}$ at a fixed pore-water velocity
of 23.3 cm h$^{-1}$ through ballotini. Figure 6 shows representative experimental data
and fitted BTCs; fitting was excellent, with the average coefficient of determination
being $0.998 \pm 0.002$ for the 11 displacements. These BTCs behaved as expected
(van Genuchten and Wierenga 1986) for miscible displacement in a finite column
and did not pass through $C_e = 0.5$ at $T = 1$. The retardation factor remained
constant at $1.001 \pm 0.004$, but $K$ increased from 1.06 to 20.5 cm$^2$ h$^{-1}$ and $P$ fell
from 696 to 36 as $\beta$ increased from 0.013 to 1.20 as a consequence of increasing
density difference. The values of $P$ were calculated as $P = \nu L/K$.

We also conducted 33 displacements through ballotini (11 speeds $\times$ 3 density
differences, corresponding to KBr concentrations of 0.08, 0.04, and 0.001 M) so
that $\beta$ ranged between 0.0018 and 24.7; the EC of the leachate was measured. There were 11 displacements through sepiolite (7 with 0.04 M and 4 with 0.02 M KBr) with $\beta$ ranging between 0.0614 and 8.06; the EC and the concentrations of Br$^-$ and K$^+$ in the leachate were measured. For both materials, the BTCs became steeper as the speed of displacement increased and the $P$ decreased in contrast to the behavior in stable, vertical flow in which BTCs generally become steeper as speed of displacement decreases (discussed in the next section), indicating that segregation dominated the displacements. The BTCs were well fitted by CXTFIT 2.0, with the average coefficients of determination being 0.996 (ballotini, $n = 33$) and 0.997 (sepiolite, $n = 33$). Apparent retardation factors $R$ were close to unity for the ballotini, with a small decrease as concentration decreased (Table 2). For sepiolite, $R$ was always less than unity for Br$^-$, greater than unity for K$^+$ and close to but less than unity for EC, and became more extreme as concentration decreased.

These vertical displacements were better fitted by CXTFIT 2.0, with the average coefficient of determination rising to 0.9982 ($n = 33$). The real retardation factors (Table 2) did not differ from those found in horizontal displacements, but the real dispersion coefficients differed markedly: $K$ increased in a quadratic fashion as $K = a + bv + cv^2$ (Passioura and Rose 1971; Rose et al. 2006) and did not differ between concentration ranges. Complete mathematical analysis of the combined effect of VF and GS is not only complex but also difficult to fit to experimental data, though approximate methods of analysis have met with some success (e.g., Chhatwal et al. 1973; Wilson and da Costa 1982). Rose and Abbas (Rose and
Figure 4. A horizontal displacement in which the denser liquid of KMnO$_4$ (0.08 M) is displacing deionized water at 23.3 cm h$^{-1}$. The formation of tongue of denser liquid undershooting the less dense liquid during uptake (an example of gravity segregation) is illustrated. Reprinted with permission from Rose and Abbas (Rose and Abbas 2007).
Abbas 2007) showed that it is possible to use CDE to describe and CXTFIT 2.0 (Toride et al. 1995) to analyze the net effect of these two disparate processes in terms of “effective” coefficients of hydrodynamic dispersion and effective retardation factors for a wide range of pore-water velocities, particle and column sizes, and density differences. This is a substantial advance on the work of Rose and Passioura (Rose and Passioura 1971a), who found that the lognormal transformation of a breakthrough curve (Rose and Passioura 1971b) could only be applied to displacements under small differences in concentration and therefore density.

As described earlier, during vertical miscible displacements, the displacing fluid mixes with and displaces the resident fluid. During vertical flow, an accelerated diffusion known as hydrodynamic dispersion around a stable interface between the resident and displacing liquids results (Figure 7a). In contrast, when the resident liquid in a horizontal porous bed is displaced by a liquid of a greater density, the resultant hydrodynamic dispersion is usually further enhanced by the formation of tongue of the denser liquid undershooting the less dense liquid (Figure 7b). This phenomenon, which is known as gravity segregation (Rose and Passioura 1971b), results in an extended plane interface between liquids, about which mixing occurs.

4. Conclusions

This paper presents new experimental results on the vertical and horizontal transport of miscible ionic solutions of different concentrations and densities through inert and reactive porous media. In addition to the phenomenon of viscous
fingering, we have explored in detail some consequences of gravity segregation during miscible displacement, a phenomenon that occurs when saline groundwater intrudes into coastal aquifers, an increasingly common problem worldwide. We found that the apparent coefficients of hydrodynamic dispersion vary much less with pore-water velocity in horizontal than in vertical flow, but retardation factors are not influenced by the orientation of flow. We observed that (i) hydrodynamic dispersion has a mitigating effect on segregation so that the mixed zone is smaller than predicted for immiscible displacement, (ii) breakthrough curves may be analyzed with CXTFIT 2.0 with confidence, and (iii) segregation substantially modifies the effect of pore-water velocity on dispersion. As a

<table>
<thead>
<tr>
<th>Materials for porous media</th>
<th>Solution/ion</th>
<th>C (M)</th>
<th>Retardation factor</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Vertical</td>
<td>Horizontal</td>
</tr>
<tr>
<td>Sepiolite</td>
<td>KBr bulk solution</td>
<td>0–0.04</td>
<td>0.991</td>
<td>0.991</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0–0.02</td>
<td>0.988</td>
<td>0.988</td>
</tr>
<tr>
<td></td>
<td>Potassium ion</td>
<td>0–0.04</td>
<td>1.033</td>
<td>1.034</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0–0.02</td>
<td>1.046</td>
<td>1.048</td>
</tr>
<tr>
<td></td>
<td>Bromide ion</td>
<td>0–0.04</td>
<td>0.973</td>
<td>0.974</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0–0.02</td>
<td>0.953</td>
<td>0.954</td>
</tr>
<tr>
<td>Ballotini</td>
<td>KBr bulk solution</td>
<td>0–0.08</td>
<td>—</td>
<td>1.006</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0–0.04</td>
<td>—</td>
<td>1.002</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0–0.001</td>
<td>—</td>
<td>0.993</td>
</tr>
</tbody>
</table>

Figure 6. Representative experimental data and fitted BTCs showing the effect of gravity segregation number $\beta$ on breakthrough of KBr solution through ballotini at $v = 23.3$ cm h$^{-1}$. Shown are fitted BTCs (solid lines), $\beta = 0.014$ (squares), $\beta = 0.299$ (triangles), and $\beta = 1.198$ (circles). Reprinted with permission from Rose and Abbas (Rose and Abbas 2007).
consequence of (i) above, use of the simple theory for immiscible displacement will provide a safety margin when predicting the extent and timing of miscible segregation. These experimental findings have practical implications on the understanding of the preferential flow of viscous liquids such as liquid fertigation in agricultural fields and the intrusion of saline water into the freshwater of coastal aquifers.

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References


