Effects of Finger-Growth Velocity on Reactive Miscible Viscous Fingering

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This article is our additional research on reactive miscible viscous fingering previously published (Nagatsu and Ueda, 2001). In our previous article, by experiment and theory, the effects were studied of reactant concentrations on reactive miscible viscous fingering in which chemical reaction is instantaneous and passive for flow motion, when finger-growth velocity is low enough to neglect the convective transport effect where chemical species are transported primarily by molecular diffusion. Two types of reaction patterns were shown depending on the dimensionless parameter ϕ_v (ϕ_v is an initial reactant concentration ratio normalized by a stoichiometric ratio of a chemical reaction.) When $\phi_v \ll 1$, the product spreads in a relatively broad area inside fingers. On the other hand, when $\phi_v \gg 1$, the product concentrates at the fingertips. The flow of recent publications in this field (Jahoda and Hornof, 2000; Böckmann and Müller, 2000; De-Wit, 2001; Fujita et al., 2000) shows its continuing importance. The objective of this article is, in turn, to experimentally elucidate the effects of finger-growth velocity on the relationship between fingering and reaction patterns.

Experimental Apparatus and Procedure

The experimental apparatus and procedure, the liquids, and the chemical reaction used in the present study are the same as those previously reported. The gapwidth of the cell b is constantly set as $b = 0.24$ mm as well. The present experiments have been conducted by varying the bulk finger-growth velocity. The bulk finger-growth velocity is defined as a characteristic velocity of the fingering U. It is defined as

$$U = \frac{q}{2\pi R b} \tag{1}$$

while the Peclet number Pe_v is defined as follows

$$Pe_v = \frac{LU}{D_v} = \frac{R\dfrac{q}{2\pi R b}}{D_v} = \frac{q}{2\pi b D_v} \tag{2}$$

Here, the notations q, R, L, and D_v and the value of D_v are the same as those in our previous article. Thus, the variation in the bulk finger-growth velocity corresponds to the variation in Pe_v and is proportional to q because of constant b. Pe_v indicates the ratio between the transport rate by convection and that by diffusion on the mixing of glycerin and water. As mentioned earlier, two reaction patterns are observed depending on ϕ_v when the bulk finger-growth velocity is low enough to neglect a convective effect. Thus, the present experiments have been conducted under two different reactant concentrations: $\phi_v = 0.1$ ($c_{m0} = 0.2$ mol/L, $c_{l0} = 0.01$ mol/L) and $\phi_v = 10$ ($c_{m0} = 0.04$ mol/L, $c_{l0} = 0.2$ mol/L). Experiments with nonreactive liquids have been performed as well in order to provide a reference for the fingering patterns of the reaction cases.

Results and Discussion

The effects of Pe_v on miscible viscous fingering pattern without chemical reaction are shown in Figures 1a and 1b. Figures 1a and 1b are the cases of $Pe_v = Pe_{v0} = 3.9\times10^3$ ($q = q_0 = 0.94$ mm^3/s) at $t = 480$ s and $Pe_v = 8Pe_{v0}$ ($q = 8q_0$) at $t = 60$ s, respectively, where the total volume of the injected and less viscous liquid is set to be the same. A quarter of the image is shown in order to recognize the fingering pattern more clearly. The lower Pe_v is the same as that in our previous article. In both cases, a well-defined interface is formed between two miscible liquids since Pe_v is essentially large (Nagatsu and Ueda, 2001). These figures show that as the bulk finger-growth velocity is increased, the fingertip-splitting is enhanced and the finger width becomes thinner. Tan and Homsy showed that the stretching of the fingertips associated with the straining flow near the fingertips plays an important role for a finger-splitting mechanism in miscible viscous fingering (Tan and Homsy, 1988). When the less-viscous liquid reaches the tip of the finger, it encounters very high viscous liquid and consequently turns sideways because of its incompressibility. Therefore, a stagnated flow is formed around the fingertips, resulting in the fingertips being subject to the stretching. When the fingertip stretch is increased, the fingertip splitting is enhanced and the finger width becomes thinner. It shows that the fingertips in Figure 1b are exposed to

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more stretching than those in Figure 1a. This result indicates that the velocity of the approaching flow to the fingertips in the case shown in Figure 1b is higher than that in the case shown in Figure 1a.

The depth of the blood-red color is changed around the fingertip, as shown in both Figures 1a and 1b, which indicates that the thickness of the less viscous-liquid layer in the depth direction in the cell is varied around the fingertips; in other words, a three-dimensional (3-D) structure appears (Lajeunesse et al., 1999, 2001). At $Pe_v = Pe_{v0}$, the configuration of the interface in the cell's depth direction can be drawn, as shown in Figure 1c, which is supposed to correspond to a preceding foot configuration (Lajeunesse et al., 1999), since the depth of the blood-red color suddenly lightens at the fingertips, as shown in Figure 1a. At $Pe_v = 8Pe_{v0}$, the configuration can be drawn, as shown in Figure 1d, which is supposed to correspond to a tongue-like shape (Lajeunesse et al., 2001), since the depth of the blood-red color is almost uniform around the fingertips and the color depth around the fingertips is lighter than that around the finger base, as shown in Figure 1b. These results indicate that the bulk finger-growth velocity influences the configuration of the interface in the cell's depth direction around the fingertips as well.

The effects of Pe_v on the chemical reaction pattern in reactive miscible viscous fingering under the conditions of $\phi_v =$ 0.1 and 10 are exhibited in Figure 2. In these figures, the blood-red region indicates where the product exists and the depth of the color corresponds to the quantity of the product. Figures 2a and 2b show the cases at $Pe_v = Pe_{v0}$ ($t = 480$ s) and $Pe_v = 8Pe_{v0}$ ($t = 60$ s), respectively, under the condition of $\phi_v = 0.1$. At $Pe_v = Pe_{v0}$, as reported in our previous article, the product spreads in a relatively broad area inside the fingers, since the reaction zone is located far from the interface between two liquids in the less viscous liquid, and the product can widely diffuse in the less viscous liquid. At $Pe_v =$ $8Pe_{v0}$, the chemical reaction pattern varies significantly as compared to the case of $Pe_v = Pe_{v0}$. In this case, the product exists remarkably around the fingertips. This is because the reaction zone shifts toward the interface in the less viscous liquid and the product is difficult to diffuse in the less viscous liquid compared to the case of $Pe_v = Pe_{v0}$ due to the convective effect mentioned earlier, that is, the approaching flow velocity in the less viscous liquid toward the interface is higher. In other words, the convective effect causes the product to exist significantly around the fingertips. Figures 2c and 2d show the cases at $Pe_v = Pe_{v0}$ ($t = 480$ s) and $Pe_v = 8Pe_{v0}$ ($t = 60$ s), respectively, under the condition of $\phi_v = 10$. At $Pe_v = Pe_{v0}$, as reported in our previous article, the product concentrates at the fingertips, since the reaction zone is located close to the interface in the more viscous liquid, and the product diffuses only negligibly in the more viscous liquid. At $Pe_v = 8Pe_{v0}$, the reaction pattern changes little as compared to the case of $Pe_v = Pe_{v0}$, where the product significantly exists around the fingertips. This is because the re-

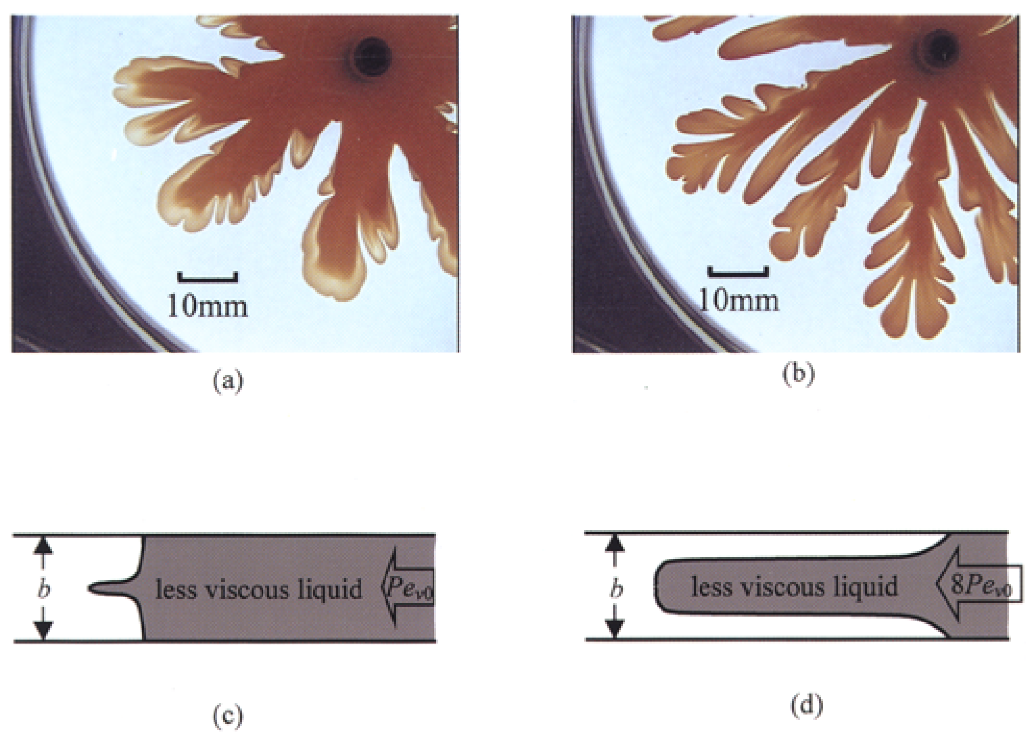

Figure 1. Miscible viscous fingering patterns without chemical reaction and configuration of the interface in the cell's depth direction around the fingertips in miscible viscous fingering.

(a) Fingering pattern at $Pe_v = Pe_{v0}$ ($t = 480$ s); (b) fingering pattern at $Pe_v = 8Pe_{v0}$ ($t = 60$ s); (c) configuration of the interface in the cell's depth direction around the fingertips at $Pe_v = Pe_{v0}$; (d) configuration of the interface in the cell's depth direction around the fingertips at $Pe_v = 8Pe_{v0}$.

action zone is hardly shifted in the more viscous liquid even in the case of $Pe_v = 8Pe_{v0}$, as compared to the case of $Pe_v = Pe_{v0}$; thus, the product diffuses only negligibly in the more viscous liquid as similar to the case of $Pe_v = Pe_{v0}$. This is caused by the viscosity of the more viscous liquid being very large, indicating that the diffusivity in the more viscous liquid is very small, which results in the diffusive zone being limited in a thin region close to the interface. These results show that as the bulk finger-growth velocity is increased, the effects of ϕ_v on the reaction pattern of the fingering becomes insignificant, and the product exists significantly around the fingertips regardless of the initial reactant concentrations in both liquids. In addition, it should be noticed that the fingering pattern with reaction and without reaction for each Pe_v condition are similar, indicating that the reaction can be treated as passive.

The effects of the variation in the 3-D structure around the fingertips produced by an increase in the finger-growth velocity on the reaction pattern should be taken into account. The configuration of the interface and that of the reaction zone in the cell's depth direction is shown in Figure 3, along with the region in which the product exists significantly. The top views of the fingering and reaction patterns are illustrated as well. Under the condition of $\phi_v = 0.1$, at $Pe_v = Pe_{v0}$, the reaction zone is formed two-dimensionally and the product spreads widely in the less viscous liquid (Figure 3a), since the reaction zone is located far from the interface and the product can widely diffuse in the less viscous liquid, as briefly mentioned in our previous article. In contrast, at $Pe_v = 8Pe_{v0}$, the reaction zone is located along the tongue-like-shaped interface in the less viscous liquid and a region where the product exists significantly is established along the interface (Figure 3b). This is because the convective effect causes the reaction zone to approach the interface as compared to the case of $Pe_v = Pe_{v0}$ and makes it difficult for the product to spread in the less viscous liquid. Under the condition of $\phi_v = 10$ at $Pe_v = Pe_{v0}$, the reaction zone is located along the preceding foot shaped interface and the product concentrates along the in-

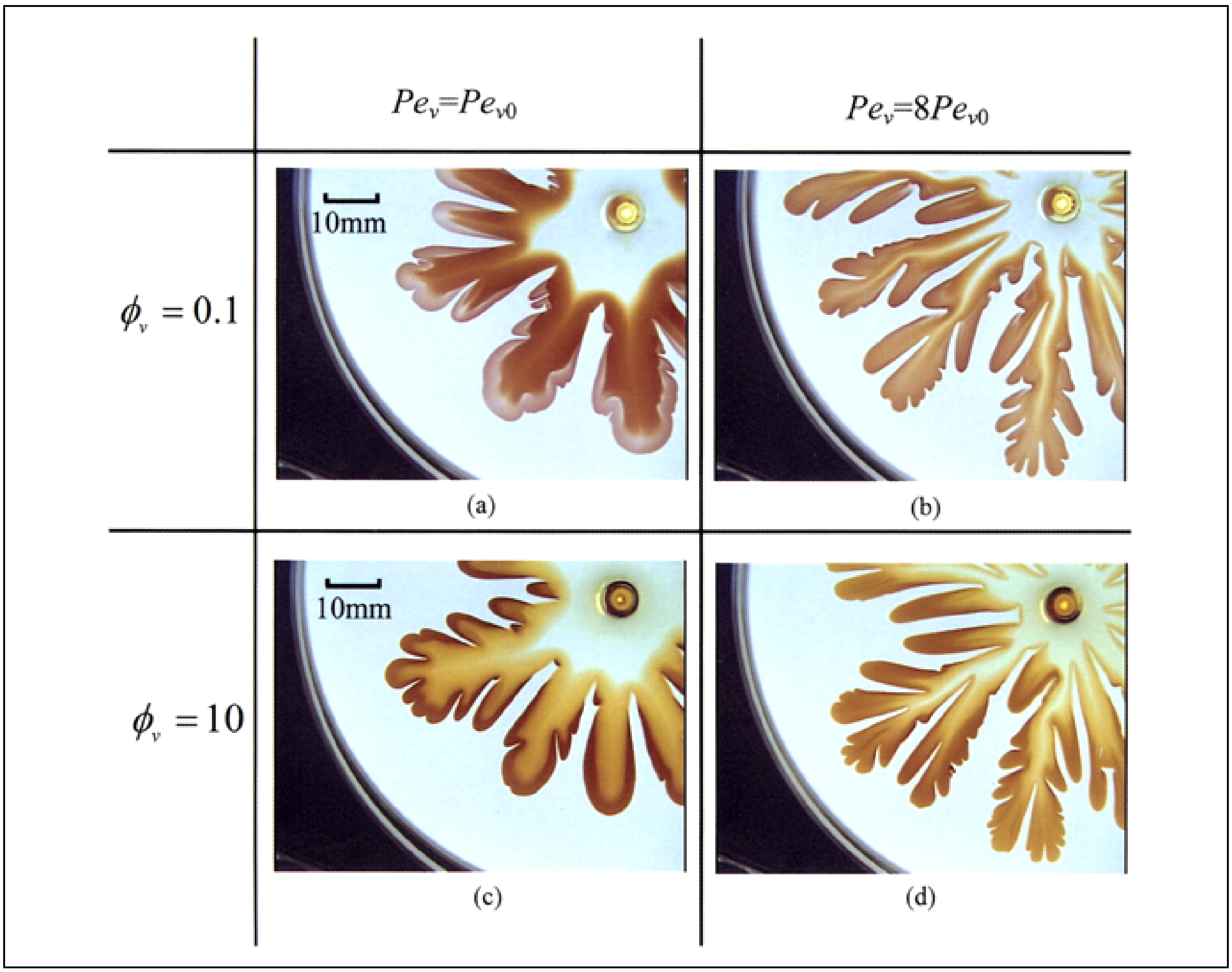

Figure 2. Miscible viscous fingering patterns with chemical reaction.

(a) $\phi_v = 0.1$ and $Pe_v = Pe_{v0}$ ($t = 480$ s); (b) $\phi_v = 0.1$ and $Pe_v = 8Pe_{v0}$ ($t = 60$ s); (c) $\phi_v = 10$ and $Pe_v = Pe_{v0}$ ($t = 480$ s); (d) $\phi_v = 10$ and $Pe_v = 8Pe_{v0}$ ($t = 60$ s).

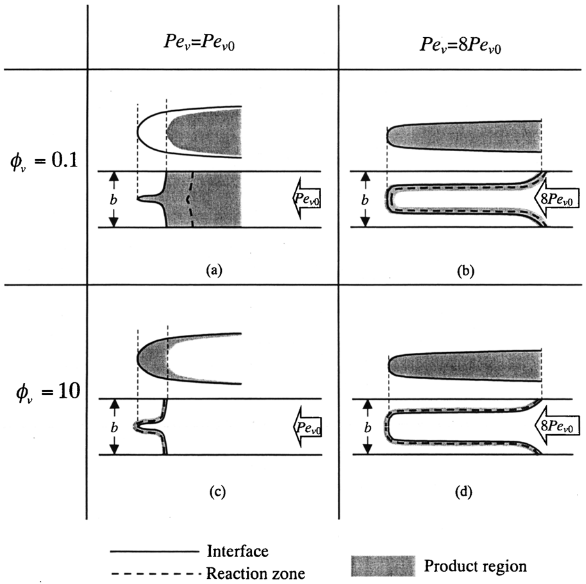

Figure 3. Interface, reaction zone, and product region in the cell's depth direction and top views of reaction pattern around the fingertip in reactive miscible viscous fingering.

(a) $\phi_v = 0.1$ and $Pe_v = Pe_{v0}$; (b) $\phi_v = 0.1$ and $Pe_v = 8Pe_{v0}$; (c) $\phi_v = 10$ and $Pe_v = Pe_{v0}$; (d) $\phi_v = 10$ and $Pe_v = 8Pe_{v0}$.

terface (Figure 3c), because the reaction zone is located close to the interface in the more viscous liquid and the product diffuses only negligibly in the more viscous liquid, as briefly mentioned in our previous article. At $Pe_v = 8Pe_{v0}$, as similar to the case of $Pe_v = Pe_{v0}$, the reaction zone is located along the tongue-like-shaped interface in the more viscous liquid, and the thin region where the product exists is formed along the interface, because the reaction zone is located close to the interface in the more viscous liquid, where the product scarcely diffuses. As a result, at $Pe_v = Pe_{v0}$, as shown in Figures 3a and 3c, the difference in the 3-D structure advances the apparent difference in the reaction pattern depending on ϕ_v. On the other hand, the locations of the reaction zone and the region where the product exists significantly are similar in both cases, $\phi_v = 0.1$ and $\phi_v = 10$, at $Pe_v = 8Pe_{v0}$, as described in Figures 3b and 3d. This similar 3-D structure at $Pe_v = 8Pe_{v0}$ indicates that the difference in the reaction pattern due to ϕ_v is less important as compared to the case of $Pe_v = Pe_{v0}$. Consequently, the variation in the 3-D structure due to an increase in the bulk finger-growth velocity also diminishes the effects of ϕ_v on the reaction pattern.

Conclusion

The effects of finger-growth velocity on the characteristics of reactive miscible viscous fingering were investigated using a Hele-Shaw cell. We have experimentally shown that the bulk finger-growth velocity affects not only the fingering pattern, but also the reaction pattern. When the bulk finger-growth velocity is increased, the effect of ϕ_v on the reaction pattern becomes insignificant, and the product exists significantly around the fingertips regardless of ϕ_v. In addition, the change in the 3-D structure around the fingertips generated by an increase in the finger-growth velocity also reduces the effect of ϕ_v on the reaction patterns.

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