Investigation of Reacting Flow Fields in Miscible Viscous Fingering by a Novel Experimental Method

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The reacting flow fields in reactive miscible viscous fingering in a Hele-Shaw cell studied by Nagatsu and Ueda had not been completely elucidated, mainly because one cannot exactly recognize where and when the reaction takes place in the reactive fingering pattern. We developed a novel experimental method that allowed us to identify the reaction region in the fingering pattern employed in the previous studies. The novel method involves switching of the less-viscous liquid injected in both the nonreactive and reactive experiments. By using the novel method, we succeeded in showing how the reaction region in the fingering pattern was affected by the initial reactant concentrations, the Péclet number, and time. We propose physical models of the reacting flow field in the cell's gap direction that can explain the obtained experimental results.

Keywords: viscous fingering, miscible displacement, reacting flow, convection-diffusion-reaction, Hele-Shaw cell

Introduction

When a more-viscous fluid is displaced by a less-viscous fluid in porous media and in Hele-Shaw cells, the interface or boundary of the two fluids becomes unstable and forms a finger-like pattern. This phenomenon is referred to as viscous fingering. Since the appearance of the pioneering works on the fluid mechanics of viscous fingering published in the 1950s,[1,2] many experimental and theoretical studies have been performed and some review articles have been published.[3–5] There are two classes of viscous fingering: fingers formed in immiscible systems and those formed in miscible systems. The dimensionless number that controls the fingering dynamics in immiscible systems is the capillary number, *Ca*, which is defined as the ratio between viscous and interface-tension forces. In miscible systems, the dimensionless number is the Péclet number, *Pe*, which is defined as the ratio between convective and diffusive transport rates of mass. For both systems, nonlinear propagation of viscous fingering is governed by different mechanisms of shielding spreading and splitting. Shielding is the phenomenon in which a finger slightly ahead of its neighboring fingers quickly outruns them and shields them from further growth. Spreading and splitting are the phenomena in which a finger that spreads until it reaches a certain width becomes unstable and splits.[3]

Viscous fingering accompanied by chemical reactions is observed in processes such as petroleum recovery,[6] chromatographic and adsorptive separation,[7] polymerization,[8] and the flow of gastric mucus[9] and is confirmed as playing an important role in these processes. Therefore, the coupling between hydrodynamics and chemistry in viscous fingering with chemical reactions has been studied. Jahoda and Hornof[10] conducted a numerical investigation of concentration fields in an immiscible viscous finger involving a chemical

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reaction. They showed that the reactant concentration in the less-viscous liquid decreased in the fingertip area as a result of the reaction at the interface. Fernandez and Homsy[11] performed experiments on immiscible viscous fingering with a chemical reaction acting to reduce interfacial tension in a Hele-Shaw cell and found that the reaction made the fingers wider. They characterized the effects of the reaction on the reactive fingering pattern using the Damköhler number, *Da*, which is defined as the ratio between a characteristic time of fluid motion and that of a chemical reaction. DeWit and Homsy[12,13] performed a numerical simulation on reactive miscible viscous fingering in porous media by assuming that the fluid's viscosity is given as a function of a chemical species concentration and by using a specific chemical kinetics. They found a new mechanism of viscous fingering that they call the 'droplet' mechanism, which involves the formation of isolated regions of either less- or more-viscous fluids in connected domains of the other. Nagatsu et al.[14] performed experiments on miscible viscous fingering with instantaneous chemical reactions that increase or decrease the viscosity of the more-viscous fluids in a Hele-Shaw cell. They found that the shielding effect was suppressed and the fingers were widened when the viscosity of the more-viscous fluid was increased by the reaction. In contrast, the shielding effect was enhanced and the fingers were narrowed when the viscosity of the more-viscous fluid was decreased by the reaction.

Nagatsu and Ueda[15] performed experiments on reactive miscible viscous fingering in a Hele-Shaw cell. They used 99 wt % glycerin solutions that included potassium thiocyanate (KSCN) and iron nitrate ($Fe(NO_3)_3$) solutions as the more- and less-viscous liquids, respectively. The instantaneous chemical reaction between Fe^{3+} and SCN^- resulted in a blood-red-colored product. This reaction had no influence on the hydrodynamics of the fingering. The product distribution was highly dependent on the ratio between the reactant concentrations initially included in the more- and less-viscous liquids normalized by a stoichiometric ratio of the chemical reaction, φ_v, which was expressed as follows:

$$\varphi_v = \frac{ac_{l0}}{c_{m0}}. \tag{1}$$

In this equation, c_{m0} and c_{l0} were the molar reactant concentrations initially included in the more- and less-viscous liquids, respectively, and a was the molar stoichiometric ratio of the chemical reaction. For $\varphi_v \ll 1$, the product was present in large quantities in a relatively broad area within the interior of the fingers, whereas for $\varphi_v \gg 1$, it concentrated around the tip of the fingers. For $\varphi_v \sim 1$, the product was equally distributed among the interior and tip of the fingers. Nagatsu and Ueda[15] also conducted a theoretical analysis on a concentration field of chemical species in two miscible liquids with different viscosities using a simplified one-dimensional diffusion-reaction model, in which a distinct interface was assumed for two miscible liquids. The analytical results revealed that the reaction plane was located in the less-viscous liquid far from the interface between the two liquids for $\varphi_v \ll 1$, but it was in the more-viscous liquid close to the interface for $\varphi_v \gg 1$. Therefore, they concluded that the product distribution was caused by differences in the location of the reaction plane due to φ_v. In another study, Nagatsu and Ueda[16] showed that the dependence of the product distribution on φ_v was diminished with an increase in the bulk finger growth velocity. In these cases, the product was present in a relatively broad area from the inside to the tip of the fingers, regardless of φ_v. To theoretically investigate the effects of the finger-growth velocity, Nagatsu and Ueda[17] conducted a convection-diffusion-reaction analysis where a distinct interface was assumed, and the analytical results showed that the dependence of the location of the reaction plane decreased with an increase in the convective effect. This result supported the experimental results of their earlier study.[16]

The reacting flow field in reactive miscible viscous fingering in a Hele-Shaw cell in Nagatsu and Ueda[15,16] has not yet been completely elucidated. Figure 1 shows time evolutions of the reactive miscible viscous fingering pattern for three different reactant concentration conditions under similar cell gap widths, b, and Péclet number, Pe_v, as those employed in Nagatsu and Ueda,[15] namely b = 0.3 mm and $Pe_v = 6.0 \times 10^3$, respectively. Here, the above-mentioned liquids and chemical reaction were used. Here, Pe_v is defined as follows:

$$Pe_v = \frac{UR}{D_v} = \frac{\frac{q}{2\pi Rb}R}{D_v} = \frac{q}{2\pi b D_v}, \tag{2}$$

where U is an increase rate of the radius of the circular pattern, R, when the less-viscous liquid completely displaces the more-viscous liquid, which means the layer of the more-viscous liquid in the gap width is absent as the pattern propagates, and the boundary remains circular. In this equation, q is the volumetric injection rate of the less-viscous liquid. D_v is the average value of the coefficient of diffusion between glycerin and water, which was measured by Petitjeans and Maxworthy,[18] and its value is $D_v = 1.6 \times 10^{-4}$ mm^2/s. The case of typical reactant concentrations in which the product is present in large quantities in a relatively broad area within the interior of the fingers is shown in (a), where c_{m0} = 0.3 mol/l and c_{l0} = 0.015 mol/l. The case of typical reactant concentrations in which the product is equally distributed among the interior and tip of the fingers is shown in (b), where c_{m0} = 0.06 mol/l and c_{l0} = 0.03 mol/l. The case of typical reactant concentrations in which the product it concentrated around the tip of the fingers is shown in (c), where c_{m0} = 0.03 mol/l and c_{l0} = 0.15 mol/l. The reaction between Fe^{3+} and SCN^- is expressed in Eq. 3:

$$Fe^{3+} + nSCN^- \rightarrow [Fe(SCN)_n]^{(3-n)^+} (\text{blood red}). \tag{3}$$

It is known that the development of the blood-red color is closely related to the formation of the $[Fe(SCN)_n]^{(3-n)+}$ (n = 1–6) complexes in aqueous solution, and has been interpreted in terms of the presence of the colored $[Fe(SCN)]^{2+}$ complex as the main species at low SCN^- concentration, the $[Fe(SCN)_2]^+$ one at about 0.1 mol/l SCN^- concentration, and $[Fe(SCN)_6]^{3-}$ at very high SCN^- concentration.[19] In the previous studies, we set n = 2, which meant a = 2. In the system employed here, the reactants being not premixed are provided separately into the reaction region. The above-mentioned reaction can be treated as an instantaneous one. Under

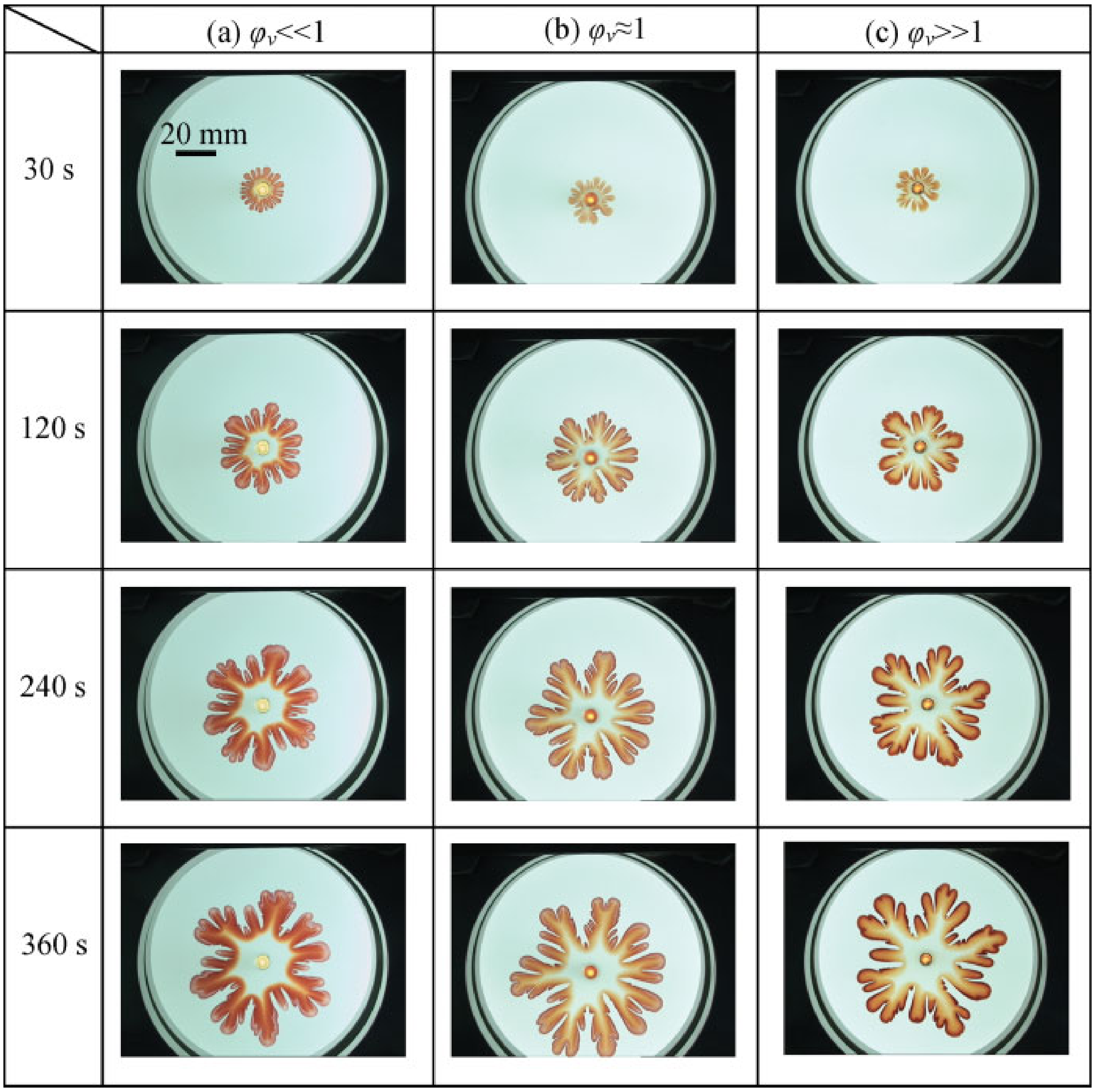

Figure 1. Time evolutions of reactive miscible viscous fingering pattern for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$ and $b = 0.3$ mm.

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the condition of instantaneous reaction, the reaction region becomes infinitesimal and thus it can be treated as reaction plane. At the reaction plane, the reactants are not present. Note that the ratio of flux of the reactants provided into the reaction plane becomes their stoichiometric ratio. On the basis of the consideration mentioned above, the concentration of SCN^- at the reaction plane is significantly low in the system employed here. This indicates that $n = 1$ ($a = 1$) can be more plausible in this system. Under reactant concentration condition shown in Figure 1a, $\varphi_v = 0.1$ when $a = 2$, whereas $\varphi_v = 0.05$ when $a = 1$. In Figure 1b, $\varphi_v = 1$ when $a = 2$, whereas $\varphi_v = 0.5$ when $a = 1$. In Figure 1c, $\varphi_v = 10$ when $a = 2$, whereas $\varphi_v = 5$ when $a = 1$. In the present study, for the reactant concentration conditions shown in Figures 1a–c, we denote $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$, respectively. In Nagatsu et al.,[20] the product concentration at the reaction plane, c_{pr}, was theoretically obtained as follows:

$$c_{pr} = \frac{c_{l0} c_{m0}}{a c_{l0} + c_{m0}} \tag{4}$$

In Figure 1, c_{l0} and c_{m0} were determined as c_{pr} was almost identical ($c_{pr} \approx 0.015$ mol/l) for each φ_v condition when $a = 2$. When $a = 1$, c_{pr} is higher in the order of the conditions of $\varphi_v \gg 1$, $\varphi_v \approx 1$, and $\varphi_v \ll 1$ (for $\varphi_v \gg 1$, $c_{pr} \approx 0.025$ mol/l; for $\varphi_v \approx 1$, $c_{pr} \approx 0.020$ mol/l; for $\varphi_v \ll 1$, $c_{pr} \approx 0.015$ mol/l).

It should be confirmed that the blood-red-colored region indicates that the product is present there, and the depth of the color corresponds to the quantity of the product in Figure 1. The difference in the product distribution by φ_v becomes significant after $t = 120$ s, although it is hardly observed until $t = 30$ s. After $t = 120$ s, the product is present in large quantities in a relatively broad area within the interior of the fingers at $\varphi_v \ll 1$, whereas it concentrates around the

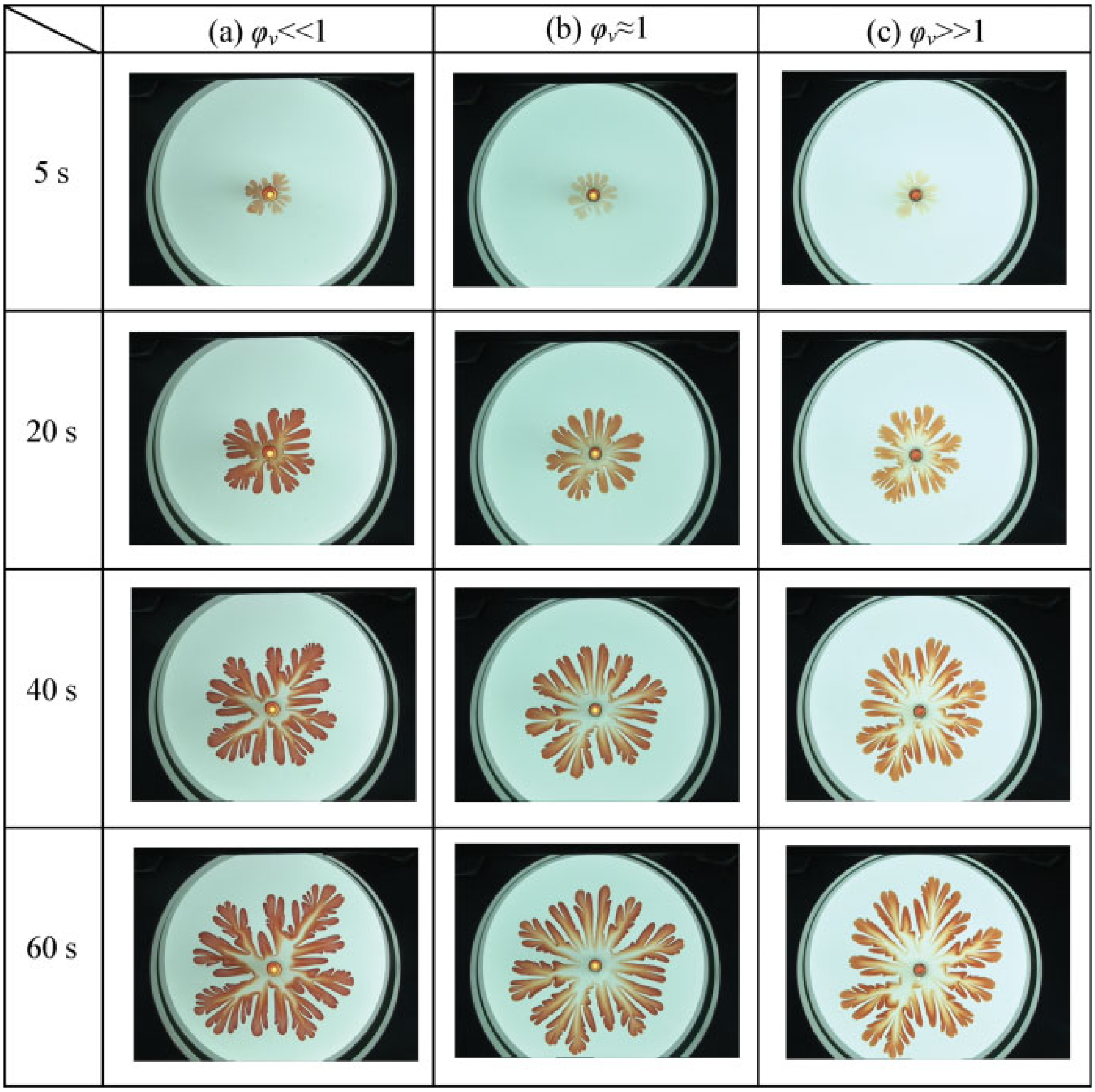

Figure 2. Time evolutions of reactive miscible viscous fingering pattern for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 3.6 \times 10^4$ and $b = 0.3$ mm.

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tip of the fingers at $\varphi_v \gg 1$. At $\varphi_v \approx 1$, the product is equally distributed among the interior and tip of the fingers. In this system, the region where the product exists does not necessarily coincide with the region where the reaction takes place, because the region where the product exists at a given time may indicate that a previously produced product remains, even though the reaction does not take place at that time. In addition, especially for $t > 120$ s, variations in the distribution and the depth of the blood-red-colored regions cannot be observed, although sizes of the fingering patterns are different. Therefore, we cannot recognize exactly where and when the reaction takes place in the fingering pattern, especially for $t > 120$ s. In addition, there are several points that have not been elucidated. For $t > 120$ s, the product is somewhat present in the fingertips at $\varphi_v \ll 1$, whereas the product is within the interior of the fingers at $\varphi_v \gg 1$. At the present we do not have a clear answer to the question of why the difference in the product concentration in the fingering pattern in each φ_v arises.

Figure 2 shows time evolutions of the reactive miscible viscous fingering pattern when $\varphi_v \ll 1$ ($c_{m0} = 0.3$ mol/l and $c_{l0} = 0.015$ mol/l), $\varphi_v \approx 1$ ($c_{m0} = 0.06$ mol/l and $c_{l0} = 0.03$ mol/l), and $\varphi_v \gg 1$ ($c_{m0} = 0.03$ mol/l and $c_{l0} = 0.15$ mol/l) under similar cell gap widths, b, and Péclet number, Pe_v, as those employed in Nagatsu and Ueda,[16] namely $b = 0.3$ mm and $Pe_v = 3.6 \times 10^4$. Also in this case, we cannot identify the reaction region in the fingering pattern, especially for $t > 20$ s, for the reason mentioned above, although we find that the dependence of φ_v on the product distribution is diminished. It is worth noting that a clear understanding about when and where the reaction takes place in the fingering pattern was not obtained in other reactive viscous fingering experiments performed by Fernandez and Homsy,[11] Nagatsu et al.,[14] or Hornof et al.[6,21,22]

In the present study, we have developed a novel experimental method that allows us to identify when and where the reaction takes place in the reactive fingering pattern employed in previous studies[15,16] to completely clarify the reacting flow field. We have investigated how the location of the reaction region in the fingering pattern is affected by φ_v, Pe_v, and time by applying the novel method under the experimental conditions described in Figures 1 and 2. Furthermore, discussion of the reacting flow field is made based on the identification of the reaction region.

Experimental Procedures

The experimental setup used in this study was the same as that previously reported,[15,16] except for the configuration of the syringe pump. The gap width of the cell was set to b = 0.3 mm, as mentioned in the Introduction. In the present study, a syringe pump carrying two syringes was employed for the novel method.

We performed nonreactive experiments and reactive experiments. For the nonreactive experiments, a 99 wt % glycerin solution was used as the more-viscous liquid. Two solutions dyed with different colors were used for the less-viscous liquid: blue 0.1 wt % indigo carmine (IC) solution and blood-red 0.025 mol/l iron thiocyanate solution. The blue IC solution is injected until $t = t_{sw}$. At $t = t_{sw}$, the injected liquid is switched, and after $t = t_{sw}$ the blood-red iron thiocyanate solution is injected. We focus our attention on the flow of the less-viscous liquid, injected second. From the difference in the blood-red areas at $t = t_1$ and $t = t_2$ ($>t_1$), we can determine to what extent the less-viscous liquid injected after t_{sw} reaches in the fingering pattern during the period of t_1–t_2.

For the reactive experiments, the more-viscous liquid was a colorless 99 wt % glycerin solution including KSCN at a concentration of c_{m0}. Two solutions were used as the less-viscous liquid. One was a blue 0.1 wt % IC solution that does not react with the more-viscous liquid, and the other was a $Fe(NO_3)_3$ solution at a concentration of c_{l0}. The second solution is light yellow but is essentially colorless in the Hele-Shaw cell because of its significantly thin gap, and it reacts with the more-viscous liquid. The chemical reaction is expressed as Eq. 1, and a blood-red product is again produced. As mentioned in the Introduction, this reaction can be regarded as instantaneous and does not influence the fluid dynamics of the fingering. First, the nonreactive blue IC solution is injected until $t = t_{sw}$. After $t = t_{sw}$ the reactive $Fe(NO_3)_3$ solution is injected. We pay attention to the distribution and depth of the blood-red color of the product after $t = t_{sw}$. If the blood-red color is present at $t = t_2$ ($>t_1$) in a region where the color does not exist at $t = t_1$, this indicates that the chemical reaction takes place in the region during the period of t_1–t_2. Also, if the blood-red is remarkably deeper at $t = t_2$ than at $t = t_1$ in a region, even if the color is present during the period of t_1–t_2, this indicates that the chemical reaction takes place in the region during the period of t_1–t_2. If the product is still not present at $t = t_2$ in a region where the product does not exist at $t = t_1$, this indicates that the chemical reaction does not take place in the region during the period of t_1–t_2.

The reactive and nonreactive experiments reveals where the less-viscous liquid injected after $t = t_{sw}$ reaches and reacts with the more-viscous liquid during the period of t_1–t_2. The difference between the experiments involving the switch of the injected liquids and the experiments involving the continuous injection of less-viscous liquid should be discussed. Here, we refer to the former and latter experiments as "switching experiments" and "continuous experiments," respectively. For the nonreactive system, there is no substantial difference between the continuous and switching experiments, although the switching experiments is different from the system of classical viscous fingering with two solutions in a sense that a less-viscous solution displaces another which then displaces the more viscous third. This is because the two less-viscous solutions have the same viscosity. For the reactive system, in the switching experiments, when the first less-viscous liquid is injected ($t < t_{sw}$) the reaction does not take place. Therefore, the reactant initially included in the more-viscous liquid is less consumed in the switching experiments than in the continuous experiments after $t > t_{sw}$. This leads to φ_v being apparently smaller in the switching experiments than in the continuous experiments. In the novel experiment, we can recognize the region where the less-viscous liquid injected after t_{sw} reacts the more-viscous liquid during the period of t_1–t_2 in the switching experiments as that in the continuous experiments although there is a difference between the switching and continuous reactive experiments as mentioned above. The reason for this is discussed later. In the novel experiment, t_{sw} was varied. On the basis of no substantial difference for the nonreactive system and difference being overcame for the reactive system between the switching and continuous experiments, the reaction region during the period of t_1–t_2 in the continuous reactive experiments appears to be the superimposition of the reaction regions identified in the various switching reactive experiments by varying t_{sw}.

Results

For the low Pe_v condition

In the present study, the condition shown in Figure 1 where the product distribution significantly depends on φ_v is referred to as "the low Pe_v condition," in contrast, the condition shown in Figure 2 where the difference in the product distribution by φ_v is not significant is referred to as "the high Pe_v condition." We first describe the results obtained by applying the novel method to the experimental condition shown in Figure 1. We set t_1 = 120 s and t_2 = 180 s, respectively. Figure 3 shows the fingering patterns without and with the reaction when t_{sw} = 90 s. The nonreactive experiments (a, b) show that the less-viscous liquid injected after t_{sw} = 90 s reaches the region from the trough to the inside near the trough of the fingers during the period of t = 120–180 s. In the reactive experiments, regardless of φ_v, (c ~ h), the blood-red area is hardly present at t = 120 s, whereas it can be observed in the region from the trough to the inside near the trough of the fingers at t = 180 s. These nonreactive and reactive experimental results indicate that the less-viscous liquid injected after t_{sw} = 90 s reaches a region from the trough to the inside near the trough of the

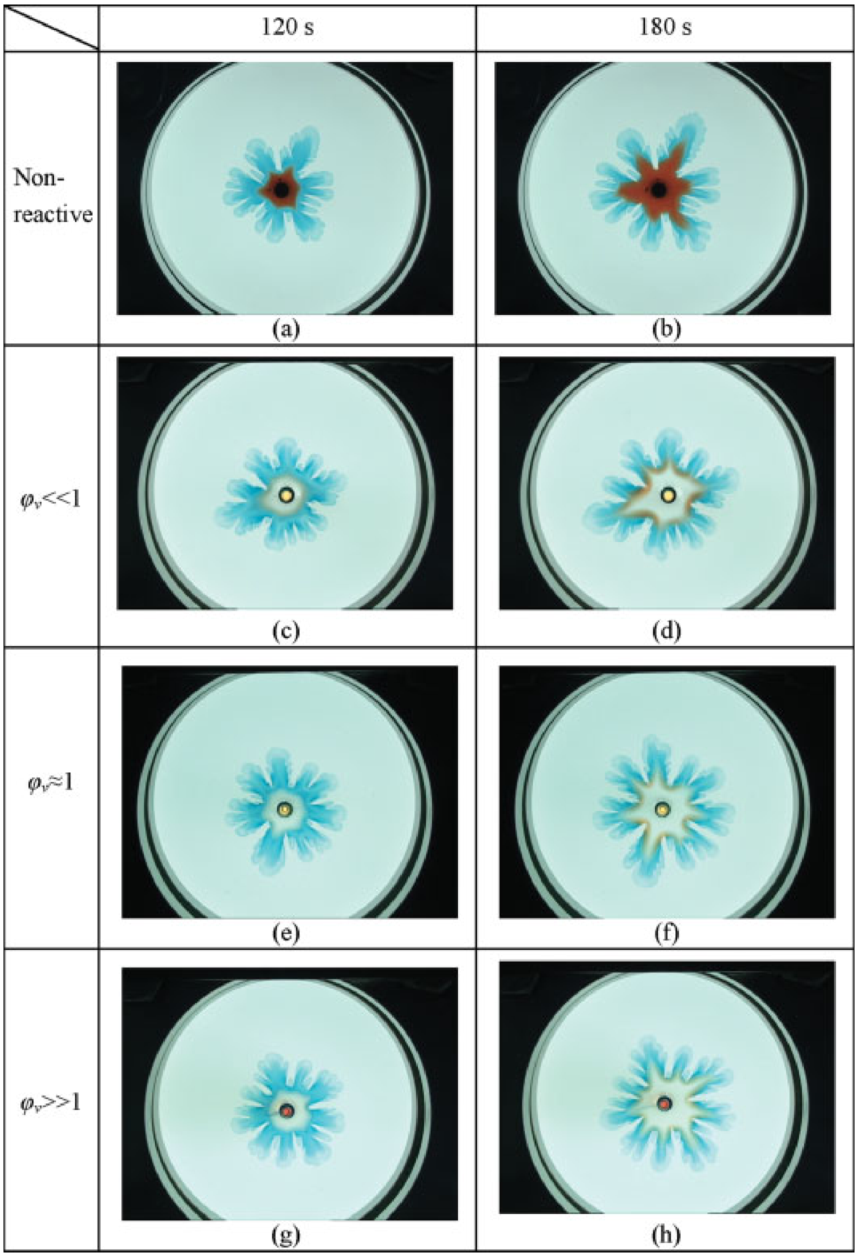

Figure 3. Miscible viscous fingering patterns without and with the reaction at t = 120 s and 180 s when t_{sw} = 90 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$.

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fingers and reacts with the more-viscous liquid in this region during the period of t = 120–180 s regardless of φ_v. Comparing the depth of the blood-red color by φ_v, we find it to be deeper as φ_v is smaller, which shows that the product concentration is higher as φ_v is smaller, although the initial reactant concentrations are set as c_{pr} is theoretically identical when $a = 2$ or they are set as c_{pr} theoretically increases with φ_v when $a = 1$ (this indicates that the product concentration will still increase with a decrease in φ_v if c_{pr} is set to be identical for various φ_v under the condition where the stoichiometric ratio is 1) in the previous steady convection-diffusion-reaction analysis.[20] Since the reaction time is independent of φ_v, the difference in the product concentration is caused by the difference in the overall reaction rate (the quantity of the product produced per unit time and per unit volume). In other words, the overall reaction rate in the region from the trough to the inside near the trough of the fingers is higher as φ_v is smaller under the condition in which c_{pr} is constant. In this figure, the blue color of the less-viscous liquid first injected abruptly becomes light around the tips of the fingers. The color of the dyed less-viscous liquid in the viscous fingering pattern being abruptly light at the fingertips was observed in Nagatsu and Ueda[15] as well. The authors suggested that this was caused by the thickness of the less-viscous liquid's layer in the cell's gap direction becoming abruptly thin at the fingertips, which is analogous to the spike observed in miscible

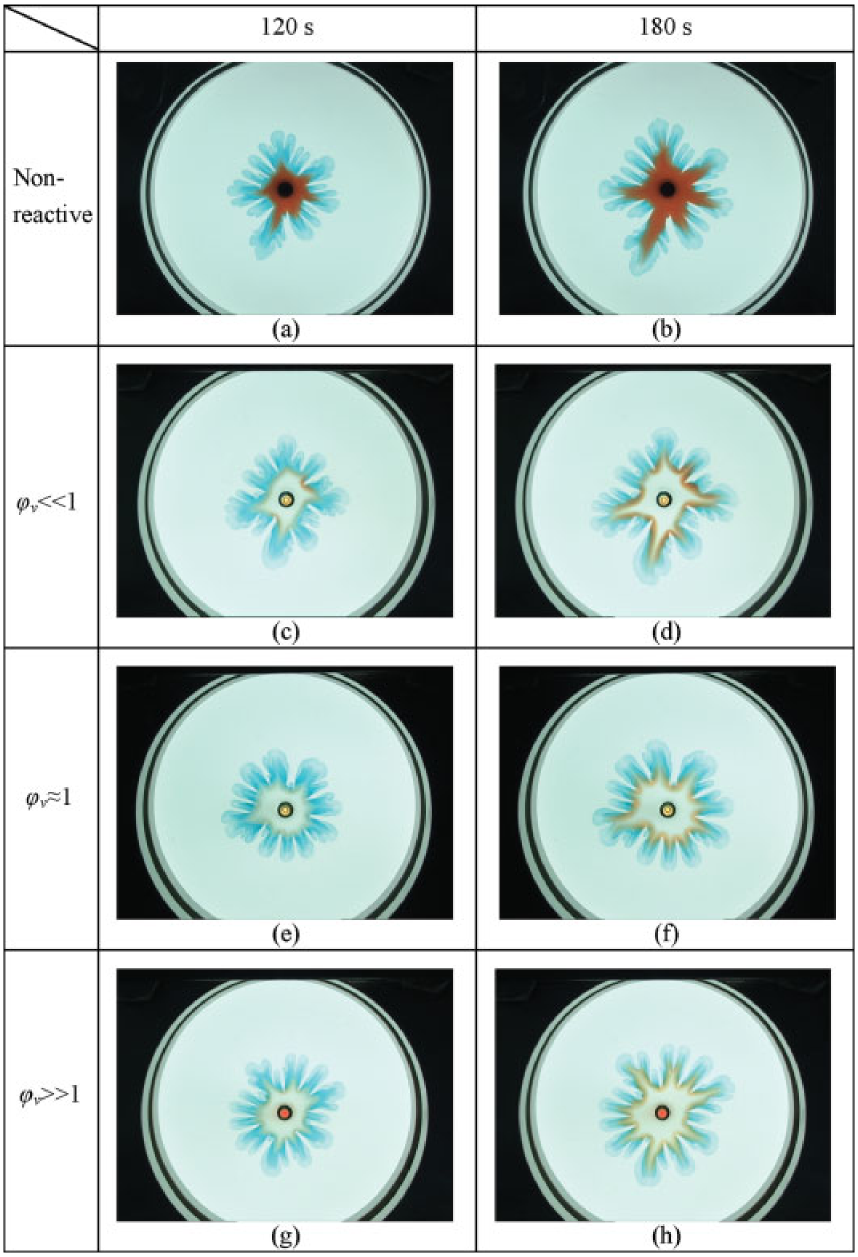

Figure 4. Miscible viscous fingering patterns without and with the reaction at t = 120 s and 180 s when t_{sw} = 60 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$.

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displacement in a capillary tube.[18,23,24] In the present study, the proposed structure of the fingertips in the Hele-Shaw cell is referred to as a "sheet structure."

Figure 4 shows the results when t_{sw} = 60 s. The nonreactive experiments (a, b) show that the less-viscous liquid injected after t_{sw} = 60 s reaches the middle of the advancing fingers during the period of t = 120–180 s. In the reactive experiments, regardless of φ_v, (c ~ h) in the middle of the advancing fingers, the blood-red color is not present at t = 120 s, whereas it can be observed at t = 180 s. The results of these nonreactive and reactive experiments indicate that the less-viscous liquid injected after t_{sw} = 60 s reaches the middle of the advancing fingers and reacts with the more-viscous liquid at least in this region during the period of t = 120–180 s regardless of φ_v. Again, the shade of the blood-red color is darker as φ_v is smaller, which indicates that the overall reaction rate is higher as φ_v is smaller in the middle of the advancing fingers under constant c_{pr}.

Figure 5 shows the results when t_{sw} = 30 s. In the nonreactive experiments (a, b), the blood-red color spreads to the base of the sheet structure region at t = 120 s (a), and it reaches the tip of the advancing fingers at t = 180 s (b). These results show that the less-viscous liquid injected after t_{sw} = 30 s reaches the tip of the advancing finger during the period of t = 120–180 s. Note that the shade of the blood-red color in the region of the sheet structure is light as well.

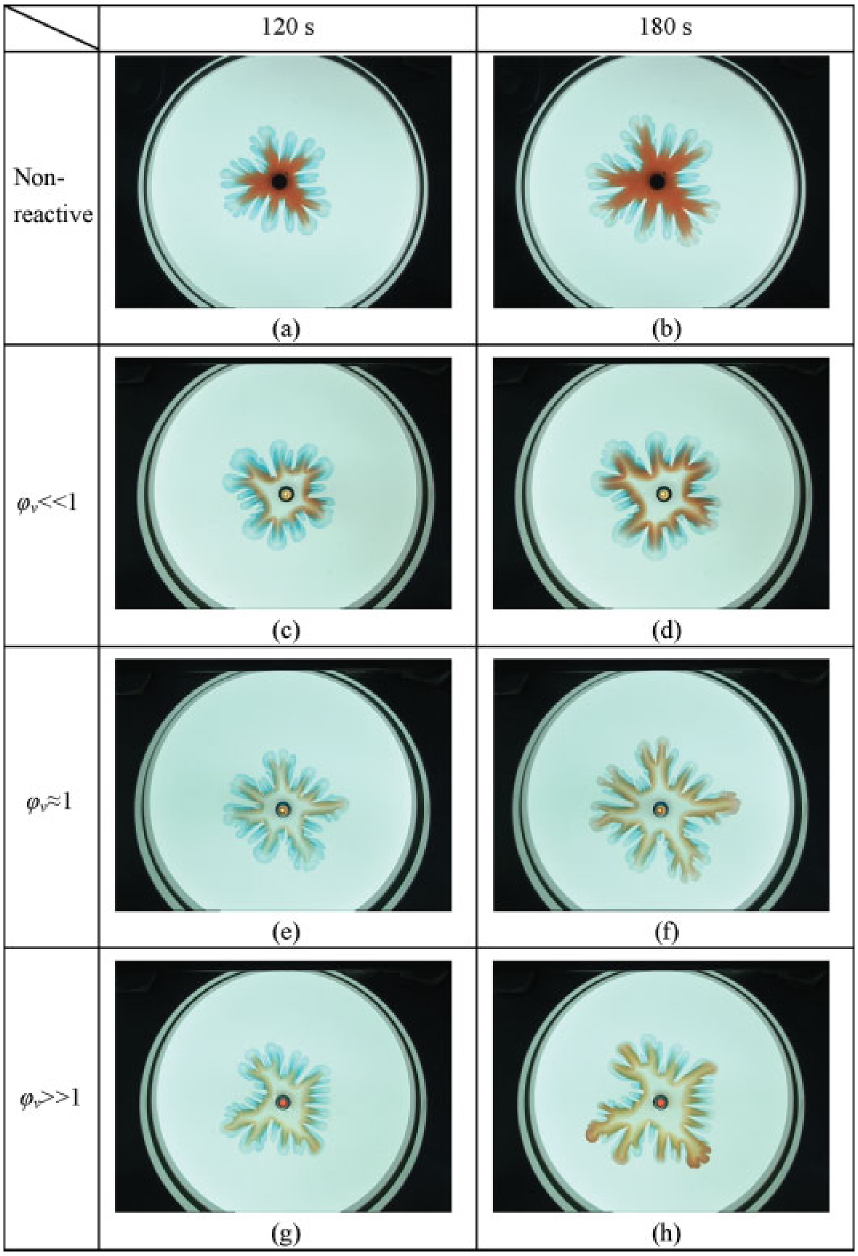

Figure 5. Miscible viscous fingering patterns without and with the reaction at t = 120 s and 180 s when t_{sw} = 30 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$.

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This indicates that the flow of the less-viscous liquid around the fingertips is not plug flow, but a flow having an approaching velocity toward the fingertips until at least t = 180 s. In the reactive experiments, when $\varphi_v \approx 1$ (e, f), in the region from the front to the tip of the advancing fingers, the blood-red color is not present at t = 120 s, but it can be observed at t = 180 s, or the shade is deeper at t = 180 s than at t = 120 s. This indicates that the less-viscous liquid injected after t_{sw} = 30 s reacts with the more-viscous liquid at least in this region during the period of t = 120–180 s. When $\varphi_v \ll 1$ (c, d), the blood-red color is not present in the front of the advancing fingers at t = 120 s, but it can be observed at t = 180 s, or the shade is deeper at t = 180 s than at t = 120 s. This indicates that the less-viscous liquid injected after t_{sw} = 30 s reacts with the more-viscous liquid at least in the front of the advancing fingers during the period of t = 120–180 s. In the sheet structure of the advancing fingers, an extremely light blood-red shade is observed at t = 180 s. Combined with the nonreactive experimental result, in the sheet structure of the advancing fingers, we conclude that no reaction takes place, although the less-viscous liquid injected after t_{sw} = 30 s reaches the tip of the advancing fingers during the period of t = 120–180 s, while the previously formed product is transported by the flowing of the less-viscous liquid. When $\varphi_v \gg 1$ (g, h), in the front of the advancing fingers, the blood-red color is not present at

t = 120 s, but it can be observed at t = 180 s, or the shade is deeper at t = 180 s than at t = 120 s. The blood-red color is remarkably deep at t = 180 s in the sheet structure of the advancing fingers. These findings indicate that the less-viscous liquid injected after t_{sw} = 30 s reacts with the more-viscous liquid from the front to the tip of the advancing fingers during the period of t = 120–180 s. Furthermore, the reaction is more remarkable in the sheet structure, which indicates that the overall reaction rate is higher in the sheet structure than in the front of the advancing fingers. The shielded fingers remain blue during the period of t = 120–180 s in both the nonreactive and reactive experiments (a ~ h), which shows that the less-viscous liquid injected after t_{sw} = 30 s does not penetrate into the shielding fingers, indicating that the reactant included in the less-viscous liquid is not introduced into the shielding fingers during the period of t = 120–180 s. Thus, no reaction takes place there during the period of t = 120–180 s.

We deduce the reaction region during the period of t = 120–180 s for the continuous reactive experiments based on the results obtained in Figures 3–5. The region where the less-viscous liquid injected after t = 90 s reacts with the more-viscous liquid during t = 120–180 s in the continuous reactive experiments is considered to be the same as that identified in Figure 3, even though φ_v is apparently smaller in the switching reactive experiments than in the continuous reactive experiments. This is because the reaction regions identified in Figure 3 are independent of φ_v. This situation is true in the results in Figure 4. In other words, the region where the less-viscous liquid injected after t = 60 s reacts with the more-viscous liquid during t = 120–180 s in the continuous reactive experiments is considered to be the same as that identified in Figure 4. In the case of t_{sw} = 30 s in the switching experiments, the amount of nonreactive liquid first injected is just one-sixth of the total amount of the less-viscous liquid injected when t = 180 s. Thus we consider the effects of φ_v being apparently smaller in the switching reactive experiments than in the continuous reactive experiments on the identification of the reaction region to be unimportant. Thus, the region where the less-viscous liquid injected after t = 30 s reacts with the more-viscous liquid during t = 120–180 s in the continuous reactive experiments can be considered to be the same as that identified in Figure 5. On the basis of the discussion, the reaction region during the period of t = 120–180 s in the continuous reactive experiments can be regarded as superimposition of the reaction regions identified in Figures 3–5.

When $\varphi_v \approx 1$, the identified reaction region is the trough, inside, and tip of the advancing fingers. When $\varphi_v \ll 1$, it is the trough and inside of the advancing fingers, while the reaction does not take place in the tip of the advancing fingers. When $\varphi_v \gg 1$, it is the trough, inside, and tip of the advancing fingers, and the overall reaction rate is higher in the sheet structure than in the trough and inside areas. In addition, we find that the overall reaction rate on the inside of the fingers depends on φ_v and increases with a decrease in φ_v under constant c_{pr}. Without regard to φ_v, the reaction does not take place in the shielded fingers during the period of t = 120–180 s. The above-identified reaction region is shown in Figure 6 by using the reactive viscous fingering patterns in the continuous reactive experiments at t = 180 s. In Figure 6, the blood-red region indicated by blue and red circles is where the reaction takes place. In Figure 6c, the overall reaction rate in the region indicated by the red circles is higher than that in the region indicated by blue circles. The blood-red region indicated by black circles is the region where the reaction does not take place even though the product exists during t = 120–180 s. The black circles show that the reaction region at a given time does not exactly coincide with the region where the product exists at the given time. The red circles show that the overall chemical reaction rate is different in the reactive fingering pattern when $\varphi_v \gg 1$. In the blood-red region indicated by the blue circles, except for that in the fingertips when φ_v = 1, the overall reaction rate depends on φ_v and increases with a decrease in φ_v under constant c_{pr}. The identification of the reaction region answers the question of why the difference in the product concentration in the fingering pattern when $\varphi_v \ll 1$ and that when $\varphi_v \gg 1$ arise, respectively. The difference in the product concentration in the fingering pattern when $\varphi_v \ll 1$ is caused by whether the reaction takes place inside the fingers or not in the sheet structure region, whereas that when $\varphi_v \gg 1$ is caused by the difference in the overall reaction rate inside the fingers and in the sheet structure region.

The reaction region in the fingering pattern may vary as time proceeds. Thus, we next applied the novel method by setting t_1 = 300 s and t_2 = 360 s. The identified reaction region in the continuous reactive experiments during the period of t = 300–360 s is shown in Figure 7 by using the reactive fingering pattern in the continuous reactive experiments at t = 360 s. In Figure 7, like in Figure 6, the blood-red region indicated by blue and red circles is the region where the reaction takes place. In Figure 7c, the overall reaction rate in the region indicated by the red circles is higher than that in the region indicated by blue circles. The blood-red region indicated by black circles is the region where no reaction takes place during t = 300–360 s. In the blood-red region indicated by the blue circles except for that in the fingertips when φ_v = 1, the overall reaction rate depends on φ_v and increases with a decrease in φ_v under constant c_{pr}. These show that the results obtained in Figure 7 are essentially the same as those obtained in Figure 6. Figures 6 and 7 show that the characteristics of reactive flow fields remain unchanged during t = 120–360 s. As shown in Figure 1, at t = 30 s the blood-red color is observed around the tips of all fingers regardless of φ_v. This indicates that the reaction region in the advancing fingers changes with time when $\varphi_v \ll 1$. Around the fingertips, the reaction occurs during the initial stage but not thereafter. When $\varphi_v \approx 1$ and $\varphi_v \gg 1$, in the advancing fingers, the reaction takes place regardless of time under the present experimental conditions. In the shielded fingers, as time proceeds, the reaction does not take place but the product produced at the initial stage remains.

For the high Pe_v condition

We will now describe the results obtained by applying the novel method to the experimental condition shown in Figure 2. We set t_1 = 50 s and t_2 = 60 s. Figure 8 shows the results when t_{sw} = 35 s. The nonreactive experiments (a, b) show that the less-viscous liquid injected after t_{sw} = 35 s reaches the region from the trough to the inside near the trough of

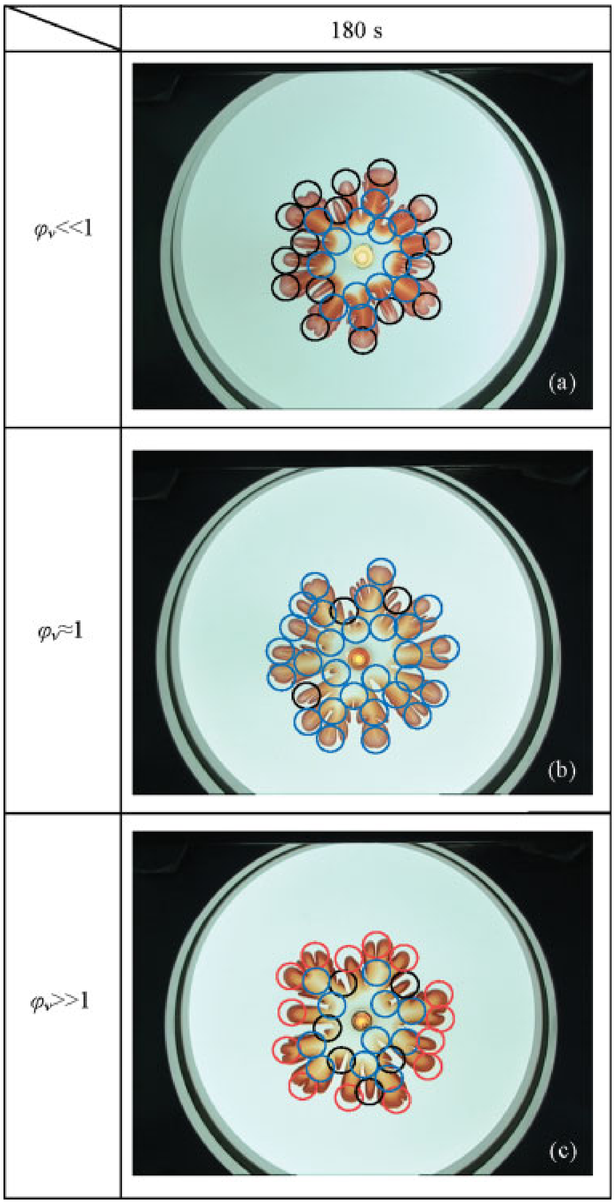


Figure 6. Identified reaction region in reactive miscible viscous fingering patterns in the continuous reactive experiment at t = 180 s for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$.

The meanings of the blue, black, and red circles are drawn in the text. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the advancing fingers during the period of t = 50–60 s. In the reactive experiments, regardless of φ_v (c–h), in the region from the trough to the inside near the trough of the advancing fingers, the blood-red color is not present at t = 50 s, whereas it can be observed at t = 60 s. These nonreactive and reactive experiments indicate that the less-viscous liquid injected after t_{sw} = 35 s reaches a region from the trough to the inside near the trough of the advancing fingers and reacts with the more-viscous liquid in this region during the period of t = 50–60 s regardless of φ_v. Comparing the depth of the blood-red color according to φ_v, we find a deeper shade of blood-red as φ_v grows smaller, which indicates that the overall reaction rate is higher as φ_v is smaller in this region under constant c_{pr}. In this figure, the blue color of the less-viscous liquid first injected around the fingertips at a relatively long distance away is lighter than that inside of the fingers. The color depth being lighter around the fingertips at a relatively long distance from the base of the fingers compared with the color around the base was observed in Nagatsu and Ueda[16] as well. The authors considered the con-

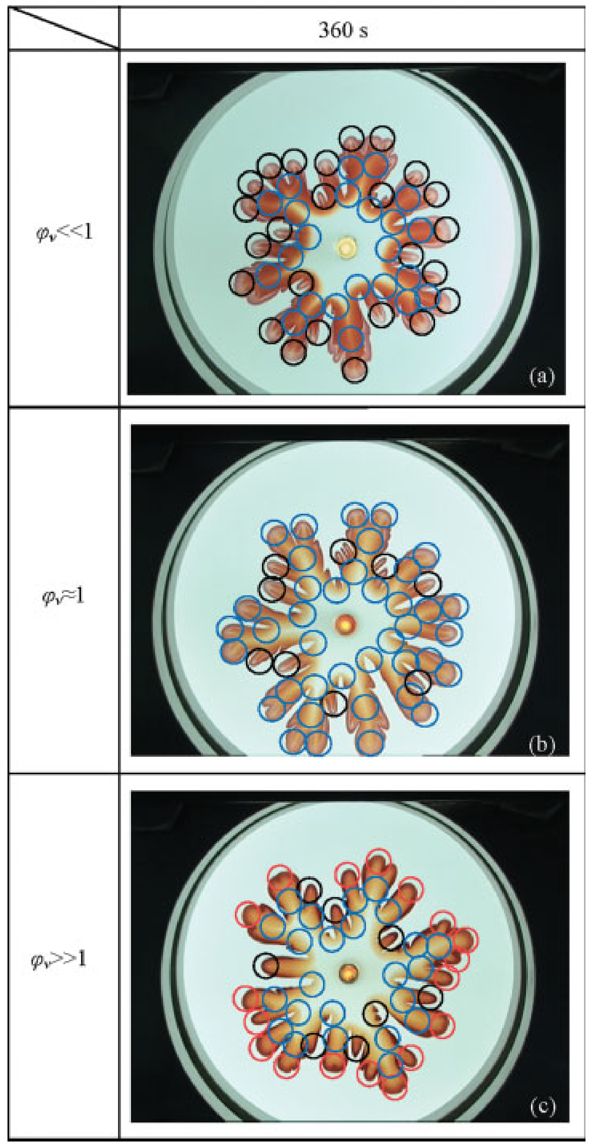


Figure 7. Identified reaction region in reactive miscible viscous fingering patterns in the continuous reactive experiment at t = 360 s for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 6.0 \times 10^3$.

The meanings of the blue, black, and red circles are drawn in the text. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

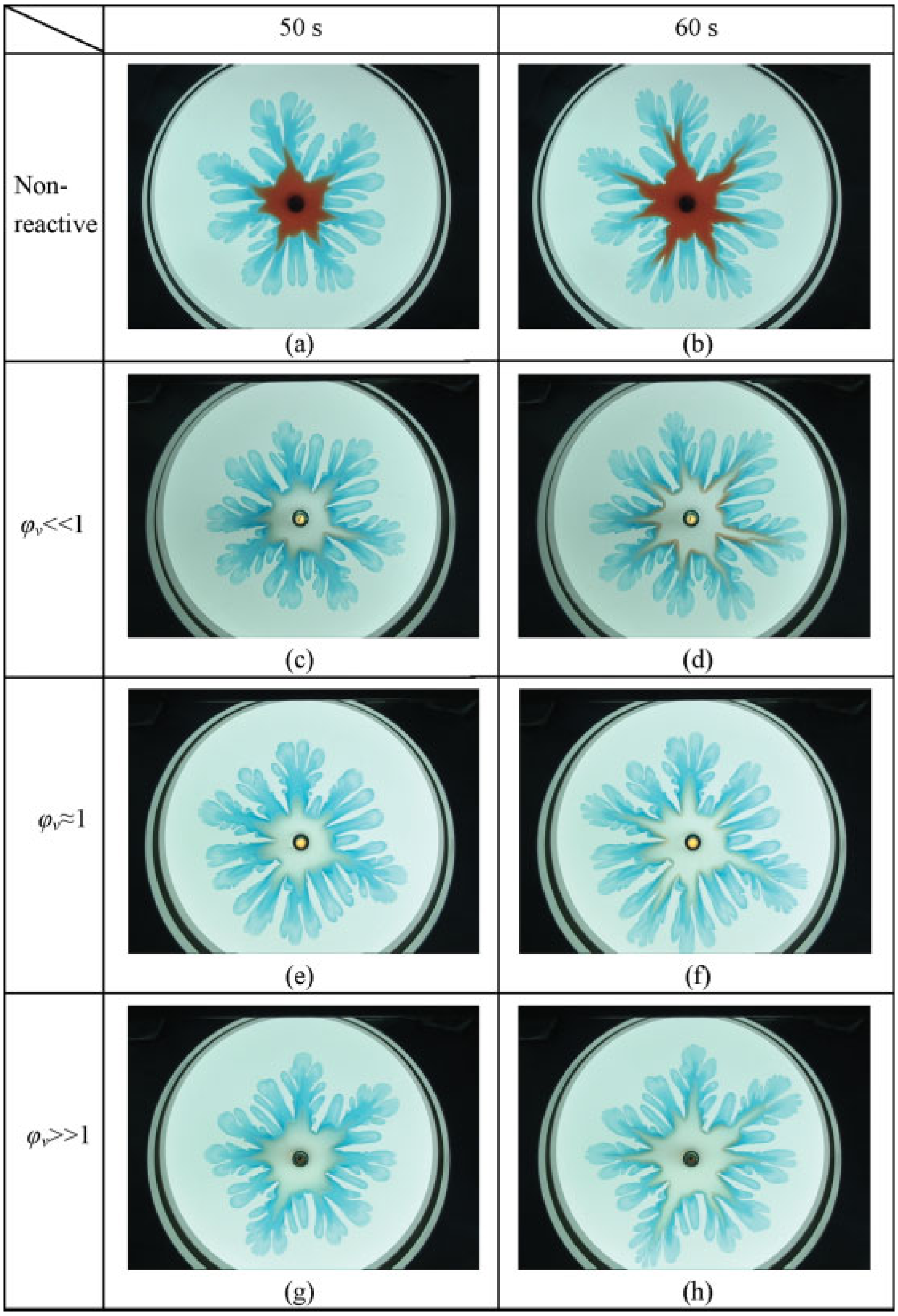


Figure 8. Miscible viscous fingering patterns without and with the reaction at t = 50 s and 60 s when t_{sw} = 35 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 3.6 \times 10^4$.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

figuration of the less-viscous liquid's layer in the cell's gap direction to be a tongue-like shape. In the present study, this is referred as to a "tongue structure."

Figure 9 shows the results when t_{sw} = 30 s. The nonreactive experiments (a, b) show that the less-viscous liquid injected after t_{sw} = 30 s reaches the middle of the advancing fingers during the period of t = 50–60 s. In the reactive experiments, regardless of φ_v (c–h), in the middle of the advancing fingers, the blood-red color is not present at t = 50 s, whereas it can be observed at t = 60 s. The results of these nonreactive and reactive experiments indicate that the less-viscous liquid injected after t_{sw} = 30 s reaches the middle of the advancing fingers and reacts with the more-viscous liquid at least in this region during the period of t = 50–60 s regardless of φ_v. Again, the shade of the blood-red color is deeper as φ_v is smaller, which indicates that the overall reaction rate is higher as φ_v is smaller in this region under constant c_{pr}.

Figure 10 shows the results when t_{sw} = 20 s. The nonreactive experiments (a, b) show that the less-viscous liquid

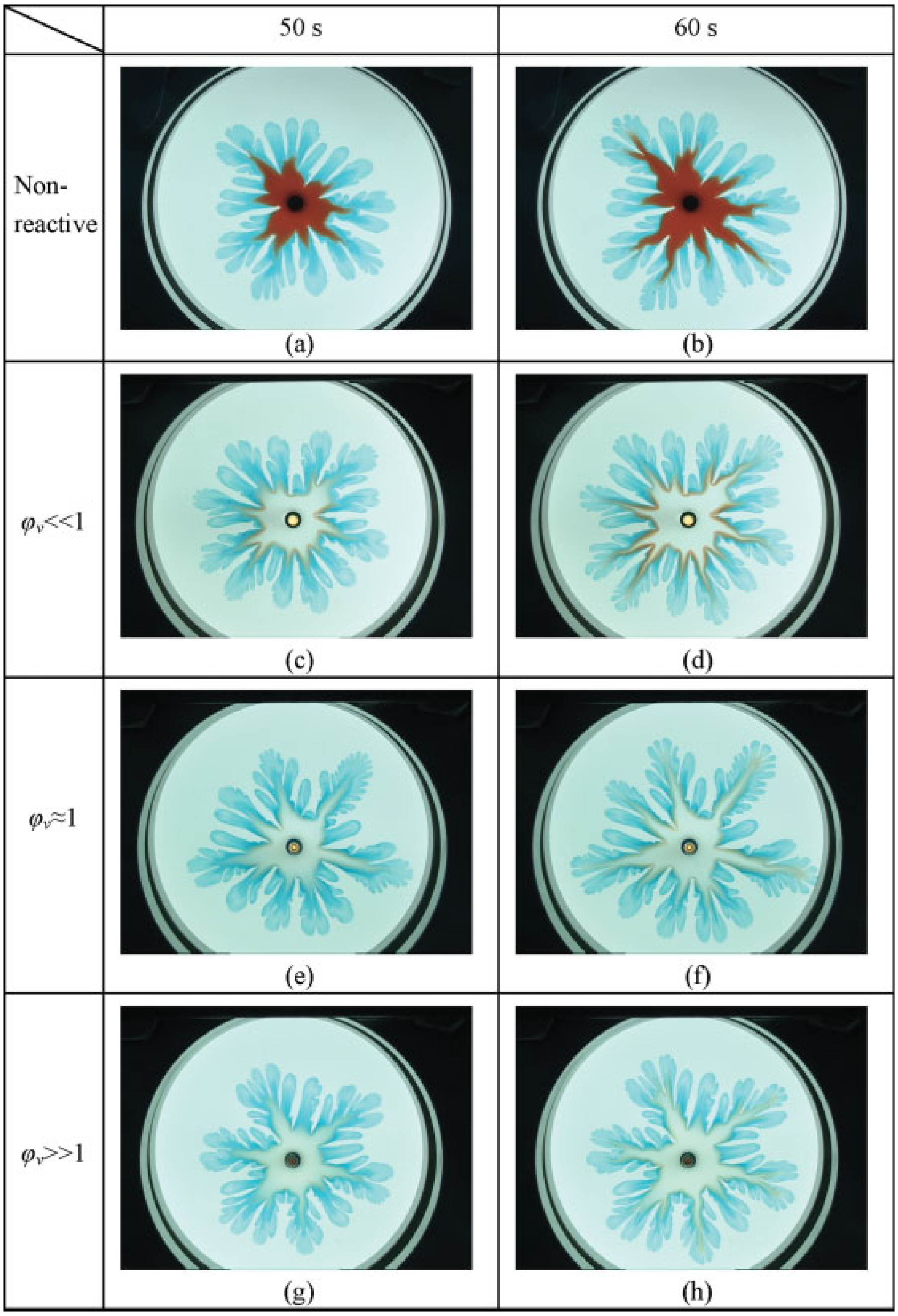


Figure 9. Miscible viscous fingering patterns without and with the reaction at t = 50 s and 60 s when t_{sw} = 30 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 3.6 \times 10^4$.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

injected after t_{sw} = 20 s reaches the tip of the advancing fingers during the period of t = 50–60 s. This indicates that the flow of the less-viscous liquid around the fingertips is not plug flow, but a flow with an approaching velocity toward the fingertips until at least t = 60 s. In the reactive experiments, without regard to φ_v, in the region from the front to the tip of the advancing fingers, the blood-red color is not present at t = 50 s, whereas it can be observed at t = 60 s, or the shade is deeper at t = 60 s than at t = 50 s. This indicates that the less-viscous liquid injected after t_{sw} = 20 s reacts with the more-viscous liquid at least from the front to the tip of the advanced fingers regardless of φ_v. We want to emphasize that the reaction takes place around the tips even when $\varphi_v \ll 1$, which differs from the case of low Pe_v. Again, around the front of the advancing fingers, the shade of the blood-red color is deeper as φ_v is smaller, which indicates that the overall reaction rate is higher as φ_v is smaller in this region under constant c_{pr}. The shielded fingers remain blue during the period of t = 50–60 s in both the nonreactive and reactive experiments. This suggests that the less-viscous

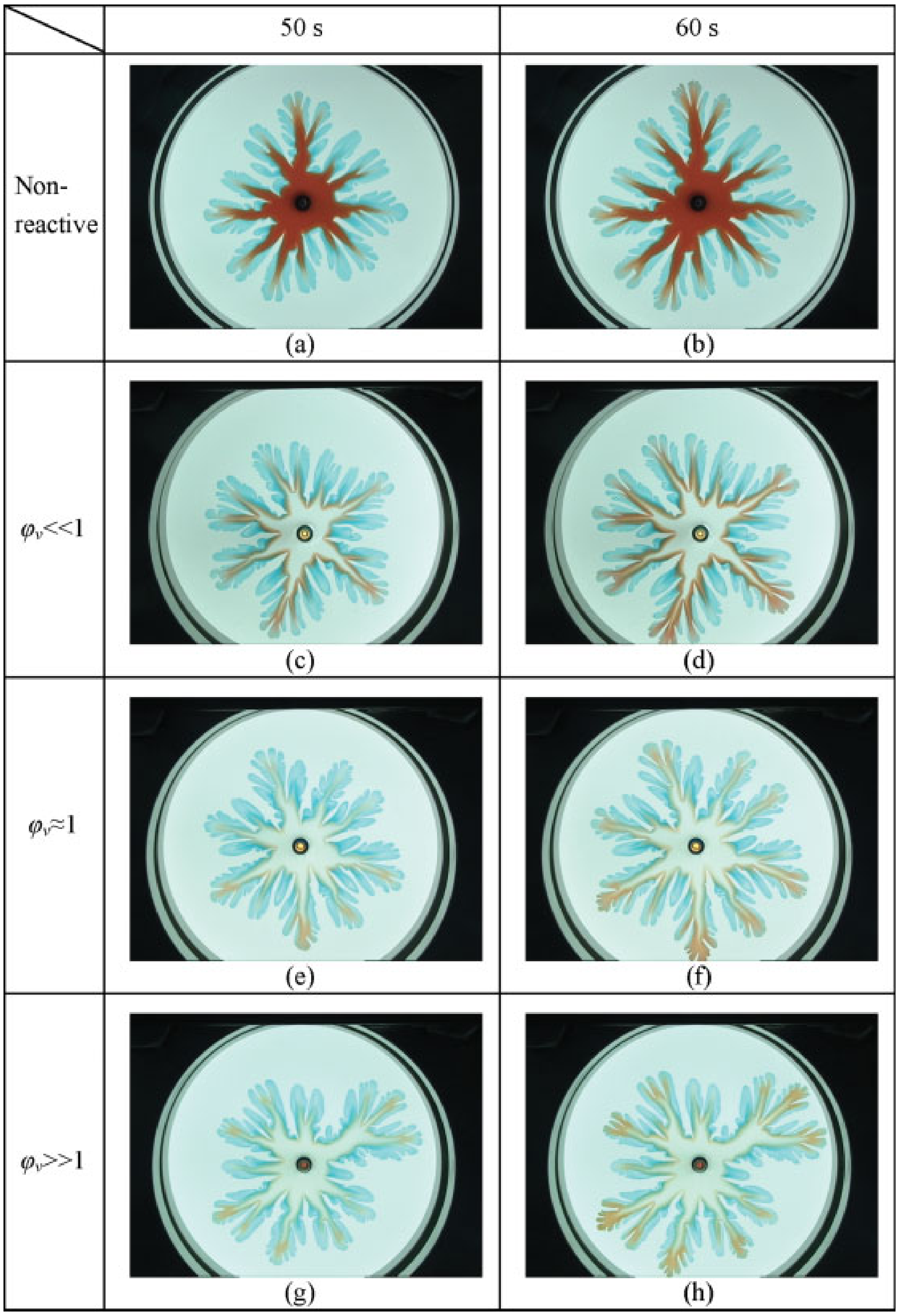


Figure 10. Miscible viscous fingering patterns without and with the reaction at t = 50 s and 60 s when t_{sw} = 20 s, for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 3.6 \times 10^4$.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

liquid injected after t_{sw} = 20 s does not penetrate into the shielding fingers. Thus, the reaction does not take place there during the t = 50–60 s time period.

The reaction region in the continuous reactive experiments during the period of t = 50–60 s can be regarded as the superimposition of the reaction region identified by Figures 8–10 on the basis of the discussion presented above in association with Figure 6. The reaction region during the period occurs in the trough, inside, and tip of the advancing fingers regardless of φ_v. However, we find that the overall reaction rate on the inside of the fingers depends on φ_v and increases with a decrease in φ_v under constant c_{pr}. These results show that the dependence of the product distribution on φ_v is present on the inside of the fingers although that in the whole fingering pattern on φ_v is diminished. Without regard to φ_v, the reaction does not take place in the shielded fingers during the period of t = 50–60 s. The above-identified reaction region is shown in Figure 11 by using the reactive viscous fingering pattern in the continuous reactive experiments at t = 60 s. In Figure 11, the blood-red color region, except for

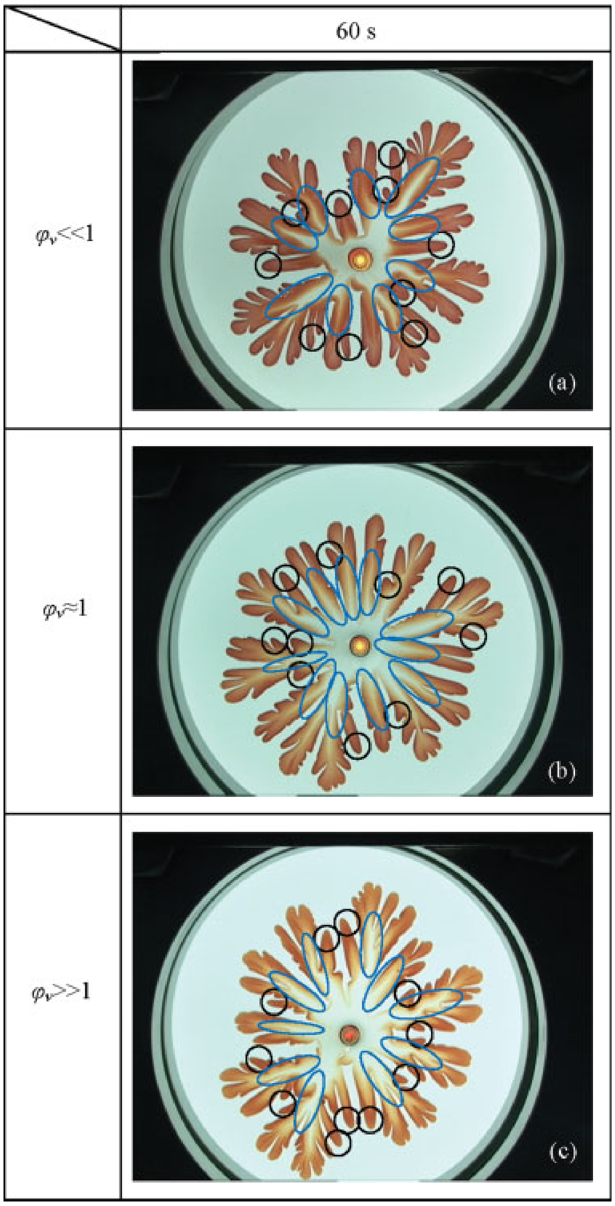


Figure 11. Identified reaction region in reactive miscible viscous fingering patterns in the continuous reactive experiment at t = 60 s for for $\varphi_v \ll 1$, $\varphi_v \approx 1$, and $\varphi_v \gg 1$ under the condition of $Pe_v = 3.6 \times 10^4$.

The meanings of the blue and black circles are drawn in the text. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

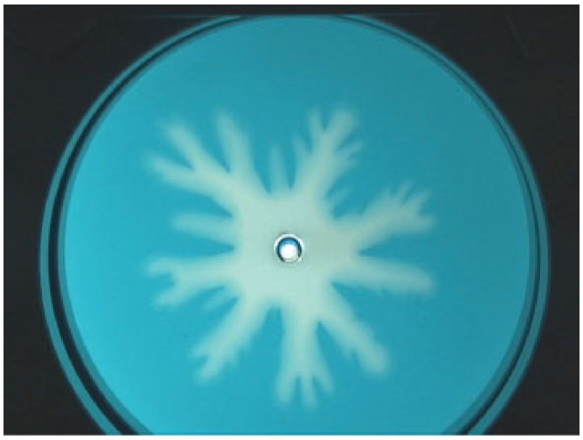
the region identified by the black circles, indicates the reaction region, whereas shielded fingers indicated by the black circles represent the region in which the reaction does not take place even though the product exists during the t = 50–60 s period. In the blood-red region indicated by the blue circles, the overall reaction rate depends on φ_v and increases with a decrease in φ_v under constant c_{pr}. As shown in Figure 2, at t = 5 s the blood-red color is observed around the tips of all fingers regardless of φ_v. This indicates that in the advancing finger the reaction takes place in the trough, inside, and tip regardless of φ_v and time under the present experimental conditions. In the shielded fingers, as time proceeds, the reaction does not take place but the product produced during the initial stage remains.

Discussion

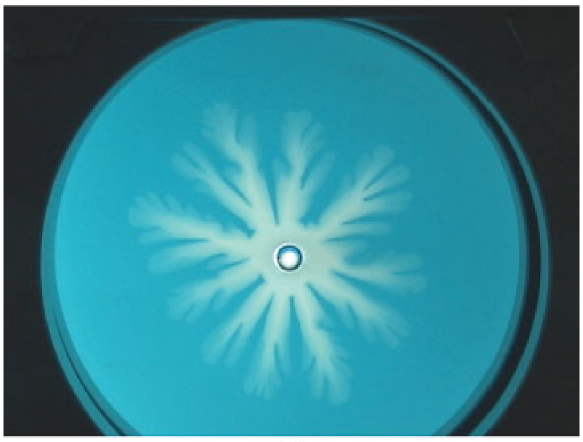
Relationship between the identified reaction region in the fingering pattern and the structure in the cell's gap direction

Here, we discuss the relationship between the identified reaction region in the fingering pattern and the structure in the cell's gap direction. Figures 12a, b show the nonreactive viscous fingering patterns under the same conditions as those in Figure 1 (the low Pe_v case) and 2 (the high Pe_v case) in which colorless water displaces 99 wt % glycerin solution dyed blue by IC solution. In these fingering patters, the thickness of the less-viscous liquid's layer in the cell's gap direction is larger as the blue color is lighter. In Figure 12a, the fingering pattern is colorless around the injection hole, which indicates the less-viscous liquid completely displaces the more-viscous liquid over the cell's gap there. The blue color becomes extremely light at the fingertips due to the sheet structure. We can observe the light-blue color in other regions, i.e., the trough and inside of the fingers. In this region, the more-viscous liquid is not completely displaced. On the basis of the observation results, we can divide the fingering pattern into three regions in terms of the structure in the cell's gap direction. The region around the injection hole where the more-viscous liquid is completely displaced is referred to as the "complete displacement region," the region in the trough and inside of the fingers where the more-viscous liquid is not completely displaced is referred to as the "partial displacement region," and the remaining region is the "sheet structure region." In Figure 12b, the fingering pattern is colorless around the injection hole, which indicates the less-viscous liquid completely displaces the more-viscous liquid over the cell's gap there. The shade of blue in the trough and inside of the fingers is similar to that in the low Pe_v case in Figure 12a. In this region, the more-viscous liquid is not completely displaced. The shade of blue in the tongue structure is deepest in the fingering pattern. However, the shade is lighter than that in the sheet structure region in Figure 12a. On the basis of the observation results, we can divide the fingering pattern into three regions in terms of the structure in the cell's gap direction as we did in the low Pe_v case. That is, there is a "complete displacement region" around the injection hole, a "partial displacement region" in the trough and inside of the fingers, and a "tongue structure region."

Comparing Figure 12a with Figure 7, we find that the blood-red region indicated by blue circles except for the sheet structure region for $\varphi_v \approx 1$ coincides with the partial displacement region. Comparing Figure 12b with Figure 11, we find that the blood-red region indicated by blue circles coincides with the partial displacement region. These results show that the overall reaction rate increases with a decrease in φ_v under constant c_{pr} in the partial displacement region for both the low and high Pe_v cases. Furthermore, this shows that the structure in the cell's gap direction is responsible for the product distribution in the fingering pattern. Note that the



(a)



(b)

Figure 12. Miscible viscous fingering pattern without the reaction where the more-viscous liquid is dyed under the condition of b = 0.3 mm. (a) $Pe_v = 6.0 \times 10^3$ at t = 360 s, (b) $Pe_v = 3.6 \times 10^4$ at t = 60 s.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

region where the product is not present coincides with the complete displacement region for both the low and high Pe_v cases.

Model of steady convection-diffusion-reaction in two miscible liquids

In Nagatsu et al.,[20] we conducted a steady convection-diffusion-reaction analysis that was considered to be an improved version of the previous analyses performed by Nagatsu and Ueda[15,17] where a distinct interface between two miscible fluids was assumed. In other words, the analysis in Nagatsu et al.[20] took miscibility into account. The analytical results showed that when the convective effect was weak, the reaction plane was located relatively far from the boundary in the less-viscous liquid for $\varphi_v \ll 1$, whereas it was located in the boundary for $\varphi_v \approx 1$ and $\varphi_v \gg 1$. As the convective effect increased, the location of the reaction plane was shifted toward the boundary for $\varphi_v \ll 1$, whereas the change in the location of the reaction plane was negligible for $\varphi_v \approx 1$ and $\varphi_v \gg 1$. Thus, the dependence of the location on the reaction plane on φ_v was decreased as the convective effect increased. These analytical results were essentially similar to those in the previous analyses,[15,17] although there was a difference in that when $\varphi_v \geq 1$, the reaction plane was located in the boundary region in Nagatsu et al.,[20] whereas it was located in the more-viscous liquid region in Nagatsu and Ueda.[15,17]

Physical model of reacting flow fields in the cell's gap direction

In this section, we discuss the reacting flow fields in the cell's gap direction on the basis of the reaction region identified in the present study and arguments given in the preceding sections of the Discussion section. We propose physical models of the reacting flow fields in an advancing finger in the identified reaction region independent of time. In making the physical models, we postulate the following:

(1) Diffusion is dominant for mass transfer in the cell's gap direction. Note that convection is dominant for the mass transfer in the direction in which the fingers penetrate, since $Pe_v \gg 1$ in the present system.

(2) Since the flow of the less-viscous liquid is not plug flow but a flow with an approaching velocity toward the fingertips, the reactant in the less-viscous liquid is supplied to the reacting flow field.

(3) In contrast, there is no supply of the reactant in the more-viscous liquid into the reacting flow field.

The cases of low Pe_v are shown in Figure 13. This figure is drawn with regard to a single finger. The configuration of the boundary between two liquid can be drawn on the basis of the spike structure in the capillary tube observed in Lajeunesse et al.[23] and Nagatsu et al.[20] and the study of miscible displacement in a Hele-Shaw cell by Lajeunesse et al.[23] (They experimentally showed three types of interface shape depending on the viscosity ratio and flow rate in vertical displacements, referring to them as the absence of shock, the presence of an internal shock, and the presence of a frontal shock, where "shock" means an abrupt decrease in the thickness of the less-viscous liquid layer in the direction of the cell's gap. This type of internal shock corresponds to the spike observed in the miscible displacement in the capillary tube.) When $\varphi_v \gg 1$, we know from the present study that the reaction takes place both in the partial displacement region and in the sheet structure region. Furthermore, the overall chemical reaction rate is higher in the sheet structure region. The reacting flow field is proposed in Figure 13c. The reaction plane is formed in the boundary, as predicted by the previous analysis. In the partial displacement region, the amount of reactant in the more-viscous liquid is significantly smaller than that in the less-viscous liquid because of the significant thinness of the more-viscous liquid's layer and its lower initial concentration. Based on postulation (3), the depletion of the reactant in the more-viscous liquid appears in this region. In other words, the reactant concentration at the edge of the film (boundary film) in the more-viscous liquid decreases with time. This leads to a decrease in the flux of the reactant in the more-viscous liquid provided to the reaction plane with time. Therefore, the overall reaction rate decreases with time. Since the more-viscous liquid's layer is

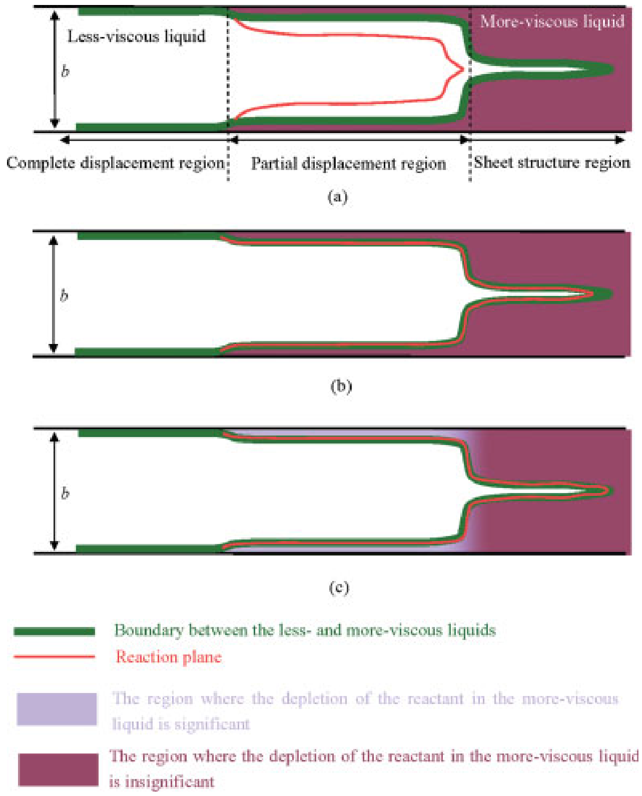


Figure 13. Physical model of the reactive flow fields in the reactive miscible viscous finger in the cell's gap direction for the low Pe_v cases: (a) $\varphi_v \ll 1$, (b) $\varphi_v \approx 1$, (c) $\varphi_v \gg 1$.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

significantly thick in the sheet structure region, the amount of reactant in the more-viscous liquid is large. Thus the depletion of reactant in the more-viscous liquid with time is much less significant in this region than in the partial displacement region. This results in much less of a decrease in the overall reaction rate with time. On the basis of the above-mentioned discussion, the difference in the overall reaction rates in the partial displacement region and in the sheet structure region occurs. It should noted that the product was distributed equivalently in the partial displacement region and in the spike region in the previous capillary tube experiment when $\varphi \gg 1$.[20] This is considered to be due to the thickness of the more-viscous liquid's layer in the partial displacement region being significantly larger in the capillary tube experiment than in the Hele-Shaw cell experiment (the diameter of the capillary tube previously used was 3 mm, which is 10 times as great as the present cell's gap width).

When $\varphi_v \approx 1$, the present study shows that the reaction takes place in both the partial displacement region and the sheet structure region. The reacting flow field is proposed in Figure 13b. The reaction plane is formed in the boundary as predicted by the previous analysis and when $\varphi_v \gg 1$. In the partial displacement region, the depletion of the reactant in the more-viscous liquid is less significant than when $\varphi_v \gg 1$ because the initial reactant concentration in the more-viscous liquid is comparable to that in the less-viscous liquid. This is the reason why the overall reaction rate is higher when $\varphi_v \approx 1$ than when $\varphi_v \gg 1$ in this region. No significant difference between the overall reaction rate in the partial displacement region and that in the sheet structure region when $\varphi_v \approx 1$ can be explained by assuming that the depletion of the reactant in the less-viscous liquid occurs even though the reactant is supplied by the flow. In other words, the depletion of the reactants in the less-viscous liquid until the less-viscous liquid reaches the sheet structure is more significant when $\varphi_v \approx 1$ than when $\varphi_v \gg 1$. This corresponds to the overall reaction rate in the partial displacement being higher when $\varphi_v \approx 1$ than when $\varphi_v \gg 1$.

When $\varphi_v \ll 1$, the present study shows that the reaction takes place in the partial displacement region, whereas it does not take place in the sheet structure region. The reacting flow field is proposed in Figure 13a. The reaction plane, which was predicted to be located relatively far from the boundary in the less-viscous liquid, is not able to form along the boundary in the sheet structure region because the less-viscous liquid's layer abruptly thins and the thin layer becomes quite long. Thus, the reaction plane is formed in the less-viscous liquid in the partial displacement region. In this region, the depletion of the reactants in the more-viscous liquid with time is much less significant when $\varphi_v \ll 1$ than when $\varphi_v \approx 1$ or $\varphi_v \gg 1$ because of its higher initial concentration, in spite of the thinness of the more-viscous liquid's layer. Therefore, the decrease in the overall reaction rate with time is much less significant when $\varphi_v \ll 1$ than when $\varphi_v \approx 1$ or $\varphi_v \gg 1$, even if some depletion of the reactant in the less-viscous liquid occurs. This is the reason why the

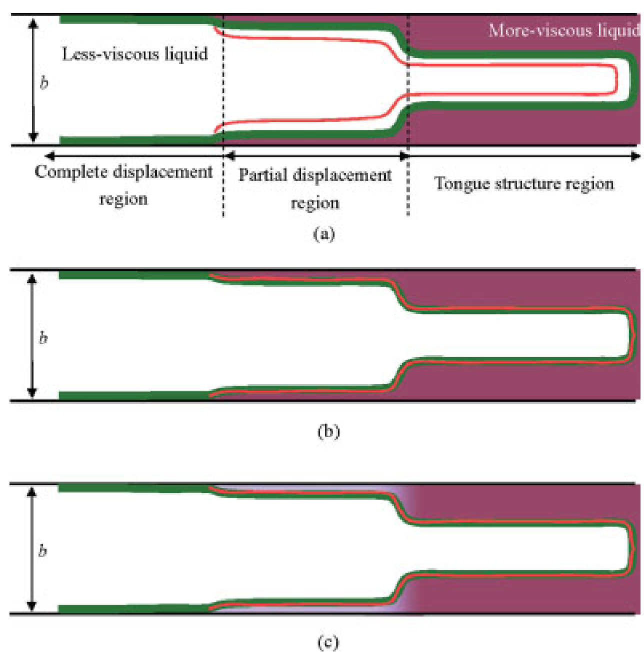


Figure 14. Physical model of the reactive flow fields in the reactive miscible viscous finger in the cell's gap direction for the high Pe_v cases: (a) $\varphi_v \ll 1$, (b) $\varphi_v \approx 1$, (c) $\varphi_v \gg 1$.

The meanings of each color are the same as those in Figure 13. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

overall reaction rate in the partial displacement region is higher when $\varphi_v \ll 1$ than when $\varphi_v \approx 1$ or $\varphi_v \gg 1$.

The cases of high Pe_v are shown in Figure 14. This figure is drawn with regard to a single finger. The configuration between the two liquid can be drawn on the basis of Lajeunesse et al.[25] (the tongue-like configuration was schematically drawn in Figure 1 in their article). Although this schematic does not involve the partial displacement region, we add it based on the observation in Figure 12b. The configuration in the partial displacement region is drawn in this case as well as it is in Figure 13. The present study shows that the reaction takes place in the partial displacement and tongue structure regions regardless of φ_v. Furthermore, the overall reaction rate in the partial displacement region is higher as φ_v is smaller under constant c_{pr}. The reaction plane is formed in the boundary as predicted in the previous analysis when $\varphi_v \approx 1$ and when $\varphi_v \gg 1$ (b, c). When $\varphi_v \ll 1$, the reaction plane, which was predicted to be located in the less-viscous liquid closer to the boundary compared with the low Pe_v case by the previous analysis, can be formed along the boundary in the less-viscous liquid around the fingertip because of the tongue structure (a). These findings show that the reaction plane is located along the boundary regardless of φ_v, which results in a decrease in the dependence of the product distribution on φ_v. In the partial displacement region, on the basis of postulations (2) and (3), the overall reaction rate appears to depend on the ratio between the reactant concentration in the more-viscous liquid and that in the less-viscous liquid. The depletion of the reactants is more significant as the initial reactant concentration in the more-viscous liquid is lower compared with that in the less-viscous liquid. This is the reason why the overall reaction rate is higher as φ_v is smaller under constant c_{pr}.

We want to emphasize that an unsteady concentration field, which means that the reactants are depleted with time, plays an important role in the present physical model of the reacting flow field. However, the unsteady feature does not have significant effects on the difference in the location of the reaction plane by φ_v predicted by the previous steady analysis.[20] This is because the unsteady feature is thought to appear without regard to φ_v. The depletion of the reactant in the less-viscous liquid through the formation of the reactive viscous fingering was also pointed out by Hornof et al.[10,21,22]

Conclusions

In previous studies,[15,16] we experimentally investigated miscible viscous fingering with a chemical reaction in a Hele-Shaw cell, where the reaction was instantaneous and did not affect the hydrodynamics of the fingering. We showed that the product distribution in the fingering pattern significantly depended on φ_v when Pe_v was small.[15] For $\varphi_v \ll 1$, the product was present in large quantities in a relatively broad area within the interior of the fingers, whereas for $\varphi_v \gg 1$, it was concentrated around the tip of the fingers. For $\varphi_v \approx 1$, the product was equally distributed among the interior and tip of the fingers. Also, we showed that the dependence of φ_v on the product distribution was diminished when Pe_v was large.[16] In these cases, the product was present in a relatively broad area from the inside to the tip of the fingers regardless of φ_v. However, the reacting flow field in these phenomena has not been completely elucidated yet. We were not able to exactly recognize where and when the reaction takes place in the fingering pattern. In addition, we did not have a clear answer to the question of why the product is somewhat present in the fingertips at $\varphi_v \ll 1$, whereas it remains within the interior of the fingers at $\varphi_v \gg 1$ when Pe_v is small. In the present study, we developed a novel experimental method that allows us to identify when and where the reaction takes place in the viscous fingering pattern generated in the previous system.[15,16] We applied the novel method under similar experimental conditions to those employed in Nagatsu and Ueda[15,16] to completely elucidate the reacting flow field. The novel method involves switching of the less-viscous liquid injected in the nonreactive and reactive experiments.

The location of the reaction region in the fingering pattern under the condition of small Pe_v in the continuous reactive experiments, in which the product distribution significantly depends on φ_v, has been identified as follows. In the early stage of the fingering formation, the reaction takes place around the whole fingertips where the product is present. As time elapses, the location of the reaction region becomes dependant on φ_v. When $\varphi_v \ll 1$, the reaction takes place in the partial displacement region (in this region the more-viscous liquid is not completely displaced in the cell's gap direction, and thus a thin layer of the more-viscous liquid is left as the fingers propagate) from the trough to the inside of the advancing fingers where the product is present. The reaction does not take place in the sheet structure region around the tip of the advancing fingers, although the product is present there. When $\varphi_v \approx 1$, the reaction takes place in the partial displacement region from the trough to the inside and in the sheet structure around the tip of the advancing fingers where the product is present. When $\varphi_v \gg 1$, the reaction takes place in the partial displacement region from the trough to the inside and in the sheet structure around the tip of the advancing fingers where the product is present. The overall reaction rate is significantly higher in the sheet structure region around the tips than in the partial displacement from the trough to the inside. In addition, the overall reaction rate is higher as φ_v is smaller in the partial displacement region from the trough to the inside of the advancing the fingers under constant c_{pr}. Regardless of φ_v, the reaction does not take place in the shielded fingers, although the product is present there, which shows that the reaction region at a given time does not exactly coincide with the region where the product exists at the given time. These characteristics of the reacting flow field are unchanged with time under the present experimental condition. The identification of the reaction region gives an answer to the question of why the difference in the product concentration in the fingering pattern when $\varphi_v \ll 1$ and that when $\varphi_v \gg 1$ arise, respectively. The difference in the product concentration in the fingering pattern when $\varphi_v \ll 1$ is caused by whether the reaction takes place inside the fingers or not in the sheet structure region, whereas that when $\varphi_v \gg 1$ is caused by the difference in the overall reaction rate inside the fingers and in the sheet structure region. The location of the reaction region in the fingering pattern under the condition of large Pe_v, in which the dependence of the product distribution on φ_v is diminished, has been identified as follows. In the early stage of the fingering formation, the reaction takes place around the whole fingertips where the product is present. As time elapses, the reaction takes place in the partial displacement region from the trough to the inside and in the tongue region around the tip of the advancing fingers, regardless of φ_v. However, the overall reaction rate is higher as φ_v is smaller in the partial displacement region from the trough to the inside under constant c_{pr}. These results show that the dependence of the product distribution on φ_v still appears in the inside of the fingers, although the dependence in the whole fingering pattern on φ_v is diminished. Without regard to φ_v, the reaction does not take place in the shielded fingers, although the product is present there. We propose the physical model of reacting flow fields in the cell's gap direction to explain our experimental findings, in which the dependence of the location of the reaction plane on φ_v, the boundary configuration, and the depletion of the reactant with time are important.

Acknowledgments

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