Title: MORPHOLOGICAL CROSSOVER FOR VISCOUS FINGERING PATTERNS IN POLYMER SOLUTIONS

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MORPHOLOGICAL CROSSOVER FOR VISCOUS FINGERING PATTERNs IN POLYMER SOLUTIONS

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Viscous fingering is the process by which a complex interface develops when a low viscous fluid is injected into a high viscous one. Now, the viscous fingering problem for simple Newtonian fluids is well understood, whereas our understandings for non-Newtonian fluids are far from complete. In this paper, we report the results of an attempt to relate the polymer chain entanglements of polymer solutions to the patterns produced when the solution is driven radially in the Hele-Shaw cell.

Hele-Shaw cell was made of a pair of plane-parallel glass plates with a separation of 0.5 mm. The high viscous fluid was an aqueous solution of hydroxypropyl methyl cellulose (HPMC), and the solution was displaced from the center of the cell by injection of air. The injection pressure was varied from 1.0 to 5.0 kPa. Two different molecular weights were used for the HPMC, 74 x 10^4 and 25 x 10^4 determined by the intrinsic viscosity measurements. The high and low molecular weight HPMC samples are designated as HPMC-1 and HPMC-2, respectively. HPMC concentrations were 0.4, 0.5, and 0.6 g/100mL for HPMC-1 and 1.5 g/100mL for HPMC-2, respectively and these solutions were dyed with methylene blue. The rheology measurements of the HPMC solutions were performed by a Rheometrics Fluid Spectrometer RFS-II. The growing patterns were observed with a CCD video camera and recorded on a videotape. The images of the recorded patterns were analyzed by a digital-image-processing system.

Figure 1 shows the fingering patterns of the 0.5 g/100mL HPMC-1 solution as a function of the injection pressure. The resulting patterns are drastically changed with an increase in the injection pressure. At the injection pressure of 1.0 kPa the resulting pattern is typical highly branched one. At 2.0 kPa the pattern is characteristic of tip-splitting one, and the fingers become more rounded shape as the injection pressure increases. At 5.0 kPa the mode in the finger growth is quite different from both the side-branching and tip-splitting patterns: the growing tip becomes broad and flattening and around tip a bump is gone out and grown up, and then we will denote it as the skewering pattern. Similar morphological crossovers in the fingering pattern were observed for the 0.4 and 0.6 g/100mL HPMC-1 solutions. The injection pressure for the morphology crossover increases with an increase the concentration of HPMC.

On the other hand, the fingering patterns of the HPMC-2 solution are only highly branched one in the entire injection pressure ranges as shown in Figure 2. However, with an increase in the injection pressure the finger width becomes thinner.

We expect that the crossover in the pattern morphology of the fingering experiments should be correlated with the rheological properties of the polymer solutions. The 0.4, 0.5, and 0.6 g/100mL HPMC-1 solutions displayed shear thinning behavior at lower shear rate than 4.0 s^{-1} and their zero-shear rate shear viscosities are 0.050, 0.107, and 0.193 Pa·s, respectively. In contrast, the HPMC-2 solution has 0.140 Pa·s in the lower shear rates than 50 s^{-1}, and above which the shear viscosity gradually decreases with an increase in the shear rate. Difference in the shear thinning on-set between the HPMC-1 and HPMC-2 samples should stem from the more frequent chain...
entanglements in the HPMC-1 solution than that in the HPMC-2 one due to the longer chains. Furthermore, chain entanglement may lead to an exponent less than 2 for the frequency dependence of the dynamic storage modulus, indicating that some phantom aggregates form in the HPMC-1 solutions.

In order to understand such a morphological crossover, we measured the tip velocity and the finger width of the fingering patterns. The tip velocity showed a drastic change accompanied with the morphological transition. For the tip-splitting pattern formation, the tip velocity gradually increased with an increase in the injection pressure. For the skewering pattern, the tip velocity was rapidly increased. Whereas, the finger width also changed with the morphological crossover from the side-branching pattern to the skewering one through the tip-splitting one. A drastic decrease was observed for the crossover from the tip-splitting to the skewering one.

In summary, we have performed viscous fingering experiments using HPMC solutions as a function of the injection pressure and demonstrated a morphological crossover for the solution of high molecular weight HPMC. Moreover, analysis of the fingering pattern showed a good correlation between the morphological crossover and the characteristic quantities, such as tip velocity and finger width.

Figure 1. Changes in pattern morphology for the 0.5 g/100mL HPMC-1 solution as a function of the injecting pressure $p$: a, $p = 1.0$ kPa; b, $p = 2.0$ kPa; c, $p = 3.0$ kPa; d, $p = 5.0$ kPa.

Figure 2. Typical patterns of a 1.5 g/100mL HPMC-2 solution as a function of the injecting pressure $p$: a, $p = 1.0$ kPa; b, $p = 2.0$ kPa; c, $p = 3.0$ kPa; d, $p = 5.0$ kPa.

References