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Miscible displacements between silicone oils in capillary tubes

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Abstract

An experiment in which one viscous silicone oil displaces another more viscous silicone oil in capillary tubes, leading to a version of so-called fingering displacement, is reported here. The fractional volume of the more viscous fluid left on the tube wall after the finger front had passed was measured. This fraction, m, defined as $m = 1 - U_{\mathrm{m}}/U_{\mathrm{tip}}$, is a critical diagnostic parameter used to identify finger patterns. Here, U_{m} is the mean velocity of the Poiseuille flow ahead the finger front and U_{tip} the propagation speed of the finger tip. In the present case, the fraction m is a function of the Peclet number, *Pe*, a viscous Atwood number, *At*, and a gravitational parameter, F. In this experiment, m was obtained by measuring the finger propagation speed, U_{tip}, for a range of injection rates, tube diameters and orientations and fluid viscosities to cover a range of *Pe*, *At* and F. For large *Pe*, the results show the fraction m reached a constant value that depended only on Atwood number. For small *Pe*, gravitational effects were substantial as measured by the magnitude of F, which is the ratio of gravitational to viscous effects. In this case m is a function of all three parameters and it becomes possible to interpolate the results to obtain the value of m when gravity is absent.

Keywords: Miscible displacement; Fingering; Instability; Molecular diffusion; Viscous flow; Porous media; Experiment

1. Introduction

This experimental programme is an extension of that of Petitjeans and Maxworthy [1, hereinafter PM] on miscible displacement in capillary tubes. One of the major questions raised by that work was whether the unexpected behaviour at low Peclet numbers ($Pe = U_{\mathrm{m}}d/D$, where U_{m} is the mean velocity of the Poiseuille flow far ahead of the propagating finger, d the tube diameter and D the molecular diffusion coefficient), was typical of many fluid pairs and had more general application or was unique to the glycerine/water mixtures chosen. Thus, at small *Pe* one would expect a trend towards a single-fluid Poiseuille profile in which case injected fluid would move at the centreline velocity and m would approach 0.5. Such a result has been found by Kuang and Maxworthy [2] when using a single fluid and generating the viscosity difference using temperature. There, thermal diffusivity replaced species diffusivity and very low values of *Pe*, of order 25, could be attained. By contrast the PM experiment found values of m slightly less than 0.5 at low *Pe* and the slope of the m vs. *Pe* curve gave no indication that a value of 0.5 was likely to be reached. In what follows these experiments are repeated using a more precise experimental method and using silicone oils of different molecular weights and hence densities and viscosities. These fluids were chosen because they have similar molecular structures and some of the concerns raised by PM's experiments could be eliminated. Moreover, these fluids are to be used in an extended experiment to be placed on board the International Space Station (ISS) [3] and it was necessary to show that the arguments used to justify the space experiment were valid for the fluids to be used.

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When a viscous fluid injects into a cylindrical tube filled with another fluid of higher viscosity, the low viscous fluid will channel through the high viscous one and leave a layer of high viscous fluid on the wall of the tube. This phenomenon, called fingering displacement, is created by a combination of viscous instability between the two fluids and the no-slip condition at the tube wall. Taylor [4] studied such a phenomenon by injecting air into a horizontal tube filled initially by a viscous fluid. A fraction m, which is the fraction of viscous fluid left on the wall behind the finger, was defined as:

$$m = 1 - U_{\mathrm{m}}/U_{\mathrm{tip}},$$

where U_{tip} is the propagation speed of the finger.

For immiscible fluids, the appropriate control parameter that describes the viscous fingering is the Capillary number, *Ca*, the ratio of viscous to surface tension forces. Taylor [4] found the fraction m increased as $Ca^{1/2}$ for very small *Ca* when $Ca < 0.1$, and then for large *Ca*, asymptotes to a constant value of 0.56. In a later experiment, Cox [5] extended [4] to an even larger *Ca* and found an asymptotic value of 0.60. Corresponding numerical simulation by Reinelt and Saffman [6] agreed well with these experiments.

For miscible fluids with a finite viscous ratio, the fraction m can be defined as above and represents the volume of viscous fluid left on the wall behind the finger tip (see Appendix). The interfacial tension between two fluids tends to zero and the Peclet number replaces *Ca* as one of the governing parameters. The other is the Atwood number, $At = (\mu_2 - \mu_1)/(\mu_2 + \mu_1)$, which is a measure of the viscosity contrast between the two fluids. PM extended the experiment of [4] to miscible fluids by using various glycerine/water mixtures. For large Peclet number ($Pe > 10^4$), the fraction m asymptoted a value that depended only on the Atwood number, *At*. When *At* was close to unity the fraction m increased to a value of 0.61, which was consistent with the value found by Cox [5] for immiscible fluids. Also, their results for large *Pe* agreed very well with corresponding simulations conducted by Chen and Meiburg [7, hereinafter CM]. For small Peclet number ($Pe < 10^3$), PM found that the value of m depended strongly on both the tube diameter and orientation since the two fluids had different densities. A gravitational parameter $F = gd^2\Delta\rho/\nu_2 U_m \rho_2$, which is a measure of the relative importance of gravitational and viscous forces, was introduced to quantify these gravitational effects. Here g is gravitational acceleration, $\Delta\rho = \rho_1 - \rho_2$ the density difference, ρ_2 and ν_2 the density and kinematic viscosity of displaced fluid, respectively. Their results showed an increase of m for $F > 0$ and a decrease for $F < 0$. The fraction m was found to be a useful parameter to categorise the flow patterns generated by the finger motion. This was hypothesized originally by Taylor [4] and extended by PM. They suggested two probable streamline patterns in a reference frame moving with the finger tip. For $m > 0.5$, there was only one stagnation point at the finger tip. While for $m < 0.5$, there are two stagnation points on the centreline, one at the finger tip and the other within the finger, together with a stagnation ring on the finger surface. For this case, PM argued that there must be toroidal recirculation inside the finger head. Moreover, their experiment, and a later experiment by Lajeunesse et al. [8] and simulations by CM, showed a thin 'spike' at the tip of the 'mother' finger for this case. Details of direct measurement of these flow patterns are given in Kuang, Maxworthy and Petitjeans [9], while in the present experiments we concentrate on detailed measurements of m as a function of *Pe*, F and *At* in capillary tubes using silicone oils as the working fluids. One advantage of this is that silicone oils with large viscosity ratios invariably have small density differences so that experiments could be performed to lower velocity (smaller *Pe*) than in PM.

2. Experimental apparatus

The present experimental apparatus is similar, but not identical, to that of PM. A sketch for the case of a horizontal tube is shown in Fig. 1. It consisted of a precision-bore glass tube of 1 m long with an inner diameter of 1, 2, 3 or 4 mm. The

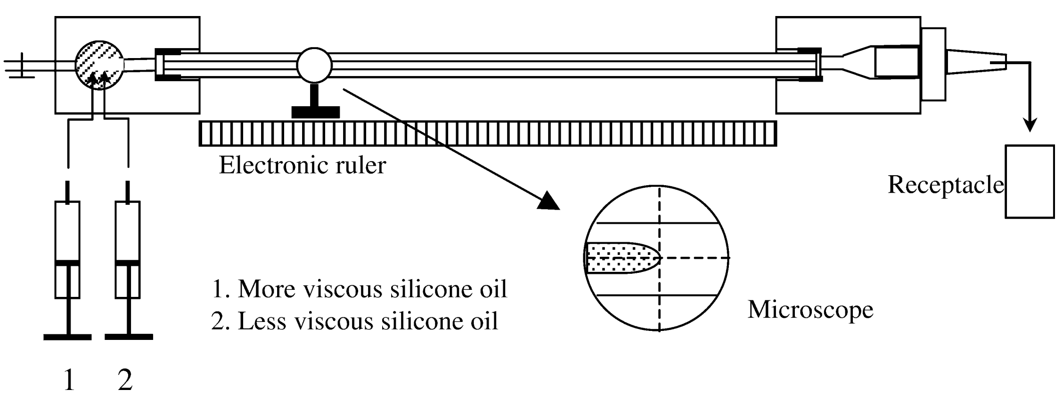


Fig. 1. Sketch of the experimental apparatus for the measurement of fraction, m (horizontal case).

tube was first filled with more viscous silicone oil, and then, less viscous silicone oil was introduced into the tube to displace the more viscous one, using a precision Hamilton glass syringe driven by a precision Cole–Parmer syringe pump to provide accurate control of the desired flow rates. The less viscous silicone oil was seeded with tiny particles of about 5 μm diameter so that the mobile finger could be observed. The outlet of the tube was extended using a flexible tube to a fluid receptacle. The finger front of the less viscous fluid was observed with a low-powered microscope mounted onto an electronic ruler that gave the position of the interface to an accuracy of ±0.5 mm. A stopwatch was used to record the time for each measured finger front position. An averaged celerity of the finger front was then calculated to give U_{tip}, the tip velocity of the less viscous fluid displacement. The flow ahead the finger front has a Poiseuille profile to give a mean velocity U_{m} for a given flow rate and tube inner diameter. Finally, it was possible to calculate the fractional amount $m = 1 - U_{\text{m}}/U_{\text{tip}}$. Experiments were run for silicone oil pairs 1000 cs/10 cs and 1000 cs/100 cs with $At = 0.98$ and 0.82, respectively. These Atwood numbers are close to the values of 0.985 and 0.79 used by PM for the glycerine–water system. The tubes were oriented horizontally and vertically to investigate gravity effects, especially at small Peclet number. For vertical displacements, the less viscous fluid was injected either upward or downward to study either a gravitationally unstable or a stable fingering displacement.

3. Results and discussion

Fig. 2 (a) and (b) show measurement results of m as a function of Pe for $At = 0.98$ (silicone oils 1000 cs/10 cs) and 0.82 (silicone oils 1000 cs/100 cs), respectively. When Pe is large, e.g., $Pe > 2 \times 10^4$, diffusion and gravity effects become very small ($F \to 0$) and viscous forces become dominant. The fraction m for different tube diameters and orientations tends to a constant value that depends on At. From the figures we have the constant value of $m = 0.6$ for $At = 0.98$ and $m = 0.56$ for $At = 0.82$. These values agree reasonably well with the data from PM and CM. When Pe is small, e.g., $Pe < 2 \times 10^4$, viscous forces become less important and diffusion and gravitational effects are dominant. The results in Fig. 2 show a large variation of fraction m with Pe in which F varies for each measurement. Solid and dotted lines plotted in Fig. 2 (a) and (b) are the fitted results of PM for glycerine–water mixtures, and they also agree reasonably well with the present measurements using the same diameter tubes. Note that the Atwood numbers are a little different for silicone oils 1000 cs/10 cs ($At = 0.98$) and 1000 cs/100 cs ($At = 0.82$) in the present experiment and for glycerine–water mixtures of 55.69% ($At = 0.985$) and 86.135% concentration ($At = 0.79$) in PM's work, respectively. However, the influence of this difference appears to be too small to be discernable in Fig. 2. Numerical simulation results by CM for $At = 0.987$ are plotted in Fig. 2(a) as well. Their data give an approximately constant value of m at Pe of order $O(10^3)$, which is quite different from the experiments. This result will be discussed later. Fig. 2 shows that the Peclet number in the present experiment could be as low as about 300, which was much smaller than that of PM. For an even slower displacement, the finger was very hard to distinguish and no good data was obtained.

For the case of gravitationally stabilizing fingering displacement ($F < 0$), i.e., a lighter and less viscous fluid displacing a heavier and more viscous fluid downwards, the more viscous fluid on the tube wall behind the finger tip has a tendency to move downward due to gravity. For a slower displacement, i.e., a smaller Pe, the heavier fluid has more time to move downward, leading to a larger finger diameter so that m is smaller. For the extreme case $Pe = 0,\ F \to -\infty$, a flat interface with $m = 0$ results. Consequently, the fraction m has a value $0 \leqslant m \leqslant (m)_{\max}$ for negative F, where $(m)_{\max}$ is the value of m as $Pe \to \infty$ and $F \to 0$. PM & CM gave a curve of $(m)_{\max}$ as a function of At as shown in Fig. 3 with the present results added. For $At < 0.5,\ (m)_{\max} \approx 0.5$, which indicates a Poiseuille flow in the tube. For higher $At,\ (m)_{\max}$ increases to a value of 0.61. Then for all values of At greater than approximately 0.5, two flow patterns may exist depending on whether $m < 0.5$ or $m > 0.5$, as indicated by [1,4] and [9] experimentally and by [7] computationally.

For the case of a gravitationally destabilizing fingering displacement ($F > 0$), i.e., a lighter and less viscous fluid displacing a heavier and more viscous fluid upwards, gravity drives the heavier and more viscous fluid downward while the lighter and less viscous fluid rises. For small Peclet number, the heavier fluid behind the finger tip has more time to move down and accumulate on the wall so that the finger diameter is smaller and fraction m becomes larger. Due to this gravitational instability, the finger continues to propagate even when there is no displacing fluid injected ($U_{\text{m}} = 0$) so that, probably, the fraction m approaches but is always less than 1. For this case, we always have $m > 0.5$ and there exists only one flow pattern.

For the horizontal tube the orientation of the finger displacement was perpendicular to gravity so that, nominally, $F = 0$. However, there were still gravitational effects, as presented by the difference between the horizontal case (star symbols) and the interpolated values corresponding to $F = 0$ for vertical cases (closed circles) in Fig. 2. When the Peclet number was large ($Pe > 5 \times 10^3$ for a 2 mm tube), the fraction m was approximately a constant and equal to the result for vertical tubes, i.e., gravitational effects were small or $F \to 0$. For small Peclet number ($Pe < 5 \times 10^3$), the value of m changed due to both a gravitational influence (F) and a low Peclet number (Pe). The finger tried to remain symmetric at its tip, but the main body, following the tip, was observed to rise towards the upper part of the tube, especially for larger tube diameter, e.g., $d = 3$ or 4 mm. The finger was no longer axisymmetric in the tube and was often cut into two longitudinal parts (cf., PM).

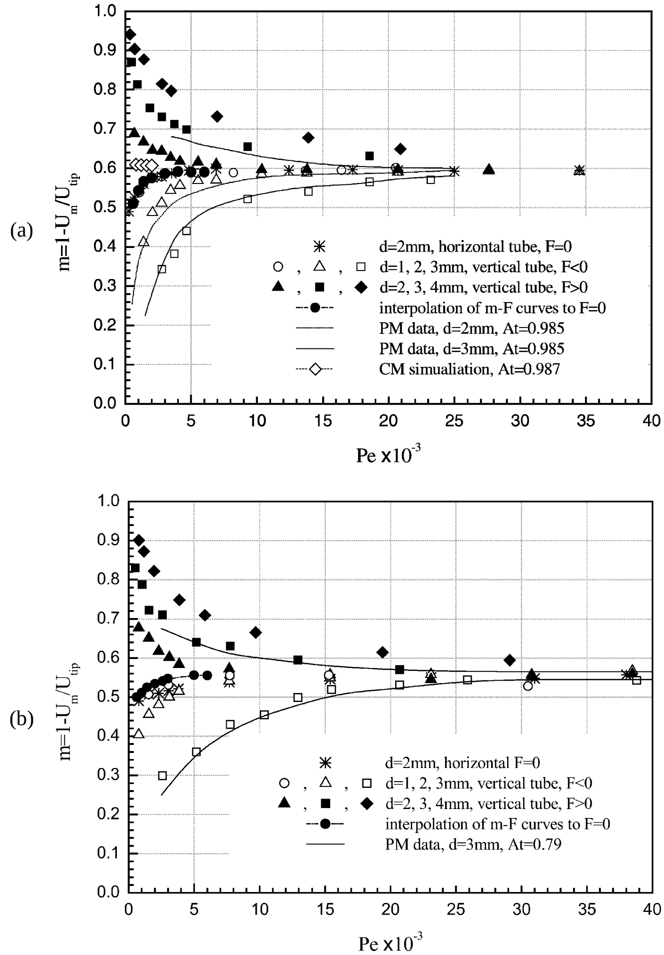


Fig. 2. Measurements of the fraction, m, as a function of *Pe*. The lines are the fitted results from PM Fig. 6. (a) $At = 0.98$, (b) $At = 0.82$.

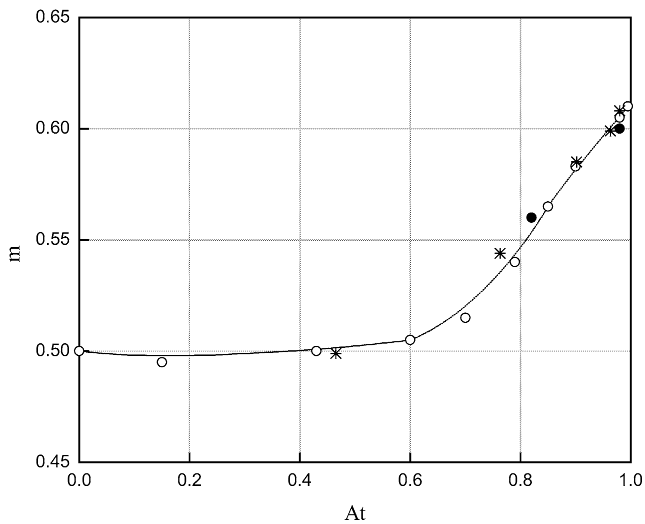
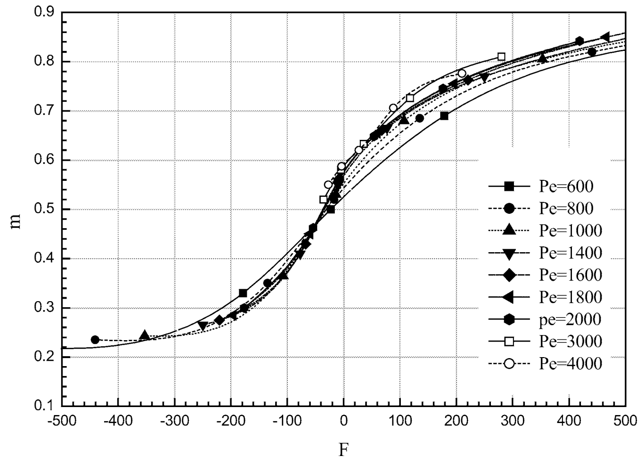
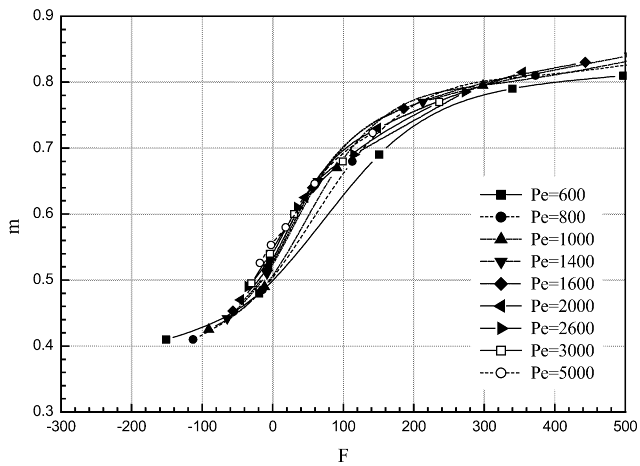


Fig. 3. Fraction m as a function of At at large Pe (from Fig. 13 of PM, with the present results added, $*$ CM's numerical results; $\circ$ PM's experimental data and $\bullet$ the present results).



(a)



(b)

Fig. 4. Fraction m as a function of the gravitational parameter F. The gravitational parameters were calculated for each tube diameter at a constant Peclet number from Fig. 2. (a) Silicone oils 1000 cs/10 cs, $At = 0.98$, (b) silicone oils 1000 cs/100 cs, $At = 0.98$.

The gravitational influence was tested more directly by cross-plotting the data of Fig. 2, at various constant values of the Peclet number, to generate curves of m vs. F, as shown in Fig. 4 (a) and (b) for $At = 0.98$ and 0.82, respectively. For $F > 0$, the gravitationally destabilizing case, we always have a fraction m greater than 0.5. With an increase of F, the fraction m increases. For $F < 0$, a gravitationally stabilizing case, the fraction m decreases when F decreases. Elementary arguments suggest that the finger front must tend to a flat surface, i.e., $m = 0$, when the injected velocity is very small and the gravitational parameter, F, becomes large and negative. Fig. 4 shows that the value of m is quite sensitive to the gravitational parameter when F is between -100 and 100. For $F = 0$, the intersections show that m increases as *Pe* increases.

The measure data presented in Fig. 2 shows strong gravitational effects for small Peclet number. A zoomed-in view of Fig. 2 is given in Fig. 5 for *Pe* less than 6000. For an upward and a downward displacement in a 2 mm diameter tube, the corresponding closed and open triangles show strong gravitation effects and the value of m changes with *Pe*. However, for a horizontal displacement in a 2 mm diameter tube and downward displacement in a 1 mm diameter tube, gravitational effects are weak. Interpolating data at $F = 0$ from Fig. 4 are also plotted on Fig. 5, and the curves are consistent with the data of the horizontal displacement in a 2 mm tube and the downward displacement in a 1 mm tube for $At = 0.98$ (Fig. 5(a)). Also plotted on Fig. 5(a) are the results of numerical simulation by CM using Stokes equations, a concentration dependent viscosity and constant diffusion coefficient but with no gravitational effects considered. Their data showed a constant value of m of about 0.61 for small *Pe* and $At = 0.987$. These simulations are significantly different from the experimental measurements for $F = 0$, indicating that another dynamical effects may be important at very small *Pe*.

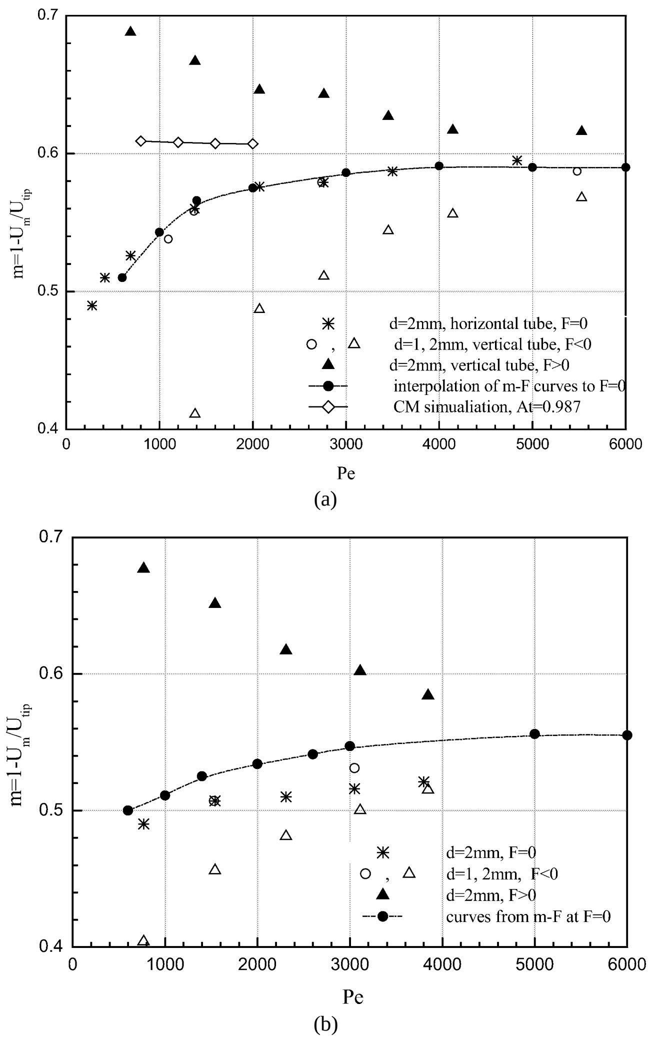


Fig. 5. Fraction m as a function of Pe for $Pe \leqslant 6000$. Including interpolated results at $F = 0$ from Fig. 4. (a) $At = 0.98$, (b) $At = 0.82$.

PM and CM noticed this difference and suggested adding additional concentration gradient dependent stresses (CDG stresses in what follows) to the Stokes equations. Joseph et al. [10] performed a theoretical study of the problem of miscible interfacial dynamics and showed that so-called Korteweg stresses [11] could be important in the region where the concentration gradients were large. In their most recent computational simulation, Chen and Meiburg [12] studied the influence of Korteweg stresses on the fingering displacement in capillary tubes. Their result shows the Korteweg stresses have significant influence on the dynamics of miscible displacement. However, our knowledge of such stresses is very limited. Paradoxically, as shown by CM, CGD stresses can become important at low Pe because they can rival in magnitude viscous stresses in a diffuse interface. Fig. 5(a) for $At = 0.98$ shows the diffusion becomes important when $Pe < 2000$.

4. Conclusions

An investigation of fingering displacement of miscible fluids in capillary tubes has been performed. Instead of glycerine/water in PM, fluid pairs with similar molecular structures, e.g., silicone oils, were selected as miscible fluids in the present experiment. The fraction, m, a measure of high viscous fluid left on the tube walls behind the finger, was measured, which was taken as a critical parameter to determine the streamline patterns of fingering displacement. Good agreement of the present results with PM indicates that the molecular structure of the miscible fluids has little influence on the behaviour of the fingering displacement.

The dependence of m on the independent, control variables Atwood number, *At*, Peclet number, *Pe*, and gravitational parameter, F, has been determined for two silicone oil pairs of 1000 cs/10 cs and 1000 cs/100 cs, corresponding to $At = 0.98$ and 0.82. Data for Peclet numbers as low as approximately 300 were obtained. At large *Pe*, m tends to a constant value that depends only on *At*. For $At = 0.98$ and 0.82, we have $(m)_{\max} = 0.6$ and 0.56, respectively. For small *Pe*, m depends on all the parameters. In particular the interpolation of the curves m vs. F to $F = 0$, shows that m tends towards small values at small *Pe*. A significant difference between these experiments and the numerical simulation of CM at small *Pe*, using only the Stokes equations, suggests that additional CGD stresses may be important at the diffusing interface. Due to these additional stresses it seems that m does not tend to the value of 0.5 that one would expect in a flow dominated by diffusion, which should asymptote a single-fluid Poiseuille flow. This latter result is somewhat surprising since one might expect CGD stresses to be more important at large *Pe* where the concentration gradients are large. However, as explained in CM, it appears that the CGD stresses become relatively more important than the viscous stresses at low *Pe*, even though their absolute magnitude decreases.

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Appendix

Assume a finger propagating in a tube at a constant speed. The tube, with an inner diameter of d, is filled with more viscous fluid. Less viscous fluid is injected into the tube at a constant flow rate, Q. By time t, the finger has propagated in the tube to a distance L. The volume of injected less viscous fluid is $V_L = Qt$, while the total volume behind the finger tip is $V_T = \pi d^2 L/4$. The volume of more viscous fluid left on the wall behind the finger tip is $V_M = V_T - V_L$. Then the fraction m is defined as:

$$m = \frac{V_M}{V_T} = 1 - \frac{V_L}{V_T} = 1 - \frac{Qt}{\pi d^2 L/4}.$$

For an incompressible fluid in a cylindrical tube, we have the flow rate $Q = \pi d^2 U_{\mathrm{m}}/4$ and the finger propagating speed $U_{\mathrm{tip}} = L/t$, which leads to:

$$m = 1 - U_{\mathrm{m}}/U_{\mathrm{tip}}.$$

Note that this result is independent of the viscosity ratio. In the limit $At \to 1$ the displaced fluid left on the tube wall behind the finger tip is stationary and m can then be related to the constant thickness of that layer, as in Taylor [4]. In general this is not true and m can only be used as a diagnostic measurement of the volume of displaced fluid left behind the advancing finger.

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