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Immiscible Liquid Displacement In A Capillary Tube: The Moving Contact Line

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The internal deformation of two immiscible liquids as one displaces the other on flowing through a circular capillary tube is examined. The existence of a toroidal-like flow adjacent to the moving interface is documented. These observations may be relevant when liquid-liquid systems are modeled in which a surfactant is present, as in the case of oil recovery. Measurements of this flow may also lead to a better understanding of the moving contact line.

Place a glass capillary tube vertically over a dish of water. The moment the lower end of the tube touches the air-water interface, a column of water (one immiscible fluid) rises up the tube and displaces the air (the other immiscible fluid) until it reaches an equilibrium height. The dynamics of the displacing fluids has been studied over the years with particular emphasis placed on liquid-gas systems. For these systems it has been found that the details of the fluid motion in the region immediately adjacent to the fluid-fluid interface can essentially be ignored when the speed at which the interface travels through the tube is analyzed. [G. D. West (1911-1912) was the first person to successfully model the flow in a capillary tube and predict the speed of propagation of the fluid-fluid interface. He assumed that the fluids undergo Poiseuille flow up to a point about two tube diameters to either side of the fluid-fluid interface. In the region near the interface the only significant pressure drop was assumed to come from its surface tension and curvature. By assuming the curvature of the interface was known, he avoided having to do any detailed analysis in the region. Others who rediscovered this include E. W. Washburn (1921) and G. D. Yarnold (1938).] However, for systems consisting of two immiscible liquids and a soluble surfactant, such simplifying assumptions may no longer be appropriate. The surfactant, in order for it to be effective, must be present at the liquid-liquid interface where it must reduce the interfacial tension to an exceedingly small value. This essentially eliminates the capillary force, whose presence, for example, impedes the movement of small slugs of oil trapped within porous rock. Does the surfactant remain at the liquid-liquid interface, or is it pushed off and forced onto the walls of the solid matrix? Is the surfactant readily accessible to the interface, or is it depleted from the surrounding

liquid thus reducing the rate at which additional surfactant can be adsorbed onto the interface? It is evident that the motion of the liquids in the region surrounding the interface is an important factor.

In this note the motion of the fluids in the vicinity of the fluid-fluid interface is examined in detail for a system consisting of two immiscible liquids flowing through a circular capillary tube. It is shown that one fluid undergoes the familiar fountain type of motion, while the other fluid contains a region adjacent to the interface where in a more complicated toroidal-like motion occurs. This latter motion has not been previously observed. [In an attempt to explain the experimentally observed fact that the curvature of the interface is constant for slow flow, Rose (1961) postulates a different flow field].

There is a more fundamental reason for studying this system. A contact line moving over a solid surface (for systems consisting of either two immiscible liquids, or a liquid and a gas) occurs in countless natural and industrial processes; however, to this day the dynamics of the moving contact line is not understood. One approach has been

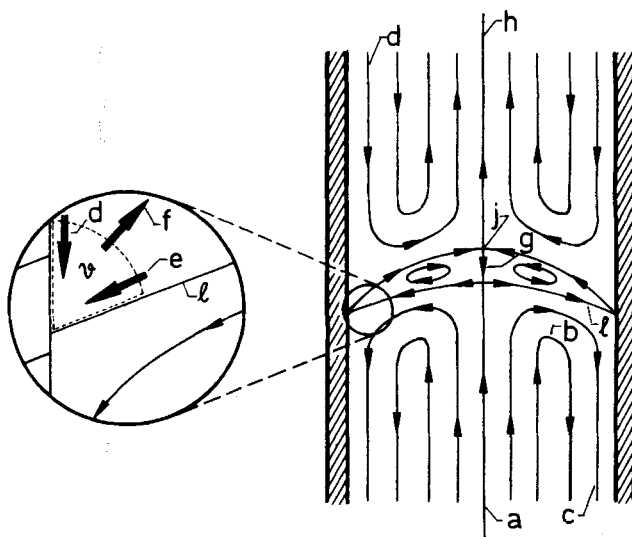


Fig. 1. Conceptualization of flow field.

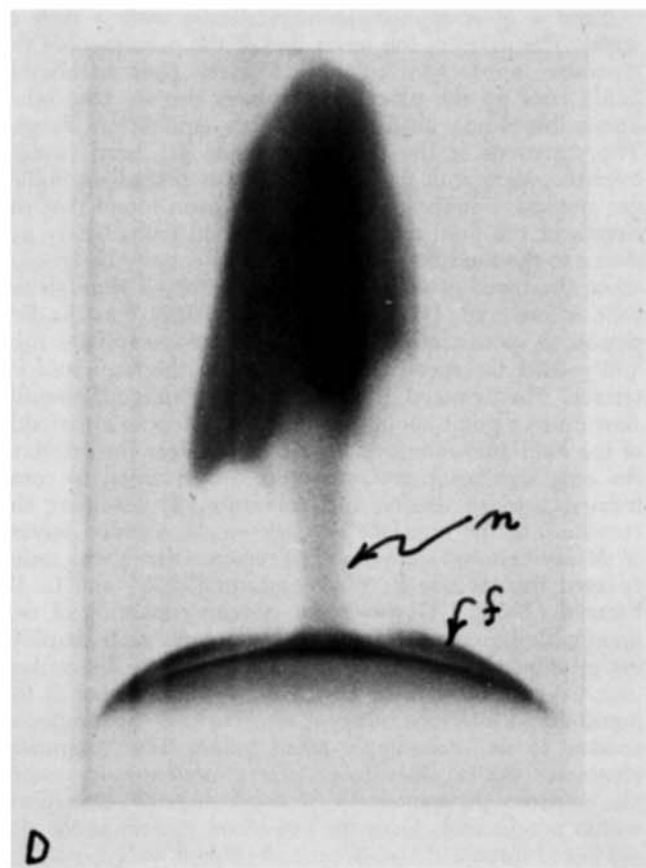
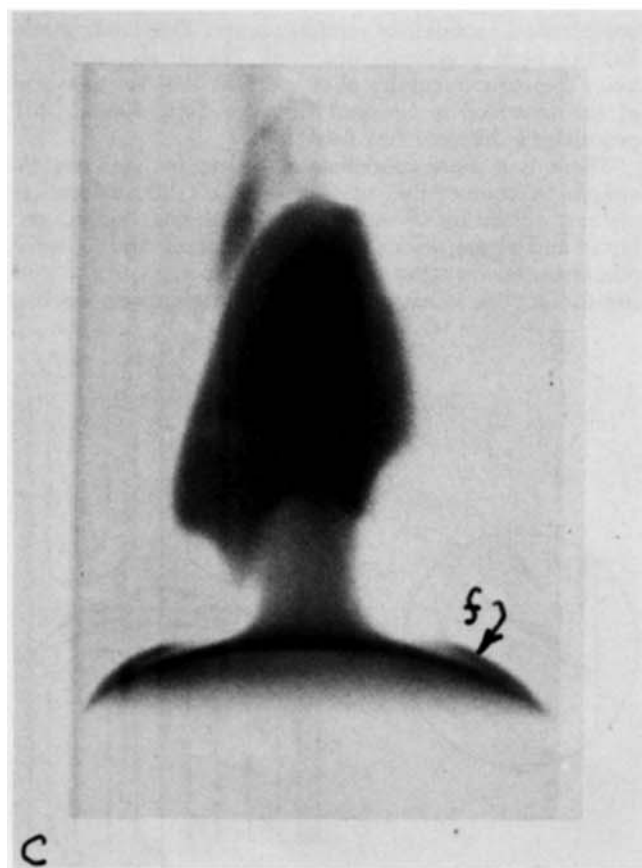
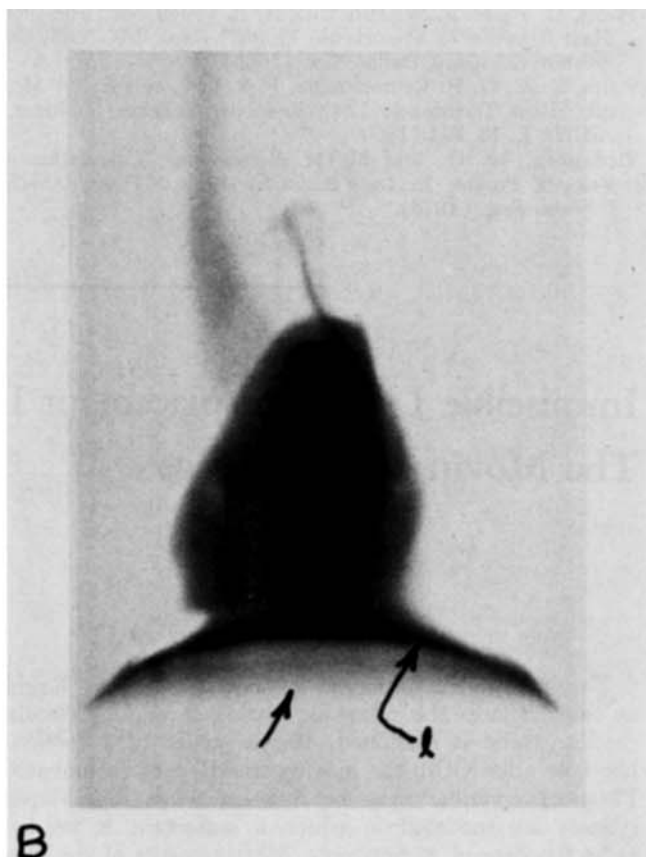
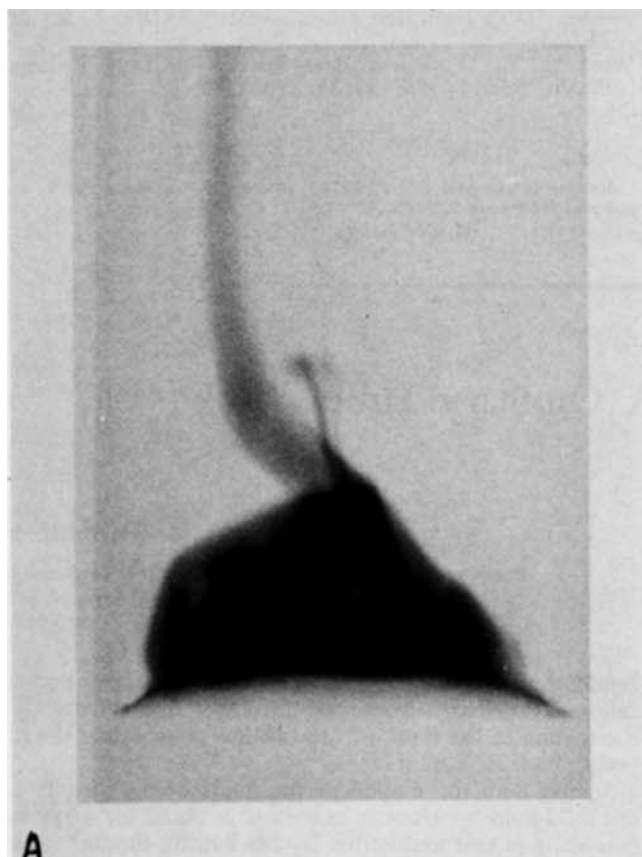


Fig. 2. Visualization of flow with dye mark.

to consider the contact line, on the continuum level, as a three-phase line, but this has led to analyses with physically unacceptable singularities [Huh and Scriven, (1971); Hansen and Toong, 1971]. Recently it has been shown that this difficulty arises from the fact that this model is not self-consistent [Dussan V. and Davis (1974). One way of avoiding the whole problem is to deny the existence of the three-phase line and assume that a thin film of the displaced fluid is left on the walls; refer to Prutow and Ostrach, and Ludviksson and Lightfoot (1968).] Flow in the capillary tube presents a tractable system from both an experimental and analytical point of view for searching for the proper modeling assumptions. Since the radius of curvature of the fluid-fluid interface is directly controlled by the moving contact line, one might anticipate that it also controls the size of the toroidal region reported on below; hence, its measurement may prove to be of fundamental value.

The system examined consists of glycerine, the more viscous and lower fluid, displacing mineral oil in an 0.00635 m inner diameter Plexiglas circular tube. (The approximate speed of the contact line is 3×10^{-5} m/s. The viscosities of the glycerine and mineral oils are 1.5 and 0.13 Pa · s, respectively.) All observations are made from a frame of reference at rest with respect to the interface. From this vantage point the interface is stationary and the tube walls are moving downward, Figure 1. It is found with the aid of dye that the lower fluid undergoes the familiar fountain type of motion; that is, there is an upwelling of fluid along the axis of the tube *a*, a radial outward flow at and just below the interface *b*, and a downward motion near the walls of the tube *c*. [This type of motion has been reported by a number of people, for example, Yarnold (1938).] In order for the upper fluid to undergo a motion consistent with that of the tube wall and lower fluid, one must postulate the existence of a toroidal-like motion in the region directly above and immediately adjacent to the interface. An explanation is: the upper fluid residing in the vicinity of the tube wall *d* is dragged in a downward direction and some of this fluid is forced to enter the pie shaped control volume *v*. Fluid is also dragged into *v* at *e* owing to the radial outward motion of the lower fluid at the interface. In order to keep the amount of mass in the control volume constant (liquids are relatively incompressible), there must also be an outward flow. The only remaining place in which this can occur is at *f*. To complete the picture, a downward motion must exist at *g* to supply the radially outward flow at *e*; however, far above the interface *h* the fluid must be in an upward motion. For these motions to be compatible a stagnation point must be located within the upper fluid (Prutow and Ostrach propose a similar flow field.)

The flow is visualized by injecting a dye mark composed of mineral oil and Sudan-Black B into the upper fluid at the center of the interface. (Care must be taken to select a dye which is not surface active. It was found in the system described above that the presence of the Sudan Black in the mineral oil did not cause a detectable change in the glycerine-oil interfacial tension.) The photograph in Figure 2*a* was taken moments after injection. Dye has been dragged along the interface from its center to its edge by the lower fluid; note the thin layer of dye covering the interface. The sequence of photographs shows that the upper part of the dye is swept upstream and hence lies outside the toroidal region. Figure 2*b* shows that the layer of dye on the interface has thickened. The perspective in the photographs is slightly different from that of Figure 1. In the photographs we are looking through a tube. The fluid-fluid interface bulges into the upper fluid; hence, in Figure 2*b* the lower arrow points to that part of the contact line (the intersection of the fluid-fluid interface and

the tube wall) which lies along the front part of the tube (out of focus), and *l* points to the uppermost part of the fluid-fluid interface (this is the same *l* as in Figure 1). The area lying between the contact line and *l* is a portion of the fluid-fluid interface. Figure 2*c* shows quite distinctly that dye has traveled from the contact line back into the interior of the upper fluid *f*. In Figure 2*d* we can see the entire upper surface of the toroidal flow *f*. The spikelike formation *n* is due to the combination of radial inward flow towards the stagnation point *j* and upward flow at *l*; compare Figure 2*a* with Figure 2*d*. (Upon reversing the direction of flow, that is, the tube moves in an upward direction in Figure 1, one would expect to produce a similar flow but with the arrows of the stream lines pointing in the opposite direction. However, the inward flow encourages surface active impurities to collect at the fluid-fluid interface which then eliminates, for the most part, any radial motion; hence, the toroidal motion is not observed.)

Thus it has been shown, for a system of two immiscible flows which forms a contact line that can move along the inside surface of a circular capillary tube, that within one of the fluids, there is a region adjacent to the interface (the toroidal-like flow) which always consists of the same material. It is not at all obvious that this always occurs in the less viscous fluid. [The important feature of the materials used (the two liquids and solid) is that the contact line can move along the solid surface. This is not true for any combination of the two immiscible fluids and solid surface; however, the above choice of materials is not unique. Also, at higher rates of displacement or as a consequence of gravity, this flow may be unstable and the interface may not have a stationary form; under such conditions, the above description would not apply.]

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