

# Rise of Liquids and Bubbles in Angular Capillary Tubes

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Received January 12, 2001; accepted November 24, 2001

We discuss the rise of a liquid inside an angular capillary tube. It is shown that for a wetting liquid, the height of the rise is (as usually) inversely proportional to the length which characterizes the confinement. The exact laws deduced from energetic considerations are found to be in excellent agreement with the data. We then show how such tubes can be used to prevent bubbles from being trapped. The rising velocity of a bubble is finally discussed, in the particular case of a square tube. © 2002 Elsevier Science (USA)

**Key Words:** capillary rise; bubble trapping.

## INTRODUCTION

Porous materials exhibit various structures (sponge-like, assembly of fibers, packed spheres, etc.), and it is interesting to know to what extent the classical laws for a capillary tube remain valid in such geometries, and how they must be adapted. Here, we consider the simple case of capillary rise in a medium having corners, a question of current interest because of the development of microfluidics, where square or rectangular channels are used for driving liquids. We successively study the rise in a square tube, and along fibers closely packed (taking into account the nature of the packing). We finally describe an application of such model systems, namely the possibility for preventing bubbles from being trapped, as often occurs in circular capillary tubes.

## RISE IN A DIHEDRON

If a corner formed from two solid plates is put in contact with a wetting liquid, a meniscus generally rises inside the corner (Fig. 1). Different questions can be addressed, such as the conditions for observing a rise and the final height and shape of the meniscus.

Neglecting the curvature of the liquid/air interface in the plane ( $x, z$ ), a balance between the Laplace pressure (related to the confinement of the liquid in the dihedral) and the hydrostatic pressure can be written,

$$\frac{\gamma}{R} = \rho g z, \quad [1]$$

$R$  is the radius of curvature of the interface at a height  $z$ ;  $\gamma$  and  $\rho$  are the liquid surface tension and density; and  $g$  is the acceleration of gravity.  $R$  is geometrically related to the distance  $d$  between the corner and the interface,

$$d = R \left( \frac{\cos \theta}{\sin(\alpha/2)} - 1 \right), \quad [2]$$

where  $\alpha$  is the corner angle and  $\theta$  the contact angle made by the liquid on the solid. The distance  $d$  is positive (which means that the liquid rises) if the angles verify the classical relation of Concus and Finn (1):

$$\alpha + 2\theta < \pi. \quad [3]$$

For  $\alpha \rightarrow 0$ , we find the classical condition of impregnation of a porous medium ( $\theta < \pi/2$ ). Note also that a rise is possible only for  $\alpha < \pi$ .

The liquid behavior inside the dihedral depends discontinuously on the value of  $\alpha + 2\theta$ . If the latter quantity is smaller than  $\pi$ , there is no solution of the Young–Laplace equation meeting the solid surface with the prescribed contact angle  $\theta$  and the liquid fingers rise (ideally) to infinity (1). Putting together Eqs. [1] and [2] shows that the profile of the interface in the plane ( $x, z$ ) is hyperbolic (1), as reported in 1712 by Taylor (the one of Taylor's expansions) and Hauksbee (2, 3),

$$d(z) \sim \frac{\kappa^{-2}}{z}, \quad [4]$$

where  $\kappa^{-1}$  is the capillary length ( $\kappa^{-1} = (\gamma/\rho g)^{1/2}$ ).

Practically, the height does not diverge because of the finite curvature of the corner at a small scale. Ramos and Cerro (4) even deduced from this remark a method for measuring the contact angle of the liquid on the tube wall. Nevertheless, the rise can be very high, and the phenomenon is amplified in a low-gravity environment, as shown by Weislogel and Lichter (5): then, the liquid fingers invading the corners can mobilize all the liquid. Tang and Tang (6) recently improved the calculation by taking into account the curvature of the liquid/vapor interface in the plane ( $x, z$ ). The liquid surface is found to be slightly raised as compared with Eq. [4], which is logical since the considered curvature has the same sign as the largest one, and thus also contributes to the rise.

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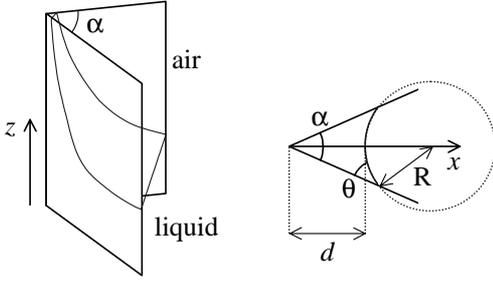


FIG. 1. Side and top views of a dihedral put in contact with a bath of wetting liquid.  $\alpha$  is the corner angle,  $\theta$  the contact angle of the liquid on the solid, and  $R$  the radius of curvature of the meniscus at a height  $z$ .

## SQUARE TUBES

### 1. Experiments

We now consider a glass tube of square section ( $\alpha = 90^\circ$ ) brought in contact with a reservoir of liquid (Fig. 2). According to Eq. [3], the liquid should invade the corner if the contact angle is smaller than  $\pi/4$ . In addition, it is observed experimentally that a meniscus stands at the center of the capillary (height  $h$ ), as in usual capillary rise. The fingers progressing along the corners can easily be detected with a blotting paper placed at the top of the tube (i.e., much higher than the position of the central meniscus): for wetting liquids, it is indeed observed that after a while, the paper becomes impregnated.

We have measured the level  $h$  reached by the visible central meniscus, using as a liquid cyclohexane which totally wets the glass ( $\theta = 0^\circ$ );  $h$  is plotted in Fig. 3 as a function of the inverse of the (inner) size  $a$  of the tube. It is found to vary linearly with the degree of confinement, as in usual capillary rise. The interesting

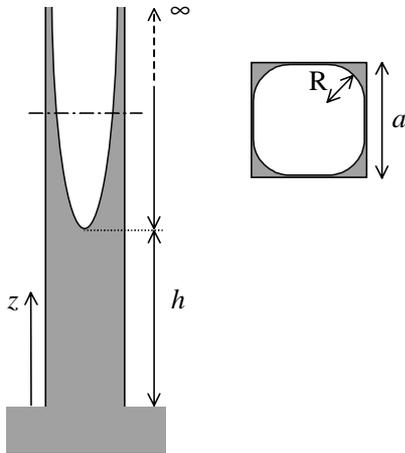


FIG. 2. Liquid rise inside a square tube (side view and cross section above the height  $h$ , showing the fingers along the corners). The main central meniscus rises to a height  $h$ , measured in Fig. 3. Above  $h$ , fingers develop along the corners under the action of the Laplace pressure associated with the radius of curvature  $R(z)$  of the liquid/vapor interface in the plane of the cross section.

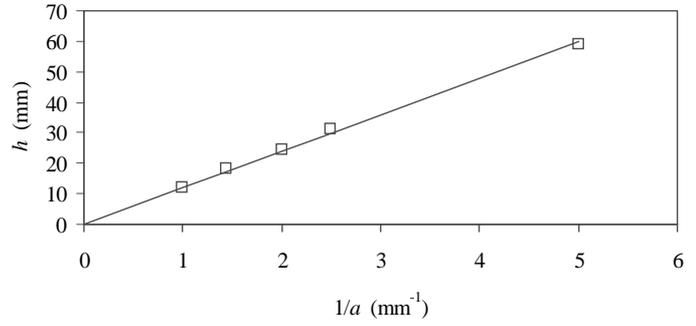


FIG. 3. Height  $h$  of the central meniscus (defined in Fig. 2) in a square tube of inner size  $a$ . The liquid used is cyclohexane, of surface tension  $\gamma = 25.5$  mN/m and density  $\rho = 820$  kg/m<sup>3</sup>. The straight line is a fit to Eq. [7].

question here is to determine and understand quantitatively the slope of this variation.

### 2. The Princen Model

Princen has proposed to model the rise in the following way (7, 8). For a capillary invariant by a translation, the height results from a balance between capillary force and gravity. Considering an infinitesimal displacement of the contact line along the axis gives the constant capillary force of the process,

$$F = p\gamma \cos \theta, \quad [5]$$

where  $p$  is the perimeter of a section of the tube ( $4a$  for a square).

Considering the weight of the liquid, we must take into account the liquid column up to the central meniscus, and the fingers along the corners above (Fig. 2). Hence the total weight  $W$  of the liquid can be written,

$$W = \rho g h a^2 + 4\rho g \int_h^\infty (1 - \pi/4) R^2(z) dz, \quad [6]$$

where the radius of curvature  $R$  obeys Eq. [1] (the other radius of curvature is neglected). Thus, the weight can be easily calculated. Balancing  $W$  with  $F$  finally yields the equilibrium height  $h$  for the central meniscus,

$$h = (2 + \sqrt{\pi}) \frac{\kappa^{-2}}{a}, \quad [7]$$

where we have taken  $\theta = 0$ . The straight line drawn in Fig. 3 is a fit to Eq. [7] ( $2 + \sqrt{\pi}$  is about 3.77) and is found to be in excellent agreement with the data. A much quicker argument for deriving the height would consist of reducing the weight to the contribution of the central meniscus, which immediately yields  $4\kappa^{-2}/a$  for the height  $h$ . The fingers' contribution is thus found to lower this quantity by about 6%. Note finally that the calculation supposes an infinite extent for the fingers, although the tubes used for experiments have a length of the order of 10 cm. Varying the total height of the tube, we observed that

the central meniscus keeps the same height (taking a shorter tube does not make the meniscus rise), which could be due to the possibility for the liquid to adjust its curvature on the sharp edges at the extremity of the tube.

### ASSEMBLIES OF FIBERS

The same kind of arguments can be used to describe the imbibition in an assembly of fibers (7, 8), as may exist in fabrics or in brushes. We restrict our study here to the case of close-packed fibers (of radius  $b$ ), and consider triangular and square lattices (Fig. 4). The liquid is still taken to be wetting ( $\theta = 0$ ).

For a triangular lattice, the perimeter of each interstice is  $\pi b$ , so that the capillary force  $F$  (per interstice) is given by the relation,

$$F = \pi b \gamma. \quad [8]$$

The weight of liquid (per interstice) still decomposes in the central meniscus contribution plus the fingers one, which can be written,

$$W = (\sqrt{3} - \pi/2) \rho g h b^2 + 3 \rho g \int_h^\infty S(z) dz, \quad [9]$$

where the section  $S$  is

$$S(z) = b^2 \left( \sqrt{\frac{R^2(z)}{b^2} + 2 \frac{R(z)}{b}} - \text{Arc cos} \frac{b}{R(z) + b} - \frac{R^2(z)}{b^2} \text{Arc sin} \frac{b}{R(z) + b} \right). \quad [10]$$

The radius of curvature  $R(z)$  is still given by Eq. [1]. Solving the equation  $F = W$  leads to the following expression for the height of the central meniscus:

$$h = 11.32 \frac{\kappa^{-2}}{b}. \quad [11]$$

For a square lattice, the section of each interface is twice larger than previously, which gives for the capillary force,

$$F = 2\pi b \gamma. \quad [12]$$

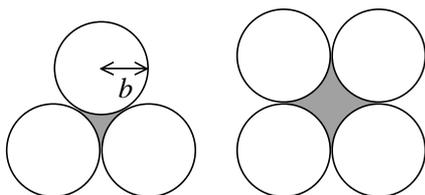


FIG. 4. Two types of close-packed assemblies of fibers or rods (of radius  $b$ ). The first lattice is taken to be triangular (maximum close packing), and the second one square.

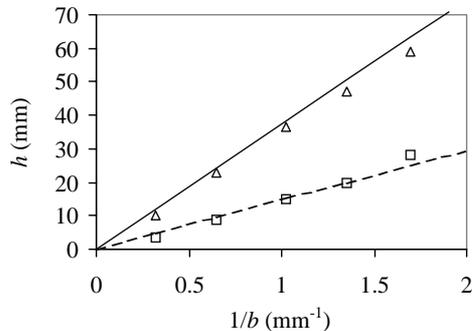


FIG. 5. Height of the central meniscus inside an assembly of close-packed cylinders of radius  $b$ . The lattice can be either triangular (maximum packing) or square, the corresponding data being respectively symbolized by triangles and squares. The full line is a fit to Eq. [11] and the dotted one Eq. [14].

The weight per interstice is also larger,

$$W = (4 - \pi) \rho g h b^2 + 4 \rho g \int_h^\infty S(z) dz, \quad [13]$$

where  $S(z)$  is given by Eq. [10]. Again, the equilibrium height is obtained by solving the equation  $F = W$ . It naturally obeys the same scaling law as previously (Eq. [11]), but the numerical value is found to be significantly smaller:

$$h = 4.49 \frac{\kappa^{-2}}{b}. \quad [14]$$

We achieved such assemblies by packing millimetric rods together. The height of the meniscus inside such assemblies is plotted in Fig. 5 as a function of the inverse of the rod radius, using once again cyclohexane as a rising liquid. The data are compared with Eqs. [10] and [14], and a good agreement is found.

### RISE OF A BUBBLE IN A CLOSED TUBE

Let us now consider a tube closed at both ends, with a bubble at the bottom (Fig. 6). We consider a bubble of volume much larger

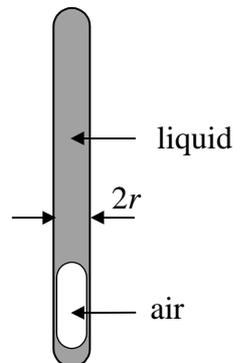


FIG. 6. Air bubble trapped (or possibly rising) in a closed tube of inner radius  $r$  filled with a wetting liquid.

than  $a^3$  (noting  $a$  the characteristic size of the tube), which forces it to elongate as drawn in Fig. 6. We still take as a liquid a wetting fluid ( $\theta = 0$ ), which remains on the wall. If this were not the case, the film would be likely to dewet, and contact lines would exist; together with the contact angle hysteresis generally associated with partial wetting, this would provide another reason for the bubble to be trapped.

In his classical paper on bubbles in a tube partially filled with a wetting liquid (9), Bretherton showed that a bubble is trapped if

$$r < 0.918\kappa^{-1}, \quad [15]$$

i.e., if the tube radius  $r$  is smaller than about the capillary length  $\kappa^{-1}$  ( $\kappa^{-1} = (\gamma/\rho g)^{1/2}$ ). This results from the fact that the liquid/air interface must deform for rising. Of course, this critical radius no longer exists if the tube is open.

In his approach, Bretherton did not take into account the possible presence of a microscopic wetting film around the bubble, which can connect both parts of the liquid as shown by Di Meglio (10). Such a film would allow the bubble to rise, but the velocity would be very small because of the thinness of the wetting films. We can evaluate a typical rise velocity. First, the thickness of the wetting film results from a balance between a Laplace pressure and a disjoining pressure, which leads to a thickness  $h$  of the order of 10 to 100 nm (10, 11). The velocity of gravitational drainage through such a film is given by a Poiseuille law, for a flow between a solid and a free surface,

$$V = \frac{h^2}{3\eta}\rho g, \quad [16]$$

where  $\eta$  is the liquid viscosity. The rising velocity is finally deduced from conservation of matter: it is of order  $hV/R$ , and thus is found to be extremely small. For typical values of the different parameters, the bubble velocity should be of the order of  $10^{-13}$  m/s (i.e., 10  $\mu$ m in 3 years)! These values can be larger if the tube walls are rough: because of the possible imbibition of the roughness, the two parts of the liquid can be connected by much thicker channels (12).

It can be of interest, in different applications, to find a solution to get rid of such bubbles in a short time. The simplest way consists of making thicker the film around them. This can be achieved by using angular tubes, which trap fingers in their corners, as emphasized above. The situation with a tube of square section is illustrated in Fig. 7.

We conducted a series of experiments with such a tube. The capillary was first filled with a wetting liquid (a silicone oil of viscosity  $\eta = 17$  mPa·s). A bubble was then introduced at the bottom of the tube, which was finally sealed. It was observed that the bubble rises at a constant velocity, of the order of 10  $\mu$ m/s. We also found that this velocity *does not vary* with the bubble length, for a bubble longer than the size  $a$  of the tube (experiments were done by varying the bubble length by about

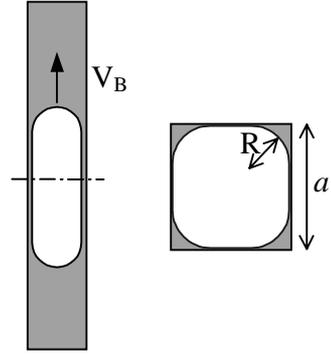


FIG. 7. Air bubble rising at a velocity  $V_B$  in a closed square tube of inner size  $a$  partially filled with a wetting liquid. The cross section shows the angular channels which help to drive the bubble.

a factor of 30, between 2 mm and 6 cm). The rising velocity  $V_B$  is plotted in Fig. 8 as a function of the size  $a$  of the tube. The smaller the size, the thinner the fingers in the corners and thus the slower the motion. Figure 8 indicates that  $V_B$  scales as  $a^2$ , in agreement with Eq. [16], providing that the mean thickness  $h$  of the liquid channel scales as  $a$ .

This behavior can be understood more quantitatively. Let us suppose that the liquid/air interface keeps a static shape despite the flow. This shape (and thus the radius of curvature of the free interface) can be deduced from a minimization of the surface of the bubble, which gives for a wetting liquid, as shown by Dong and Chatzis (13),

$$R = \frac{1}{2 + \sqrt{\pi}}a, \quad [17]$$

where the numerical coefficient is about 0.275. This radius of curvature is, of course, the same as the one calculated above for the law of capillary rise (Eq. [7]).

The drainage velocity is obtained by balancing viscous friction with gravity (inertia can very generally be neglected in such confined flows). Ransohoff and Radke determined the viscous

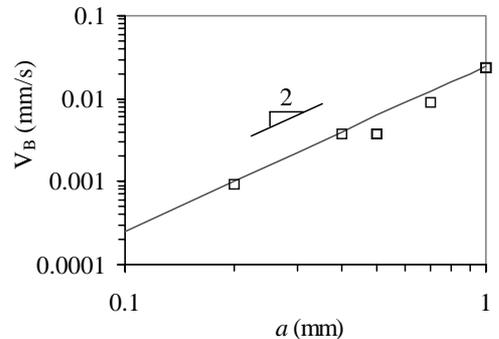


FIG. 8. Velocity  $V_B$  of an air bubble rising in a closed square tube (of inner size  $a$ ) partially filled with a wetting silicone oil of viscosity  $\eta = 17$  mPa·s and density  $\rho = 980$  kg/m<sup>3</sup>. The straight line is a fit to Eq. [20].

pressure drop along a corner (14). They found a Poiseuille-type law,

$$\nabla P = C \frac{\eta V}{R^2}, \quad [18]$$

where  $V$  is the mean velocity of the flow, and  $C$  a numerical constant which depends of the opening angle of the corner, and which is about 94 for a square angle. Balancing this pressure gradient with the gravity  $\rho g$ , we find the flow velocity,

$$V = \frac{\rho g R^2}{C \eta} = 7.5 \times 10^{-4} \frac{\rho g a^2}{\eta}. \quad [19]$$

The rising velocity for the bubble is easily deduced from conserving the flow rate. We thus obtain

$$V_B = 4.8 \times 10^{-5} \frac{\rho g a^2}{\eta}. \quad [20]$$

The latter relation can be directly compared with the experimental data in Fig. 8 (straight line). The agreement is quite good, although some values are smaller than the predicted value which could be related to the blunt nature of the corner as mentioned by Ransohoff and Radke (14). Note that in this approach, the flow was simplified by considering a quasi-static shape for the interface. This can be justified by the modest value of the capillary number  $Ca = \eta V / \gamma$ , which is written  $5 \times 10^{-5} (\kappa a)^2$ , and thus always remains smaller than  $10^{-5}$ . Bretherton (9), Ratulowski and Chang (15), and Thulasidas *et al.* (16) have described the situation where the bubble displacement is forced (for either a circular or square tubes). In square tubes, the corners fill all the more since the capillary number is high, which eventually produces an axisymmetric bubble at high forced velocity, as shown by Kolb and Cerro (17, 18). The pressure drop across the bubble scales as  $Ca^\alpha$  ( $\alpha$  ranging from 0.5 to 1.4 depending on the geometry) and is independent of the bubble length for long bubbles. If this pressure drop is balanced with a hydrostatic pressure through the bubble, the bubble velocity is found to be proportional to its length, which was not observed in our experiments. The difference between the two results results from immobility of the liquid slugs in sealed tubes. In the case of circular bubbles, the bubble velocity was calculated by Thulasidas *et al.* (Eq. [18] in (16)), and found to obey the same scaling as in Eqs. [18] and [19], but with a significantly larger numerical coefficient: in our case, as sketched in Fig. 7 and shown by Eq. [17], the radius of curvature of the fingers is smaller than  $a/2$ , which increases the friction of these nearly square bubbles compared with that of circular bubbles.

This discussion finally suggests other tricks for getting rid of trapped bubbles. For example, introducing a thin vertical fiber (radius  $b = 100 \mu\text{m}$ ) along the inside of a circular tube (radius  $r = 600 \mu\text{m}$ ) produces the same effect. In the latter example, the rising velocity was observed to be a constant, of the order of  $25 \mu\text{m/s}$  with a silicone oil of viscosity  $17 \text{ mPa} \cdot \text{s}$ . More generally, special defects (such as grooves, or threads) could be designed on the surface tube, for the cases where one wants to avoid bubble trapping in small capillary tubes.

## CONCLUSION

We have discussed the capillary rise in angular tubes. Our data were found to be in quantitative agreement with Princen's models, which suppose that the weight of the liquid can be decomposed in the weight of a central meniscus (the visible part of the rise) plus the one of the fingers which invade the corners. Because of the presence of these filaments, the final height was found to be smaller than the "classical" height of capillary rise (i.e., obtained when neglecting these fingers). We could also take advantage of these filaments for connecting two liquid regions in the tube (for wetting liquids). In this spirit, we showed that a long bubble does not remain trapped in a thin closed square tube (as it would be in a circular one), but rises because of gravity. The rise velocity was analyzed, which allowed us to measure the viscous friction associated with a flow inside the fingers. This friction was found to be in good agreement with the predictions of Ransohoff and Radke.

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