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Forces, pressures and energies associated with liquid rising in nonuniform capillary tubes

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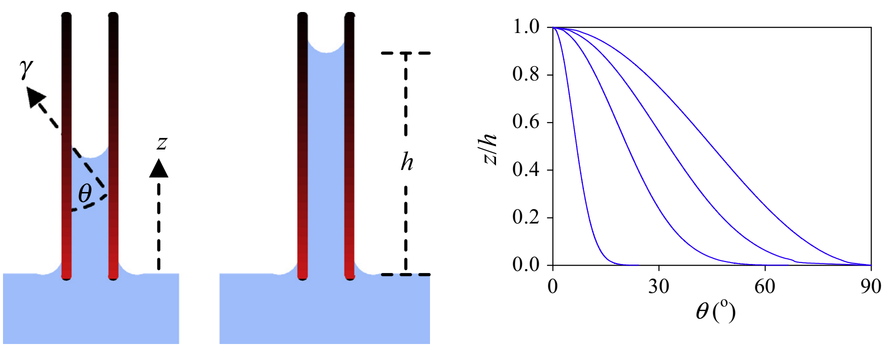
GRAPHICAL ABSTRACT



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ABSTRACT

In this theoretical study, the forces, pressures, energies and kinetics for liquid rising in three types of capillary tubes were analyzed: one type was chemically homogeneous and the other two were nonuniform with chemical gradients. The tubes with chemical gradients were "designed" such that the liquid would still rise and attain the same ultimate height as an equivalent homogenous tube, but as shown here, the energies and kinetics of these inhomogeneous tubes are anticipated to be quite different from their homogeneous counterpart.

1. Introduction

Some of the biggest names in modern science have studied and contributed to our understanding of capillary rise: da Vinci, Boyle, Newton, Young, Laplace, Gauss, Maxwell and Einstein, to name a few [1]. The earliest investigators attempted to elucidate the seemingly spontaneous rise of liquids in small diameter tubes [1a,2], but also, more broadly, the character and range of molecule interactions [1a,1e]. These pioneering studies were often framed in terms of forces and pressures. Analyses of energies [3] and kinetics [4] came later.

More recently investigators have considered capillary rise or imbibition in the absence of gravity [5], in tilted tubes [6], in non-circular tubes [7], in tapered tubes [8], in rough tubes [5b], in tubes where inertia dominates [4d,9], in tubes where the contact angle [5b,10] or viscosity [11] depends on rise velocity, in tubes with surfactants solutions [12], as well as in various types of porous media [13].

With all that has been done, there still are unanswered questions. For instance, how would the capillary rise differ if the tube were chemically heterogeneous? There are undoubtedly many

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scenarios that could be considered. One could imagine inhomogeneities inside a tube that would completely impede the rise of liquid. On the other hand, if certain types of nonuniformities allowed liquid to rise, would liquid reach the same height as in an otherwise equivalent homogeneous tube? How would forces, pressures, energies and kinetics be affected?

These questions are not solely of academic interest. A wide variety of industrial products, such as filters, purifiers, gas contactors and various micro-fluidic devices, rely on controlling the wettability inside hollow fibers and other porous materials. In order to accurately characterize and control wettability inside porous materials, it is necessary to understand the role of chemical heterogeneities.

Thus, in this study, the rise of liquid in nonuniform capillary tubes with chemical gradients is examined. First, the forces, pressures and energies associated with a wettable, homogeneous tube are analyzed. Next, the same quantities are evaluated for two types of tubes that exhibit chemical gradients, yet still allow liquid to rise to the same level as a wettable, homogeneous tube of the same diameter. The kinetics of some of these nonuniform tubes is also explored.

2. Theory

2.1. Capillary rise in a smooth vertical tube

Consider the vertical capillary tube shown in Fig. 1. It is smooth with a constant, inner diameter of D. The capillary is brought into contact with a liquid of surface tension γ, density of ρ, and viscosity of μ, such that its bottom just touches the liquid, Fig. 1a. The liquid wets the tube with an advancing contact angle of θ. If D is small and $\theta < 90°$, then a concave meniscus forms inside the tube. The curvature of the meniscus and the surface tension of the liquid create an upward "Laplace" pressure (p_c),

$$p_c = \frac{4\gamma}{D}\cos\theta. \quad (1)$$

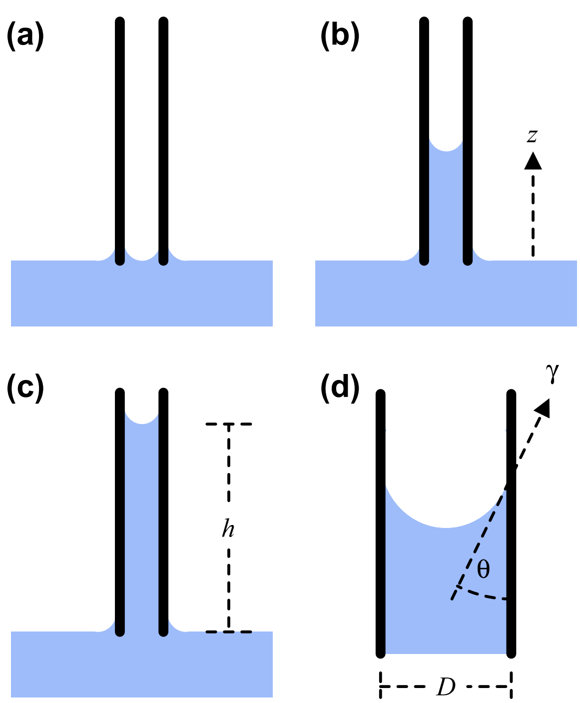

Fig. 1. Depiction of capillary rise in a small diameter tube. (a) The tube contacts the liquid and forms a concave meniscus inside the tube. (b) Liquid rises vertically. (c) The liquid stops at an equilibrium height of h. (d) A close-up view of the meniscus.

The Laplace pressure causes the liquid to rise in the vertical or z direction, Fig. 1b. As the liquid rises, the magnitude of the hydrostatic pressure inside the tube (p_h) increases with the transient height of the liquid column (z),

$$p_h = \rho g z, \quad (2)$$

where g is the acceleration due to gravity. The hydrostatic pressure is directed downward, acting against the Laplace pressure. The difference between Laplace and hydrostatic pressures (Δp),

$$\Delta p = p_c - p_h = \frac{4\gamma}{D}\cos\theta - \rho g z, \quad (3)$$

determines the rate of rise and the extent of energy dissipation within the liquid column. Flow ceases where $\Delta p = 0$ and $z = h$, Fig. 1c. Thus, from Eq. (3), the final rise height (h) can be estimated as [1a,1d,1e,14]

$$h = \frac{4\gamma\cos\theta}{\rho g D}. \quad (4)$$

If $\theta = 0°$, then Eq. (4) reduces to

$$h = \frac{4\gamma}{\rho g D}. \quad (5)$$

2.2. Wettability of the model tubes

Several wetting profiles, shown in Fig. 2, were used to explore variations in forces, pressures, energies and kinetics as liquid rises in a capillary tube. The tube shown in Fig. 2a is homogeneous and wettable with $\theta = 0°$ along its entire length. Fig. 2 also portrays two types of nonuniform tubes that exhibit a wetting gradient. The walls of these tubes are smooth with a constant diameter of D, but θ varies along their length. These tubes are relatively lyophobic near their bottoms where $z = 0$ and their wettability increases with z until $\theta = 0°$ where $z/h = 1$. Fig. 2b shows the profile of a nonuniform tube where θ varies exponentially as

$$\cos\theta = e^{z/h-1}, \quad (6)$$

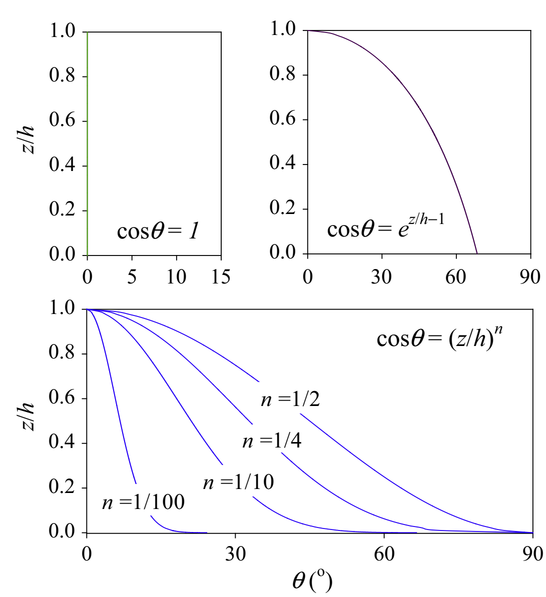

Fig. 2. Contact angle profiles inside of three types of capillary tubes: one type with homogeneous wettability where $\cos\theta = 1$ ($\theta = 0°$) and two types of heterogeneous tubes with chemical gradients where the wettability varies either as $\cos\theta = e^{z/h-1}$ or as $\cos\theta = (z/h)^n$.

from $\theta = 68°$ at the bottom of tube ($z/h = 0$) to $\theta = 0°$ at its ultimate rise height ($z/h = 1$). Fig. 2c shows a family of profiles where θ varies according to a simple power law relation,

$$\cos\theta = (z/h)^n, \tag{7}$$

from $\theta = 90°$ at the bottom to $\theta = 0°$ at the top. As $n \to 0$, the wetting gradient vanishes and its wetting profile approaches that of a wettable, homogeneous tube.

2.3. Energy balance

The energy balance for liquid slowly rising in a capillary tube consists of the three terms: the work done in lifting the liquid (w), a term that represents viscous dissipation (K) in the bulk of the flowing liquid and a potential energy term ($-\Delta U$) that accounts for energy stored in the liquid column [15],

$$w - K = -\Delta U. \tag{8}$$

2.4. Energies of homogeneous tubes

Let us begin with the simple case of a chemically homogeneous tube. Its inner surface is wettable along its entire length, such that $\theta = 0°$ for all z. The work done as surface tension lifts liquid upward through a tube (w) can be calculated by integrating the capillary force (f_c) from $z = 0$ to $z = h$ [16],

$$w = \int_0^h f_c \cdot dz, \tag{9}$$

where f_c is the product of length of the contact line and the vertical component of the surface tension,

$$f_c = \pi D \gamma \cos\theta. \tag{10}$$

Combining (9) and (10) and integrating gives

$$w = \pi D h \gamma \cos\theta. \tag{11}$$

The wetted area (A) of the inside of the capillary is

$$A = \pi D h. \tag{12}$$

Thus, the work done per wetted area (w/A) of a homogeneous, wettable tube ($\theta = 0°$) is [16]

$$\frac{w}{A} = \gamma. \tag{13}$$

The energy lost due to viscous dissipation (K) for laminar flow through a tube of constant cross section generally can be estimated from the liquid volume in the tube (V) and its hydrostatic pressure (p_h) [17],

$$K = \int V \cdot dp_h. \tag{14}$$

For capillary rise inside a wettable homogenous tube, Eq. (14) becomes

$$K = \int_0^h \frac{\pi}{4} D^2 \cdot \rho g z \cdot dz = \frac{\pi}{8} \rho g D^2 h^2. \tag{15}$$

Hence, viscous dissipation per wetted area (K/A) is

$$\frac{K}{A} = \frac{1}{8} \rho g D h = \frac{1}{2} \gamma. \tag{16}$$

According to elementary thermodynamics [18], the change in the gravitational potential energy ($-\Delta U$) can be calculated by integrating the hydrostatic force (f_h) as liquid rises from $z = 0$ to $z = h$,

$$-\Delta U = \int_0^h f_h \cdot dz. \tag{17}$$

The hydrostatic force is the product of the hydrostatic pressure (p_h) and the cross-sectional area of the tube (A_c), which in turn can be framed in terms of liquid density, height and tube diameter,

$$f_h = p_h \cdot A_c = \rho g z \cdot \frac{\pi}{4} D^2. \tag{18}$$

Combining Eqs. (17) and (18) gives an expression for a thin slice of the liquid column of height dz that has a local gravitational energy of $\rho g z (\pi D^2/4) dz$, which can be integrated to yield the change in potential energy of the full height (h) of the liquid column [3,15],

$$-\Delta U = \int_0^h \rho g z \cdot \frac{\pi}{4} D^2 dz = \frac{\pi}{8} \rho g D^2 h^2. \tag{19}$$

Subsequently, the change in potential energy per wetted area ($-\Delta U/A$) is

$$-\frac{\Delta U}{A} = \frac{1}{8} \rho g D h = \frac{1}{2} \gamma. \tag{20}$$

2.5. Energies of heterogeneous tubes

The work of wetting and the energy lost due to viscous dissipation for the model nonuniform tubes can be calculated using the z-dependent contact angle profiles. For the exponential wetting profile, Eq. (6), the work of wetting per area and dissipation per area are

$$\frac{w}{A} = (1 - e^{-1})\gamma \tag{21}$$

and

$$\frac{K}{A} = \left(\frac{1}{2} - e^{-1}\right)\gamma. \tag{22}$$

For the power law profiles, Eq. (7), they are

$$\frac{w}{A} = \frac{1}{n+1}\gamma \tag{23}$$

and

$$\frac{K}{A} = \frac{1}{2} \cdot \frac{1-n}{1+n} \gamma. \tag{24}$$

The change in potential energy, including the nonuniform ones, depends on the net change in the height of the liquid in the tube. Thus, for all cases,

$$-\frac{\Delta U}{A} = \frac{1}{2}\gamma. \tag{25}$$

2.6. Rate of rise

The rate of liquid rise in a capillary tube was first analyzed in the early part of the twentieth century [4]. Working equations can be derived by starting with the Poiseulle equation [17], which relates the volumetric flow rate (Q) to the difference between the Laplace and hydrostatic pressures (Δp),

$$Q = \frac{\pi D^4}{128 \mu z} \Delta p. \tag{26}$$

Here, only bulk viscous friction is taken into account. Any friction at the three-phase contact line is ignored. For a vertical tube of constant diameter (D), the transient liquid height (z) changes with time (t) as

$$Q = \frac{\pi}{4} D^2 \frac{\partial z}{\partial t}. \tag{27}$$

Combining Eqs. (26) and (27) and substituting Eq. (3) for Δp yields

$$\frac{\partial z}{\partial t} = \frac{D}{8\mu}\left(\gamma \cos\theta \cdot z^{-1} - \frac{1}{4}\rho g D\right). \quad (28)$$

With the initial boundary condition of $z = h$ when $t = 0$, this differential equation can be solved for a tube with constant contact angle to arrive at the classic Lucas–Washburn equation,

$$t = -\frac{32\mu}{\rho g D^2}\left[z + \frac{4\gamma\cos\theta}{\rho g D}\ln\left(1 - \frac{\rho g D}{4\gamma\cos\theta}z\right)\right]. \quad (29)$$

For the model nonuniform tubes with power law wetting dependence, modified Lucas–Washburn equations can be derived by substituting Eq. (7) into (28) and integrating. For example, if $n = ½$, then the modified Lucas–Washburn equation is

$$t = -\frac{32\mu}{\rho g D^2}\left[z + \frac{8\gamma}{\rho g D}\left[\sqrt{\frac{\rho g D z}{4\gamma}} + \ln\left(1 - \sqrt{\frac{\rho g D z}{4\gamma}}\right)\right]\right]. \quad (30)$$

3. Results and discussion

3.1. Pressure versus transient height

For a liquid to rise inside a tube, the Laplace pressure of the concave meniscus must be greater than the hydrostatic pressure of the liquid column, $p_c > p_h$. Fig. 3 shows dimensionless capillary pressure ($p_cD/4\gamma$) and hydrostatic pressure ($p_h/\rho gh$) for liquid rising inside smooth tubes with various wetting profiles. The dimensionless height (z/h) ranges from $z/h = 0$ at the bottom of the tube to $z/h = 1$ at the ultimate height of the liquid column. In all cases, the

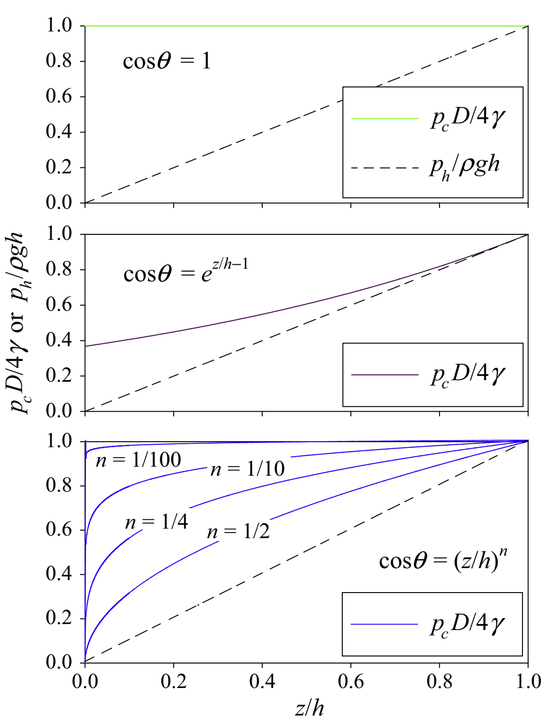

Fig. 3. Dimensionless capillary pressure ($p_cD/4\gamma$) and hydrostatic pressure ($p_h/\rho gh$) for liquid rising inside a smooth tube of diameter D. The dimensionless height (z/h) ranges from $z/h = 0$ at the bottom of the tube to $z/h = 1$ at the ultimate height of the liquid column. For $\cos\theta = 1$, the tube is homogeneous and wettable ($\theta = 0°$) from top to bottom. For $\cos\theta = e^{z/h-1}$, the contact angle varies exponentially from $\theta = 68°$ at the bottom of tube ($z/h = 0$) to $\theta = 0°$ at the top ($z/h = 1$), according to Eq. (6). For $\cos\theta = (z/h)^n$, the contact angle varies according to simple power law relation, Eq. (7), from $\theta = 90°$ at the bottom of tube ($z/h = 0$) to $\theta = 0°$ at the top ($z/h = 1$).

magnitude of the hydrostatic pressure (p_h) increases linearly with liquid height. On the other hand, the Laplace pressure depends on the type of wetting profile. For the case of the homogeneous, wettable tube ($\theta = 0°$ and $\cos\theta = 1$), the Laplace pressure (p_c) is constant as liquid rises and subsequently, the overall driving pressure (Δp), decreases linearly until $p_c = p_h$.

Similarly, for liquid to rise in the tubes with chemical gradients, $p_c > p_h$. However, because the Laplace pressure is reduced by the wetting gradient, the overall driving pressure, the energies and kinetics of the nonuniform tubes are significantly less than the geometrically equivalent homogeneous tubes. For the exponential wetting gradient, the initial value of Δp as liquid enters the bottom end of a given tube is only 37% of that found in a homogeneous, wettable tube of the same D. In order for liquid to rise in a tube with a power wetting gradient, n must be <1. Otherwise, if $n \geqslant 1$, then $p_c \leqslant p_h$ and liquid will not rise.

3.2. Energies

The work of wetting (w), viscous dissipation (K) and potential energy change ($-\Delta U$) of liquid rising in capillary tubes are extrinsic quantities that depend on the diameter of the tube and the ultimate rise height of the liquid. These quantities, derived in the Theory section, can be converted into intrinsic quantities by framing them in terms of wetted area (A). Furthermore, using Eq. (5) for final rise height, K/A and $-\Delta U/A$ can be rewritten in terms of surface energy of the liquid, which allows direct comparison to w/A values.

Table 1 lists the various intrinsic energies associated with liquid rising in homogeneous and heterogeneous tubes. For wettable, homogenous tubes, $½w/A = K/A = -\Delta U/A = ½\gamma$, which implies that half the work per area done in lifting the liquid is stored as potential energy while the other half is dissipated [16]. The same is not true for the heterogeneous tubes. Even though liquids are expected to rise to the same height and the change in potential energy is expected to be the same, chemical gradients in the heterogeneous tubes alter the capillary forces, which in turn influence the work of wetting and dissipation. The presence of lyophobic gradients reduces w/A and K/A values. For the nonuniform tube with an exponential wetting profile, $w/A = 0.63\gamma$ and $K/A = 0.13\gamma$. The difference between them is $½\gamma$, as expected from our energy balance, Eq. (8).

Values of w/A and K/A for heterogeneous tubes with power law wetting profiles vary with the exponent n, as shown in Fig. 4. Values of w/A ranged from $½\gamma$ to γ and values of K/A, from 0 to $½\gamma$. Once again, the difference between w/A and K/A was $½\gamma$ for any given value of n. Therefore, the difference between the work of wetting and the energy dissipated during capillary rise was equal to the change in potential energy.

3.3. Rate of rise

Because the presence of chemical gradients reduces the differential driving pressure, we anticipate liquid should rise more slowly in the nonuniform tubes. Indeed, the calculations presented here indicate this should be true. Fig. 5 shows dimensionless plots of transient rise height versus time for wettable, homogenous

Table 1
Energies associated with liquid rising in homogeneous or heterogeneous tubes.

Type	$\cos\theta$	θ (°)	w/A	K/A	$-\Delta U/A$	$(w-K)/A$
Homogeneous	1	0	γ	$½\gamma$	$½\gamma$	$½\gamma$
Heterogeneous	$e^{z/h-1}$	0–68	$(1-e^{-1})\gamma$	$(½-e^{-1})\gamma$	$½\gamma$	$½\gamma$
	$(z/h)^n$	0–90	$[(1/(n+1)]\gamma$	$½[(1-n)/(1+n)]\gamma$	$½\gamma$	$½\gamma$

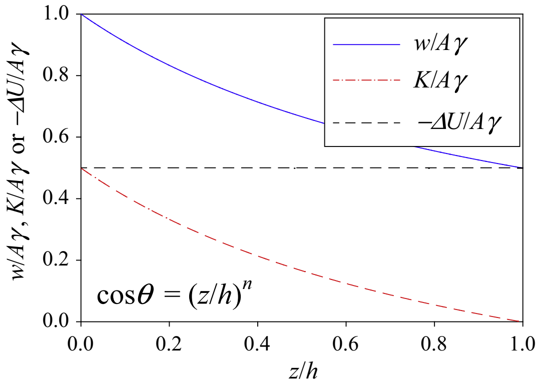

Fig. 4. Dimensionless values of work of wetting ($w/A\gamma$) and dissipated viscous energy ($K/A\gamma$) and potential energy ($-\Delta U/A\gamma$) for liquid rising inside tubes with power law wetting gradients, $\cos\theta = (z/h)^n$.

tubes, where $\cos\theta = 1$, and for heterogeneous tubes with power law wetting gradients, $\cos\theta = (z/h)^n$, where n values range between ½ and 9/10. For n = ½, the rise is slower than for a homogeneous tube of the same diameter, but not by much. If a power law gradient such that $n \to 1$ is employed, then the rate of rise is slowed significantly.

3.4. An example

Consider ethylene glycol rising in small diameter tubes with homogeneous, exponential and power law wetting profiles. Ethylene glycol has a surface tension of γ = 48 mN/m, a density of ρ = 1110 kg/m^3, a viscosity of μ = 20 mPa·s and a capillary length of $\lambda = (\gamma/\rho g)^{1/2}$ = 2 mm [19]. It has been observed that when $D \leqslant \lambda$, rise of liquid inside capillary tubes is sufficiently slow that it obeys the Lucas–Washburn equation [7e]. Accordingly, for our example assume that D = 0.2 mm. Here, the final rise height would be h = 88 mm for homogenous as well as nonuniform tubes. Because the liquid rises to the same height for the various tube types, the change in potential energy per wetted area is also the same for all of them, $-\Delta U/A$ = 24 mJ/m^2. On the other hand, wetting gradients reduced the wetting energy available to drive flow. Values of w/A ranged from 48 mJ/m^2 for a homogenous tube to 25.3 mJ/m^2 for a power law tube with n = 9/10. For a homogenous tube with D = 0.2 mm, the time to reach the maximum rise height of 88 mm (z/h = 0.999) would be 765 s or roughly 13 min. Conversely, for a power law tube with n = 9/10, it would take more than 10 times as long, about 8140 s, or more than two hrs.

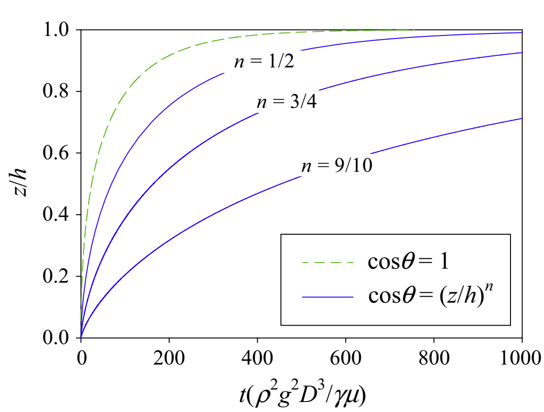

Fig. 5. Dimensionless plots of rise height versus time for: homogenous wettable tubes, $\cos\theta = 1$, and heterogeneous tubes with various power law wetting gradients, $\cos\theta = (z/h)^n$.

4. Conclusions

The forces, pressures, energies and kinetics were examined for capillary tubes with wetting gradients that ranged from relatively lyophobic at their bottom to completely wettable at their top. The profile of the wetting gradients was chosen such that the liquid would rise to the same height as in a wettable, homogeneous tube of the same diameter. Homogeneous or not, the change in potential energy per wetted area was the same. In contrast, the work of wetting, energy dissipation, and rise kinetics all depended on the wetting profiles. This analysis demonstrates that final rise height cannot be used as an unequivocal indicator of homogeneity.

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