Early regimes of capillary filling

Siddhartha Das, Prashant R. Waghmare, and Sushanta K. Mitra

Department of Mechanical Engineering, University of Alberta, Edmonton, Alberta, Canada T6G 2G8 (Received 31 August 2012; revised manuscript received 27 November 2012; published 14 December 2012)

In this paper we analyze the inviscid regime (for which viscosity is unimportant and the flow occurs due to the balance between the capillary and the inertial effects) that invariably precedes the classical century-old Washburn regime during capillary filling. We demonstrate that a new nondimensional number, namely, the product of the Ohnesorge number and the ratio between the filling length (ℓ) and the radius of the capillary (R), dictates the occurrence of this regime and the other well-known regimes in a capillary filling problem. We also identify that this inviscid regime occurs for the time that is of the order of the capillary time scale and, as has been quantified before [Quere, Eur. Phys. Lett. **39**, 533 (1997); Joly, J. Chem. Phys. **135**, 214705 (2011)], is characterized by the filling length being linearly proportional to the filling time. We establish the universality of this regime by pinpointing the existence of this regime (showing appropriate dependencies of the capillary radii and density) from existing experimental and Molecular Dynamics Simulation results that signify disparate ranges of length and time scales.

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I. INTRODUCTION

Since the pioneering contributions of Lucas [1], Washburn [2], and Bosanquet [3], efforts have been made to describe the problem of capillary filling as a function of the balance between the driving surface tension forces and resisting viscous drag, resulting in the filling length (ℓ) becoming proportional to the square root of filling time (t) [2–9]. The proposed derivation necessarily assumes $\ell \gg R$ (R is the capillary radius), so that one can consider the fully developed pressure-driven Poisseuille flow profile for explicit calculation of the viscous drag and thereby obtain the explicit form of the $\ell \sim \sqrt{t}$ variation [2,10]. One thing, however, remains seriously undiscussed. This assumption of $\ell \gg R$ is important not to ensure that a fully developed velocity profile is attained; rather it ascertains that the viscous drag (which is proportional to ℓ) becomes large enough to balance the capillary forces. Accordingly, one key question about the capillary filling problem remains unaddressed: What happens for $\ell \leq R$ or $\ell \sim R$? Alternatively, the question can be posed as Why for $\ell \leq R$ or $\ell \sim R$ viscous forces are unimportant? On the other hand, if we assume that the condition $\ell \gg R$ is necessary for the viscous forces to be able to balance the capillary forces, it is intuitive that for $\ell \sim R$, the capillary filling is driven by the balance between the capillary and the inertial forces, and this is the "inertial" or "inviscid" regime of capillary filling as pointed out by Quere [11]. This regime, as we shall demonstrate later, is applicable for $t \leq \tau_c$ or $t \sim \tau_c$ (where $\tau_c \sim \sqrt{\rho R^3/\gamma}$ is the capillary time scale, with ρ and γ being the density and surface tension of the liquid). Typically τ_c for common liquids is very small (e.g., for $R \sim 1 \text{ mm}$, we get $\tau_c \sim 3 \text{ ms}$ for water) and has remained mostly unexplored. Therefore, although several studies have mentioned about this "inertial" or "inviscid" regime (and point out that in this regime $\ell \sim t$) at the beginning of the capillary filling process [3,11–16], to the best of our knowledge this regime has not been properly analyzed due to a lack of reasonable answers to questions such as Which physical criteria demarcate this regime from the Washburn regime (where $\ell \sim \sqrt{t}$)? What is the role of the ratio ℓ/R in dictating this demarcation? Can one associate any characteristic time with this regime? etc.

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In this paper, we categorically respond to these questions. There are in essence two important contributions of this paper. First, we demonstrate that the demarcation of this inviscid regime from the well-known Washburn regime is governed by a dimensionless number, namely $Oh(\ell/R)$ (where $Oh = \eta / \sqrt{\rho \gamma R}$ is the Ohnesorge number, with η being the dynamic viscosity of the liquid). For example, for $Oh(\ell/R) \ll 1$, the viscous effects are negligible and we encounter this "inviscid" regime, whereas for $Oh(\ell/R) \sim 1$ the viscous and the capillary forces balance each other, and we have the Washburn regime. Second, we demonstrate that as an alternative to this dimensionless parameter $Oh(\ell/R)$, we can use τ_c to demarcate between this regime from the Washburn regime: For $t \sim \tau_c$ one encounters this "inviscid" regime, whereas for $t \gg \tau_c$, we get the Washburn regime. Our analysis yields the well-documented [11,16] result that in the "inviscid" regime, $\ell \sim t \sqrt{\gamma/\rho R}$. We recover this scaling dependence from existing experimental [11] (in a mm radius channel) and Molecular Dynamics Simulation (MDS) (in a nm radius channel) results [16,17]. Therefore, in a problem of capillary filling, we demonstrate the universality of this "inviscid" regime, preceding the Washburn regime, for a multitude of length and time scales.

II. SCALING ESTIMATES

The key to identify the parameter that dictates the different regimes in a capillary filling problem is to obtain the ratio between the resisting viscous forces F_v and the driving capillary forces F_c . We always have $F_c \sim \gamma R$ and $F_v \sim$ $\eta(\partial u/\partial y)\ell R \sim \eta u_0\ell$ (here y is the transverse coordinate or the coordinate perpendicular to the capillary walls), so that $F_v/F_c \sim \eta u_0\ell/\gamma R$. This ratio of the viscous to the capillary forces (except the coefficients) can be obtained by comparing the corresponding force expressions provided by Joly [16]. To use this expression for pinpointing the relative importance of the two forces (viscous and capillary), we need to express u_0 . Using $u_0 \sim \ell/t$, we get $F_v/F_c \sim \eta \ell^2/\gamma Rt$. To obtain the time t appearing in the ratio, we invoke the idea that at any instant the transport is caused by the capillary forces, so that $mdu/dt \sim F_c \Rightarrow t \sim \ell \sqrt{\rho R/\gamma}$ (using $m \sim \rho R^2 \ell$). Note that Joly [16] obtains the same scaling of the time t when the inertial and capillary effects balance each other. Also through such balance, we introduce the density ρ in the ratio of the viscous to the capillary forces. Hence using this expression for t in the F_v/F_c ratio, we get

$$F_v/F_c \sim \frac{\eta\ell}{\gamma R} \sqrt{\frac{\gamma}{\rho R}} \sim \frac{\eta}{\sqrt{\rho \gamma R}} \left(\frac{\ell}{R}\right) = Oh\left(\frac{\ell}{R}\right).$$
 (1)

Thus for $Oh(\ell/R) \ll 1$ we have the "inviscid" regime, and for $Oh(\ell/R) \sim 1$ we have the viscosity-dependent Washburn regime. This also dictates the role of the corresponding ℓ/R ratio in demarcating these two regimes. For example for $R \sim 1$ mm, Oh $\sim 10^{-2}$ – 10^{-3} for most of the common liquids. Therefore, one needs $\ell \sim 0.1-1$ m (i.e., $\ell/R \sim 10^2-10^3$) to ensure that one witnesses the Washburn regime. This regime selection based on the stated values of $Oh(\ell/R)$ is perfectly valid for horizontal capillary filling. However, for vertical capillary filling, the presence of gravity ensures that the net driving force is lower than the capillary force, and the Washburn regime is attained at a lesser value of ℓ/R ratio [or a value of $Oh(\ell/R)$ much lesser than unity [11] (in fact, in vertical capillary filling, ℓ cannot exceed the "Jurin" height [11]). At the initial stages (i.e, when $\ell \sim R$), however, gravity can be neglected even for a vertical capillary (as illustrated later), so that $Oh(\ell/R)$ dictates the "inviscid" regime even for the vertical capillary filling.

III. INERTIAL OR "INVISCID" REGIME

This is the regime where the viscous forces are negligible in comparison to the capillary forces, i.e., $Oh(\ell/R) \ll 1$, and the capillary filling is driven by a balance between the inertial and the capillary forces:

$$\gamma R \sim \frac{d}{dt} (\rho \ell R^2 u_0). \tag{2}$$

Therefore, we can obtain (using $t \sim \ell/u_0$), $u_0 \sim \sqrt{\gamma/\rho R}$, as shown by Quere [11]. This straightaway leads us to the relevant scaling law dictating the filling length ℓ and filling time t as

$$\ell \sim t \sqrt{\gamma/\rho R}.$$
 (3)

In this regime, one can assume $\ell \sim R$ (as in the inertial regime, the meniscus develops inside the tube, we have $\ell \sim R$ [11]), so that using (3), we can obtain the corresponding time scale (that characterizes this regime) as $t \sim \ell/u_0 \sim \sqrt{\rho R^3/\gamma} = \tau_c$. Therefore, this τ_c quantifies the "initial" time, where the viscous forces are subdominant, and the capillary filling is governed by (2) and (3). Hence, we can infer that $t \sim \tau_c$ is a condition, alternative to the condition $Oh(\ell/R) \ll 1$, that dictates the occurrence of the "inviscid" regime. We can discuss the relevance of this time scale in the light of the full analytical solution of the capillary filling (considering all the three forces, i.e., inertial, capillary, and viscous) provided by Joly [16]. For $t \ll \tau_v$ (where $\tau_v \sim \rho R^2/\eta$ is the viscous time scale), we get from Joly [16], $\ell \sim t \sqrt{\gamma/\rho R}$ [18]. This is the same expression that we obtain in (3), and Joly [16] proposes for the case when viscous effects are negligible, which we show to occur when $Oh(\ell/R) \ll 1$. Therefore, both conditions $Oh(\ell/R) \ll 1$ or $t \sim \tau_c$ (or $t \ll \tau_v$) equivalently represent the inertial regime and the crossover (from inertial to Washburn regimes) occurs for $Oh(\ell/R) \sim 1$ or $t \gg \tau_c$ (or $t \sim \tau_v$; this

is based on the condition that $\tau_c \ll \tau_v$ for most liquids; see Table I). Note that from the full analytical expression provided in Joly [16], we get the exact quantification of the larger time scale (τ_v), where $\ell \propto \sqrt{t}$; however, we do not get the smaller time scale (τ_c), where $\ell \propto t$.

For vertical capillary filling, where gravity is important, there will be a gravitational force $F_g \sim \rho \ell R^2 g$ (where g is the acceleration due to gravity). Therefore, with $t \sim \tau_c$ and $\ell \sim R$, we get $F_I/F_g \sim \gamma/\rho g R^2$, which for $R \leq 1$ mm, is always much larger than unity for standard liquids (e.g., for water with $R \sim 1$ mm, we have $F_I/F_g \sim 10$). Therefore, we can safely state that even for the vertical capillary filling, Eq. (3) is the scaling law for the "inviscid" regime.

IV. INTERMEDIATE OR WASHBURN REGIME

In this regime the capillary flow is driven by the balance of the viscous and the capillary forces. In effect, this implies that ℓ has become substantially large so that $Oh(\ell/R) \sim 1$ (for horizontal capillaries). Hence, one may write

$$F_v \sim F_c \Rightarrow \eta u_0 \ell / \gamma R \sim 1 \Rightarrow \ell \sim \sqrt{t} \sqrt{\gamma R / \eta},$$
 (4)

i.e., we recover the Washburn regime $(\ell \sim \sqrt{t})$, with appropriate dependence on system parameters [2]. We can obtain the same form of scaling law [as (4)] by using the full-scale solution of Joly [18]. Above, we have shown that the condition $t \sim \tau_c$ and $Oh(\ell/R) \ll 1$ are equivalent as both of them signify that the filling is in the inertial regime. Using (4), we shall demonstrate that the condition $t \gg \tau_c$ and $Oh(\ell/R) \sim 1$ are equivalent as both of them signify that the filling is in the Washburn regime. Defining $\bar{t} = t/\tau_v$, we can rewrite (4) as $Oh(\ell/R) \sim \overline{t}$. Hence for $Oh(\ell/R) \sim 1$, we have $\overline{t} \sim 1$, or $t \sim \tau_v$, i.e., $t \gg \tau_c$ since we have shown above (both from our analysis, and the full-scale solution of Joly [16]), when $t \ll \tau_v$, $t \sim \tau_c$. For this case, if gravity becomes important (vertical capillary filling) the condition $Oh(\ell/R) \sim 1$ no longer dictates the attainment of the Washburn regime. Rather, the net driving force being much smaller (since there is the retarding gravitational effect) than $F_c \sim \gamma R$, the Washburn regime is attained at a much smaller $Oh(\ell/R)$ or smaller ℓ/R [e.g., in experiments by Quere [11] with ethanol as the filling liquid, we observe the onset of Washburn regime for $Oh(\ell/R) \sim 0.1$]. The corresponding ℓ versus t variation can be obtained from the force balance, which reads $\gamma/R - \rho g \ell \sim \eta u_0 \ell/R^2$. Since $\ell < \ell_J$ (where $\ell_J \sim \gamma / \rho g R$ is the Jurin height [11]), $\gamma / R > \rho g \ell$, and hence the force balance becomes approximately $\gamma \sim \eta \ell^2 / Rt$ (using $u_0 \sim \ell/t$), yielding $\ell \sim \sqrt{t}\sqrt{\gamma R/\eta}$, i.e., the same scaling law [Eq. (4)]. Off course, how close ℓ is to ℓ_J decides the time up to which one can witness the Washburn regime.

V. VISCOUS REGIME

Once ℓ has become so large that $Oh(\ell/R) \gg 1$, the viscous effects will overwhelm the capillary forces. Therefore, now one will have the balance between the viscous and the inertial forces:

$$\frac{d}{dt}(\rho R^2 \ell u) \sim \eta \frac{\partial u}{\partial y} \ell R \Rightarrow t \sim \frac{\rho R^2}{\eta} = \tau_v, \qquad (5)$$

where τ_v is the viscous time scale.

To close the discussion on the different regimes in capillary filling, it is worthwhile to mention here that there has been a conjecture that at time scale $t \ll \tau_c$ (i.e., time preceding the "inviscid" regime), we should have $\ell \sim t^2$ [4,11,15,19], motivated by the requirement of continuity in velocity at t = 0 [11]. We would like to emphasize here that such a regime would indeed exist; however, $\ell \sim t^2$ is not mandatory; rather, $\ell \sim t^n$ (where n > 1) would satisfactorily fulfill the requirement of velocity continuity at t = 0.

VI. RESULTS AND DISCUSSIONS

The key to identify the existence of the different regimes, as illustrated above, is to estimate the value of $Oh(\ell/R)$, or to obtain τ_c and τ_v (see Table I). We find only for sufficiently large capillary radius ($R \sim 1 \text{ mm}$) that we get $\tau_c \ll \tau_v$, and τ_c is in the measurable range ($\tau_c \sim 1-10 \text{ ms}$). For smaller capillary radii, either $\tau_v \sim \tau_c$ (e.g., $R \sim 100 \text{ nm}$) or $\tau_c > \tau_v$ but τ_c is too small to measure (e.g., for water in $R \sim 10 \ \mu m$ capillary, $\tau_c \sim 1 \ \mu$ s). In fact, the time resolution of most of the classical capillary filling experiments (with $R \sim 0.1-1 \text{ mm}$) [20,21] is ~ 0.1 –1 s, so that one never encounters this otherwise universal linear regime. One of the rare exceptions is the study by Quere [11], where results are provided for $t \sim \tau_c$ (for his experiments $\tau_c \sim 4 \text{ ms}$ and $\tau_v \sim 300 \text{ ms}$). In Fig. 1 we validate our scaling law for the linear regime with the experiments of Quere [11]. We obtain an excellent match, and the linear regime is witnessed for $t \sim 10$ ms, i.e., $t \sim \tau_c$. Please note that the experiment by Quere is on a vertical capillary where the gravity effects are important. However, as we have discussed, our scaling law [see Eq. (3)] for the "inviscid" regime remains unaffected by the gravity (for the case studied by Quere, we have $F_I/F_g \sim 6$). In Fig. 1, we also validate the scaling law for the Washburn regime. As gravity is important, this regime is witnessed at $t < \tau_v$.

The other spectrum of capillary filling problems, which deal with much smaller time scales (picoseconds to nanoseconds) are the MDS studies on capillary imbibition [13,16,17,22]. In these studies $R \sim 1$ nm and $u_0 \sim 100$ m/s [16,17] (attributed to large slip lengths [23–25]). Alternatively, without considering any slip, one must consider an equivalent reduction in the viscosity [26,27] by four-to-five orders (it increases the viscous time scale by the same order). The key reason

TABLE I. Capillary and viscous time scales for different fluids. We have exponents a = 9, 6, 3 and b = 9, 5, 1 for R = 100 nm, 10 μ m, 1 mm. We take the standard liquid properties at 20 °C.

Fluid	$ au_c imes 10^a$ (s)	$\tau_v \times 10^{\rm b} ({\rm s})$
Benzene	5.5	13.3
Carbon tetrachloride	7.7	16.6
Chloroform	7.4	26.3
Ethanol	6	6.9
Hexane	6	20.1
Isopropanol	6.1	3.3
Methanol	5.5	13.3
Water	3.7	10



FIG. 1. (Color online) Match of the scaling laws for the "inviscid" (or linear) and the Washburn (or intermediate) regimes with the experimental results of Quere [11]. For the linear regime our scaling law is expressed as $\ell = M_1 t \sqrt{\gamma / \rho R}$, where M_1 is a constant, which is obtained by matching one experimental data with the scaling law (expressed above). For the present case the liquid is ethanol ($\gamma =$ 21.6 mN/m, $\rho = 780 \text{ kg/m}^3$, $\eta = 1.17 \text{ mPas}$) and $R = 689 \,\mu\text{m}$, so that we get $M_1 = 0.8774$ [using $(\ell)_{t=2.16 \text{ ms}} = 0.38 \text{ mm}$]. For the Washburn (intermediate) regime, our scaling law is expressed as $\ell = M_2 \sqrt{t} \sqrt{\gamma R/\eta}$ (the effect of gravity is accounted for in the constant M_2). With the stated parameters, we get $M_2 = 7.1348$ (using $(\ell)_{t=50.11 \text{ ms}} = 5.7 \text{ mm}$). We depict the experimental results by markers (blue circles) and scaling predictions by lines (red dashed for the inertial regime and green bold line for the Washburn regime). M_1 and M_2 dictate the slopes of the ℓ vs t and ℓ vs \sqrt{t} variations, respectively for a given fluid in a capillary of a given radius.

for such a viscosity reduction is the presence of depletion layers adjacent to the channel walls [28,29]. For water with $R \sim 1$ nm, this reduction increases τ_v from 0.1 ps to 1–10 ns.



FIG. 2. (Color online) Match of the scaling law for the "inviscid" (or linear) regime with the MDS results [17] of carbon nanotube capillary imbibition, showing the effect of density. Our scaling law is expressed as $\ell = M_1 t \sqrt{\gamma/\rho R}$, so that we can express $\ln(\bar{\ell}) =$ $\ln(\bar{\iota}) - \ln(\bar{\rho})/2 + \ln(C)$, where $\bar{\ell} = \ell/\ell_0$, $\bar{\iota} = t/t_0$, $\bar{\rho} = \rho/\rho_0$, and $C = (M_1 t_0/\ell_0) \sqrt{\gamma/\rho_0 R}$. Here ℓ_0 , t_0 , and ρ_0 are the length, time, and density scales. Procedure to obtain *C* is illustrated in Ref. [30]. We depict the experimental results by markers (blue circles for $\bar{\rho} =$ 0.11, red squares for $\bar{\rho} = 0.21$, and green triangles for $\bar{\rho} = 0.32$) and scaling predictions by lines (blue bold lines for $\bar{\rho} = 0.11$, red dashed line for $\bar{\rho} = 0.21$, and green dash-dot line for $\bar{\rho} = 0.32$).



FIG. 3. (Color online) Match of the scaling law for the "inertial" regime with the MDS results [16], showing the effect of capillary radii. Our scaling law is expressed as $\ell = A_1 t \sqrt{\gamma/\rho R}$, where A_1 is a constant [physically, the role played by A_1 is identical to that played by M_1 (see Fig. 1)] when ℓ and R are expressed in nm and t is expressed in ns. This scaling law can be expressed as $\ell = C_1(t/\sqrt{R})$, where $C_1 = A_1\sqrt{\gamma/\rho}$. Procedure to obtain C_1 is illustrated in Ref. [31]. In Ref. [32], we also compare and discuss the values of A_1 and M_1 , both of which represent the numerical prefactor to the scaling law [see Eq. (3)]. We depict the experimental results by markers (blue circles for R = 0.514 nm and red squares for R = 0.514 nm and black bold line for R = 1.87 nm).

On the contrary for the same capillary, $\tau_c \sim 10$ ps. Therefore, when $t \sim 10-100$ ps (as witnessed in MDS results [16,17]), we would expect a $\ell \sim t$ variation as predicted in Eq. (3). Figure 2 compares our scaling hypothesis with MDS results (for capillary imbibition in a carbon nanotube with $R \sim 1$ nm) in this time range [17], showing a clear $\ell \sim t$ variation with the proposed dependence on ρ [for this study, we have $\tau_c \sim 7$ ps and $\tau_v \sim 10$ ns (with a reduction in viscosity by four orders)]. In Fig. 3 we provide comparison with results of another MDS study [16] of capillary imbibition in carbon nanotubes, showing the proposed dependence on capillary radii [for this study, we have $\tau_c \sim 5$ ps and $\tau_v \sim 50$ ns (with a reduction in viscosity by four orders)]. The excellent match of our scaling laws with the MDS results in both Figs. 2 and 3 establish that our proposed scaling remains valid even for MDS time scales. Also the fact that $\tau_v \sim 1-10$ ns indicates that the viscosity-dependent (or Washburn) regime will be observed over this time scale (which is much larger than the time scales accessed in Refs. [16,17]), as indicated by other MDS results [13,22,33,34].

VII. CONCLUSIONS

In this paper we analyze the well-known "inviscid" regime (characterized by linear variation of the filling length ℓ with the filling time t [11,16]) that invariably precedes the celebrated Washburn regime in capillary filling process. We identify experiments [11] and MDS results [16,17] that establish the universality of this "inviscid" regime spanning across a multitude of length and time scales. We show that this regime is demarcated from the classical Washburn regime by a dimensionless number, namely, $Oh(\ell/R)$. Alternatively, as a more practical measure one can consider τ_c (or the capillary time scale) as the relevant time scale over which one can witness this regime.

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- [1] R. Lucas, Kolloid Z. 23, 15 (1918).
- [2] E. W. Washburn, Phys. Rev. 17, 273 (1921).
- [3] C. H. Bosanquet, Philos. Mag. 45, 525 (1923).
- [4] B. V. Zhmud, F. Tiberg, and K. Hallstensson, J. Colloid Interface Sci. 228, 263 (2000).
- [5] N. R. Tas, J. Haneveld, H. V. Jansen, M. Elwenspoek, and A. van den Berg, Appl. Phys. Lett. 85, 3274 (2004).
- [6] A. Han, G. Mondin, N. G. Hegelbach, N. F. de Rooij, and U. Staufer, J. Colloid Interface Sci. 295, 151 (2006).
- [7] J. W. van Honschoten, J. W. Berenschot, T. Ondarçuhu, R. G. P. Sanders, J. Sundaram, M. Elwenspoek, and N. R. Tas, Appl. Phys. Lett. 97, 014103 (2010).
- [8] J. M. Oh, T. Faez, S. de Beer, and F. Mugele, Microfluid. Nanofluid. 9, 123 (2010).
- [9] P. Waghmare and S. K. Mitra, Microfluid. Nanofluid. 12, 53 (2012); Anal. Chim. Acta 663, 117 (2010); A. Saha and S. K. Mitra, J. Colloid Interface Sci. 339, 461 (2009); A. Saha, S. K. Mitra, M. Tweedie, S. Roy, and J. McLaughlin, Microfluid. Nanofluid. 7, 451 (2009).
- [10] L. R. Fisher and P. D. Lark, J. Colloid Interface Sci. 69, 486 (1979).
- [11] D. Quere, Europhys. Lett. 39, 533 (1997).

- [12] A. Kutana and K. P. Giapis, Nano Lett. 6, 656 (2006).
- [13] D. I. Dimitrov, A. Milchev, and K. Binder, Phys. Rev. Lett. 99, 054501 (2007).
- [14] N. Ichikawa and Y. Satoda, J. Colloid Interface Sci. 162, 350 (1994).
- [15] M. Dreyer, A. Delgado, and H. Rath, J. Colloid Interface Sci. 163, 158 (1994).
- [16] L. Joly, J. Chem. Phys. 135, 214705 (2011).
- [17] S. Supple and N. Quirke, Phys. Rev. Lett. 90, 214501 (2003).
- [18] From Joly [16], we get the full solution of the capillary filling length as $\ell/\ell_0 = [t/\tau'_v + (1/2)(e^{-2t/\tau'_v} 1)]^{1/2}$, where (without slip at solid-liquid interface) $\tau'_v = (\rho R^2/4\eta) = \tau_v/4$ and $\ell_0 = \sqrt{2\gamma\rho R^3}/4\eta$. Hence for small time $(t \ll \tau_v \text{ or } t \ll \tau'_v)$, we can expand the exponential term (up to quadratic term) in the expression of ℓ/ℓ_0 to obtain $\ell \approx t\sqrt{2\gamma/\rho R}$. From this condition, we get the corresponding time scale as $\tau_c = \sqrt{\rho R^3/\gamma}$, by assuming that for such small time (characteristic of the inviscid regime), we have $\ell = R$. This also implicitly means $\tau_c \ll \tau_v$, as explicated for most of the common liquids (see Table I). Similarly, when $t \sim \tau_v$ or $t \sim \tau'_v$, we get from the full solution $\ell/\ell_0 \approx \sqrt{t/\tau'_v} \Rightarrow \ell \approx \sqrt{t}\sqrt{\gamma R/2\eta}$.

- [19] M. Stange, M. E. Dreyer, and H. J. Rath, Phys. Fluid 15, 2587 (2003).
- [20] F. G. Yost, R. R. Rye, and J. A. Mann, Jr., Acta Mater. 45, 5337 (1997).
- [21] M. Radiom, W. K. Chan, and C. Yang, Microfluid. Nanofluid. 9, 65 (2010).
- [22] B. Henrich, C. Cupelli, M. Santer, and M. Moseler, New J. Phys. 10, 113022 (2008).
- [23] M. Majumder, N. Chopra, R. Andrews, and B. J. Hinds, Nature (London) 438, 44 (2005).
- [24] M. Whitby and N. Quirke, Nat. Nanotech. 2, 87 (2007).
- [25] M. Whitby, L. Cagnon, M. Thanou, and N. Quirke, Nano Lett. 8, 2632 (2008).
- [26] J. S. Babu and S. P. Sathian, J. Chem. Phys. 134, 194509 (2011).
- [27] B. Xu, B. Wang, T. Park, Y. Qiao, Q. Zhou, and X. Chen, J. Chem. Phys. 136, 184701 (2012).
- [28] S. Joseph and N. R. Aluru, Nano Lett. 8, 452 (2008).
- [29] M. Majumder, N. Chopra, R. Andrews, and B. J. Hinds, Nature (London) 438, 44 (2005).
- [30] We obtain a *C* value by matching one experimental data with the scaling law (see the caption of Fig. 2) and then apply this scaling law to obtain the scaling predictions. In the present case, we obtain $[\ln (C)]_{\bar{\rho}=0.11} = -0.3609$ [using $\ln (\bar{\ell})_{\ln (\bar{\ell})=2.025} = 2.7677$], $[\ln (C)]_{\bar{\rho}=0.21} = -0.4329$ [using $\ln (\bar{\ell})_{\ln (\bar{\ell})=4.055} = 4.3529$], and

 $[\ln(C)]_{\bar{\rho}=0.32} = -0.4274$ [using $\ln(\bar{\ell})_{\ln(\bar{\ell})=4.0078} = 4.1501$]. Using the actual properties described in Ref. [17], we get $\ln(C) \approx -0.48$, i.e., of similar range as obtained from the fit.

- [31] We obtain a C_1 value by matching one experimental data with the scaling law (see the caption of Fig. 3) and then apply this scaling law to obtain the scaling predictions. In the present case, we obtain $(C_1)_{R=0.514 \text{ nm}} = 25.01 \text{ nm}^{3/2}/\text{ns}$ [using $(\ell)_{t=0.1709 \text{ ns}} = 5.9596 \text{ nm}$] and $(C_1)_{R=1.87 \text{ nm}} = 44.48 \text{ nm}^{3/2}/\text{ns}$ [using $(\ell)_{t=0.1721 \text{ ns}} = 5.60 \text{ nm}$]. Decrease in C_1 with decrease in radius can be associated with an enhancement in density ρ for a channel of smaller radius.
- [32] From the values of C_1 [31], we obtain the values of A_1 as $(A_1)_{C_1=44.48 \text{ nm}^{3/2}/\text{ns}} = C_1 \sqrt{\rho/\gamma} = 0.2$ (using $\rho = 980 \text{ kg/m}^3$ and $\gamma = 0.05 \text{ N/m}$ [16]). Hence both A_1 and M_1 (see caption of Fig. 1) are of similar order. The difference which is reflected in their values is likely to be attributed to the differences between ℓ and R values at which they are calculated; e.g., M_1 is calculated on the basis of the data corresponding to $\ell = 0.38 \text{ mm}$ and R = 0.689 mm (i.e., $\ell < R$) (see caption of Fig. 1), whereas A_1 is calculated on the basis of the data corresponding to $\ell = 5.60 \text{ nm}$ and R = 1.87 nm (i.e., $\ell > R$) (see Ref. [31]).
- [33] S. Ahadian and Y. Kawazoe, Colloid Polym. Sci. 287, 961 (2009).
- [34] S. Ahadian, H. Mizuseki, and Y. Kawazoe, J. Colloid Interface Sci. 352, 566 (2010).