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Dynamics of water imbibition through hydrogel-coated capillary tubes

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Water-absorbing materials consisting of hydrogel particles are widely used for agricultural substrates, hygienic products, drug delivery systems, and microfluidic devices. Hydrogel particles absorb water with expansion, leading to dynamics involving complicated interactions of intergranular liquid flow and hydrogel deformation. It is thus difficult to predict the water absorption rate of these materials. Water flow in hydrogel-coated capillary tubes also occurs with the deformation of flow passages, so it can be analogous to capillary flow through porous structures made of hydrogels. Here we report a combined experimental and theoretical investigation of water flow through capillaries whose inner surface is covered with a hydrogel layer. We experimentally measured the flow rate of water through hydrogel-coated capillary tubes and the volume of water absorbed by the hydrogel layer. We developed a mathematical model for capillary flow including the effects of the water absorption and swelling of the hydrogel layer. Our model is in good agreement with the measurements of the water flow rate and elucidates how the absorption and swelling of hydrogel regulate capillary flow. This study provides a theoretical framework for understanding the dynamics of capillary flow through porous structures composed of hydrogels, leading to insights into the engineering applications of porous hydrogel materials.

I. INTRODUCTION

A hydrogel is a cross-linked polymer network that can absorb water. Since hydrogels are highly biocompatible and can uptake a great amount of water, they are widely used as water-absorbing materials in a variety of applications. However, in some applications where rapid water absorption is necessary, hydrogel often suffers from low rate of water uptake, mainly because of diffusive water transport through a bulk hydrogel body [1,2]. In such cases, porous structures made of hydrogel, such as hydrogel granular materials, can be exploited to accelerate water absorption by virtue of convective flow through interconnected pores driven by capillary action. Indeed, improved absorption rate resulted in a better performance of various systems made of hydrogels, such as cell-culturing platforms [3], solvent absorbers [4], drug delivery systems [5,6], microfluidic pumps [7], and soft actuators [8].

Accurate prediction of water absorption rate through porous hydrogel materials is crucial for practical applications. Hydrogels absorb water with swelling, and water flow through a porous hydrogel structure results in the deformation of the interconnected pore structure. Understanding of flow through porous hydrogel structures thus requires a coupled analysis of flow dynamics and

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polymer physics. Researchers have analyzed liquid flows in some deformable porous materials, such as engineered sponges [9–12] and papers [13]. The theoretical frameworks used in previous studies are accompanied by mathematical analysis to describe the liquid-solid interaction and resultant deformation of the solid, which typically depends sensitively on several physical properties. However, many physical properties, such as permeability and modulus of deformation, which can vary with the water content, are very difficult to measure experimentally, often rendering the theoretical frameworks far from practical.

In a macroscopic analysis of porous media flow, geometrical features at the pore scale can be appropriately disregarded by introducing mean parameters. Specifically, capillary imbibition into porous materials can be understood on the ground of the Washburn equation that describes the capillary-driven flow through a cylindrical tube as $l = [\sigma R \cos\theta t/(2\mu)]^{1/2}$, where l is the imbibition length, σ is the surface tension, θ is the contact angle, R is the tube radius, t is the time, and μ is the liquid viscosity [14]. Considering the analogy between the flow through a porous medium and a bundle of cylindrical capillary tubes with a radius of R, one can predict the liquid imbibition rate through the porous material as $l = kt^{1/2}$ with k that scales as $(\sigma R \cos\theta/\mu)^{1/2}$. One can estimate k by experimentally measuring the interdependence of l and t, so that the Washburn equation can provide a practical analytical method despite inaccuracies resulted from blurred wetting front, intrapore structure, and swelling [15–17]. Nevertheless, water absorption through porous hydrogel materials cannot be understood based on the Washburn equation because the equation does not accommodate the deformation of hydrogels.

Water imbibition through porous hydrogel materials can be understood as analogous to a flow through a channel that deforms with water absorption, rather than to a capillary flow through a stiff channel. In this respect, capillary flow through a hydrogel channel can offer a simple, but practical, theoretical framework for better understanding of the water absorption through porous hydrogel materials. Only a few studies have been reported on water imbibition through hydrogel capillary channels. Andersson *et al.* [18] studied water flow through channels formed in a hydrogel structure, and Silva *et al.* [19] examined water flow through a capillary tube with the inner surface coated with hydrogel. Those studies reported experimental measurements of flow speed and discussed the effect of local deformation of the hydrogel boundary in the vicinity of advancing meniscus. Of interest to biological systems, e.g., water uptake in wood during the following periods of drought, in engineering and construction applications, Zhou *et al.* [20] studied water flow through a hydrogel channel as an analogous system of hygroscopic wood material. Indeed, it has been reported that many channels in plants, such as xylems, are coated by hydrogels, allowing plants to control the hydraulic resistance [21,22]. However, the effects of water absorption and swelling of hydrogel on water flow were not included in the previous studies, and the dynamics of water imbibition through hydrogel-coated channels still remains unclear.

In the present study, we examined capillary flow through hydrogel-coated capillary tubes. We experimentally observed water flow in hydrogel-coated capillary tubes and measured the volume of water absorbed by the hydrogel layer. We developed a simple mathematical model for water imbibition through hydrogel-coated capillary tubes including the effects of water absorption and swelling of the hydrogel layer. The model agrees well with experimental observations and thus reveals the role of the water absorption and expansion of the hydrogel in the dynamics of water flow. Our study provides a theoretical framework for understanding the water flow through porous hydrogel material, leading to insights into the engineering applications of those materials.

II. EXPERIMENTS

We fabricated hydrogel-coated capillary tubes as shown in Fig. 1(a). A glass capillary tube with an inner diameter of 860 μm was fixed on a three-axial stage. A Teflon-coated wire with a diameter of 290 μm (SME Korea) was inserted in the tube and tightly stretched out on both sides. We adjusted the position of the glass capillary tube so that the Teflon-coated wire was coaxially aligned with the cylindrical tube. Two hydrogel solutions were produced for coating inside of the capillary tube. We
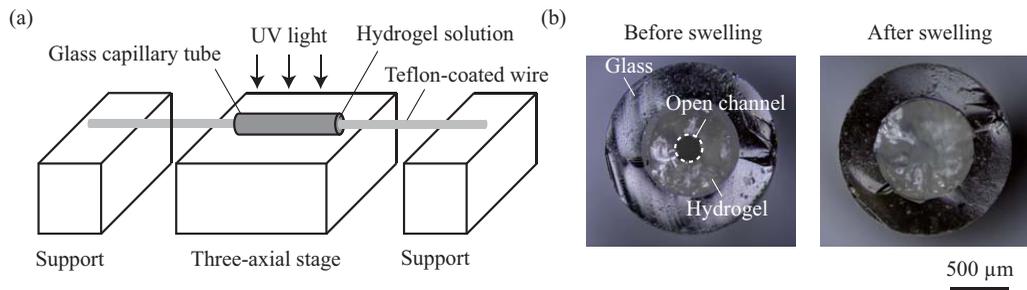
FIG. 1. (a) Schematic illustration of hydrogel-coated capillary tube production. (b) Cross-sectional images of the hydrogel-coated capillary tube before and after swelling

prepared a mixture of 5 ml acrylic acid and 5 ml sodium hydroxide solution with a concentration of 20 wt%. By adding either 100 or 200 mg of a cross-linker, N, N'-methylenebis(acrylamide), to the mixture, we prepared hydrogel solutions I and II with cross-linker concentrations of 0.01 and 0.02 g/ml, respectively. As a photoinitiator, we used 0.2 ml of 2-hydroxy-2methyl-propiophenone. One of the prepared hydrogel solutions was injected into the space between the wire and glass capillary tube. After the hydrogel was polymerized by irradiation with ultraviolet light for 1 min, the Teflon-coated wire was gently pulled out of the tube. Figure 1(b) shows the cross sections of the hydrogel-coated tube before and after swelling with water absorption.

Figure 2(a) shows the experimental setup to measure the volume of water absorbed by the hydrogel layer in a glass capillary tube. A capillary tube with an inner diameter (D) of 290 μm was connected to a syringe with a volume of 3 ml through a small hole created on the syringe. After filling the syringe and capillary tube with water, the syringe nozzle was covered with a thin tape. By cutting the fabricated hydrogel-coated capillary tubes, we created a sliced piece of tube with a length of 5 mm. When a cut tube piece was brought to the syringe nozzle, the sealing tape was broken. Water immediately flowed into the cut tube piece within 1 s, mainly by capillary action, hence filling the void in the hydrogel layer. Water was then absorbed by the hydrogel layer with expansion, leading to a change in the water column height in the glass capillary tube connected to the syringe. By measuring the temporal evolution of the water column height, we assessed the volume of water absorbed by the hydrogel and the time required for the expanded hydrogel to completely occupy the inner space in the tube.

We constructed an experimental setup to observe water imbibition through hydrogel-coated capillary tubes as shown in Fig. 2(b). A tube was placed in the horizontal direction to preclude gravitational effects on the water flow. We adjusted the horizontal position of the water reservoir so that one end of the tube came into contact with water. While the capillary tube imbibed water, the imbibition front was filmed using a DSLR camera (Canon, 90D). Figures 2(c) and 2(d) show sequential images of the advancing front of infiltrating water.

III. EXPERIMENTAL OBSERVATIONS

We begin with a description of our observations in experiments on short tube segments to determine the rate of the water absorption of the hydrogel layer, as shown in Fig. 2(a). Figure 3(a) displays the temporal evolution of the water column height difference in the experiment with cut tube pieces. The displacement of the water column height, shown in Fig. 2(a), initially increases rapidly, but later the rate of increase slows down. It is supposed that while the initial rapid change in h in a timescale of $\sim$1 s is mainly attributed to water imbibition into the space inside the cylindrical hydrogel layer in the tube by capillary action, the change in h after $\sim$1 s is associated with water absorption by the hydrogel layer. After $\sim$10 s, the expanded hydrogel layer completely occupies the inner space of the tube, limiting further expansion of the hydrogel layer and hence water uptake,
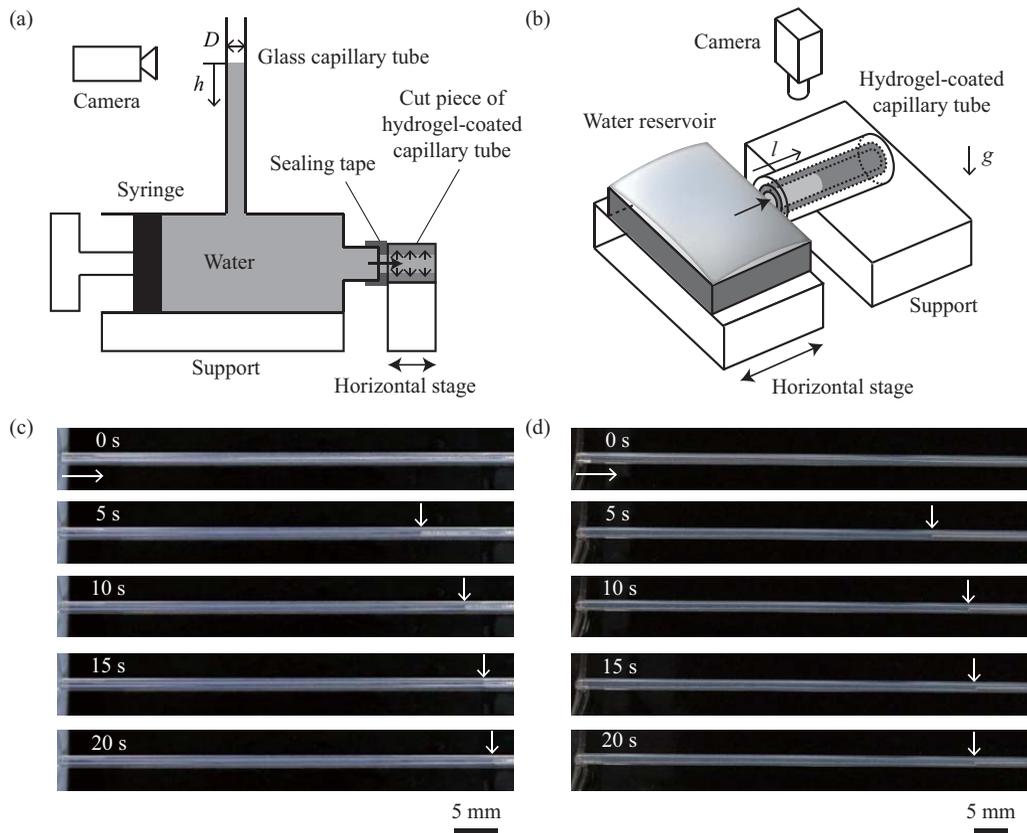
FIG. 2. Schematic illustrations of experimental setups for (a) measuring volume of water absorbed by the hydrogel layer and (b) measuring imbibition length in hydrogel-coated capillary tubes. Sequential images of imbibing water through hydrogel-coated tubes made of hydrogels (c) I and (d) II. Arrows indicate imbibition front.

resulting in only a negligible change in h. The cross sections of the hydrogel-coated capillary tubes before and after swelling are shown in Fig. 1(b). We define t_e as the time it takes for h to reach 99% of its maximum value, which is approximately 14 s and 11 s for hydrogel I and II, respectively. The volume of water ultimately absorbed by the hydrogel in the 5 mm cut tube was measured to be 0.58 and 0.47 mm^3 for hydrogel I and II, respectively.

We now proceed to describe our observations in experiments with long capillary tubes, illustrated in Fig. 2(b). Figure 3(b) presents the measurements of the water imbibition length in the hydrogel-coated capillary tubes. In the early stage ($t < 1$ s), both curves are almost overlapped and show that the square of the imbibition length increases linearly with time ($l^2 \sim t$), as observed in ordinary capillary tubes [14]. After ~ 1 s, the imbibition rate becomes slower, and the experimental results depart from the straight line showing the interdependence of $l^2 \sim t$. In the early stage, the square roots of the slopes of the curves, $(l^2/t)^{1/2}$, were measured to be 28.4 ± 0.4 mm s$^{-1/2}$ for both hydrogels. The expansion of the hydrogel layer is expected to remain insignificant in the early stage, so that the flow speed is primarily determined by capillary action, rather than swelling and water absorption. Therefore, we conjecture that the measured value of 28.4 mm s$^{-1/2}$ corresponds to k in the Washburn equation. This is supported by the fact that two channels with different cross-linker concentrations but with the same initial diameter have the same value of 28.4 mm s$^{-1/2}$. We observed much less expansion of the hydrogel layer in the axial direction, compared to that in the radial
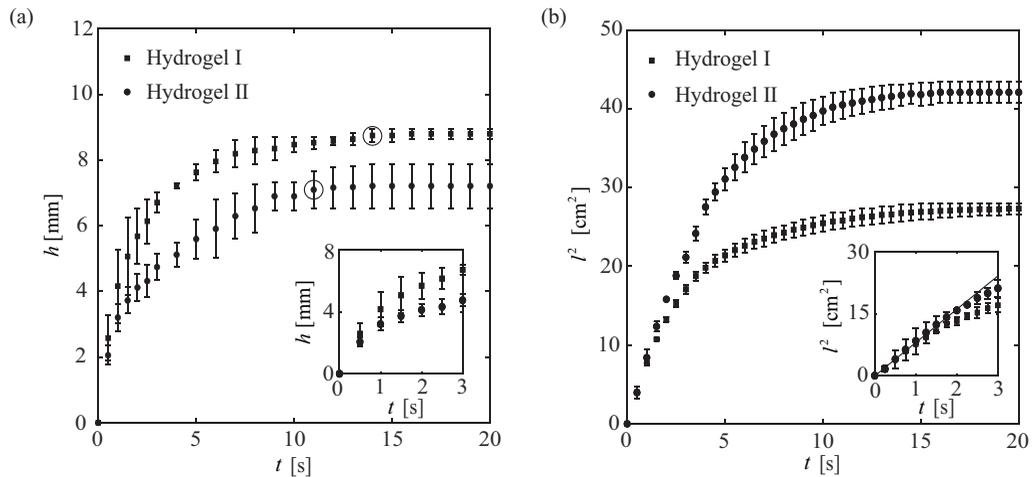
(a)

(b)

FIG. 3. (a) The dependence of the water column height difference h on time t in experiments with cut tube pieces, illustrated in Fig. 2(a). Circles indicate t_e. (b) The dependence of the square of the imbibition length l^2 on time t in long capillary tubes made of hydrogels I and II in experiments illustrated in Fig. 2(b). The experiments were repeated three times, and the error bars indicate the maxmin range.

direction. This is most likely because the hydrogel layer adheres to the rigid glass tube, and so the deformation in the axial direction is restrained.

IV. THEORETICAL ANALYSIS

Water absorption by hydrogel is a kinetic process of water diffusion involving the deformation of hydrogel, which depends on the complex interactions between the polymer network and water. A mathematical description of the process requires a number of physical parameters, such as water diffusivity in hydrogel, viscoelastic modulus of hydrogel, and enthalpy of mixing of water and hydrogel [1,23–27]. However, many of such parameters cannot be easily determined, often making the practical analysis infeasible. For poly (acrylic acid) hydrogels used in our experiments, a phenomenological description based on experimental observation of hydrogel deformation can offer an alternative analytical tool. Ogawa *et al.* [28] experimentally measured the volume change of hydrogel particles and suggested that the volume change can be well described by an exponentially decaying function of time. Similarly, Omidian *et al.* [29] reported that the rate of water mass absorbed by hydrogel particles decreases exponentially with time. The latter study further inferred that the characteristic time dependence of the water uptake rate described by the exponential function is associated with the viscoelastic properties of hydrogels, as would arise in an analogous system consisting of a spring and a dashpot in parallel.

To develop a simple mathematical model on the ground of these phenomenological descriptions of water uptake and deformation of hydrogel, we assume that the volume of water absorbed by the hydrogel layer in a capillary tube, V_w, is given by the following exponential function of time:

$$\frac{V_w}{V_w^\infty} = 1 - e^{-t/\lambda}, \tag{1}$$

where V_w^∞ is the volume of water absorbed when the hydrogel fully expands and occupies the entire volume of the capillary tube, and λ is the time constant for water uptake by hydrogel. Similarly, the increased volume of the expanded hydrogel, V_h, can be expressed as

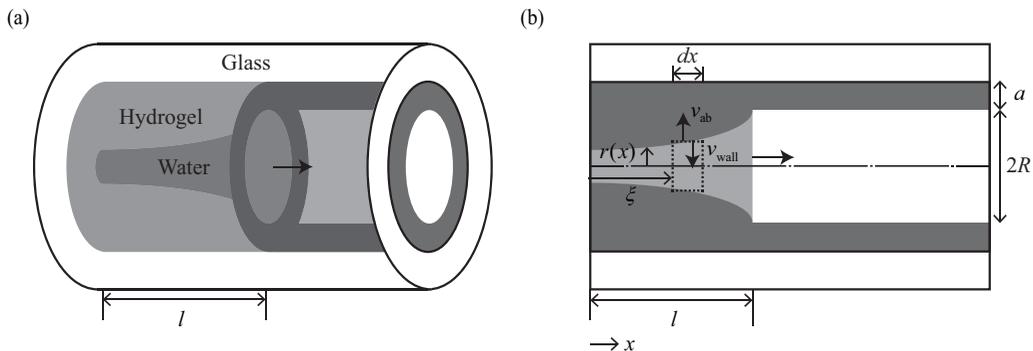$$\frac{V_h}{V_h^\infty} = 1 - e^{-t/\lambda}, \tag{2}$$

FIG. 4. Schematic illustrations of (a) a bird's eye view and (b) a cross-sectional view of the hydrogel-coated capillary tube. Dotted box indicates the control volume used for analysis.

where V_h^∞ is the final increased volume of the expanded hydrogel. Since the hydrogel layer expands and finally occupies the entire space of the tube, V_h^∞ is equal to the volume of the initial void of the tube. As the water uptake of the hydrogel can be assumed to virtually simultaneously occur with swelling [30–33], the same time constant λ is used in (1) and (2). It is noteworthy that the water volume finally absorbed by the hydrogel layer, V_w^∞, is not necessarily the same as the ultimate expanded volume of the hydrogel, V_h^∞.

Figure 4 displays the geometric model adopted to analyze the water imbibition into a capillary tube with consideration of water absorption and deformation of hydrogel. Unlike ordinary capillary tubes, the hydrogel layer in a capillary tube absorbs water and expands inward. Therefore, the tube radius r varies with time and space, so that $r = r(x, t)$ while the radius of the capillary tube at the imbibition front is constant in time.

We consider an infinitesimal segment of the channel with a length of dx at a distance of ξ from the channel inlet, indicated as the dotted box in Fig. 4(b). In the channel segment, the continuity equation for water is given as $\pi r^2 (du/dx) = 2\pi r(v_{\text{wall}} - v_{\text{ab}})$, where u is the local average speed of water flow in the axial direction, v_{ab} is the volume flux of water absorbed by the hydrogel layer, and v_{wall} is the speed of the inward motion of the channel boundary due to the swelling of the hydrogel layer. By integrating the continuity equation from ξ to l, we obtain

$$u(\xi) = l'(t) + \int_\xi^l \frac{2(v_{\text{ab}} - v_{\text{wall}})}{r} \, dx, \tag{3}$$

where l' is dl/dt, and $u(l)$ has been replaced with $l'(t)$.

We proceed with the estimation of v_{ab} and v_{wall}. The volume of the expanded hydrogel in this segment is $\pi(R^2 - r^2) \, dx$ with R being the initial radius of the channel formed in the hydrogel-coated capillary tube. The expanded hydrogel volume can be expressed as $\pi R^2 \, dx(1 - e^{-(t-\tau)/\lambda})$ in (2) with $V_h^\infty = \pi R^2 \, dx$, where τ is the time satisfying $l(\tau) = \xi$, thus implying that $t - \tau$ is the local water absorption time. Equating both expressions enables us to express the tube radius as

$$r(\xi) = Re^{-(t-\tau)/(2\lambda)}. \tag{4}$$

Since $v_{\text{wall}} = -dr/dt$, (4) provides

$$v_{\text{wall}}(\xi) = (R/2\lambda)e^{-(t-\tau)/(2\lambda)}. \tag{5}$$

The rate of the water volume absorbed by the hydrogel layer in the segment, $(2\pi r dx)v_{\text{ab}}$, is given by dV_w/dt in (1). Combining this with $V_h^\infty = \pi R^2 dx$ for the segment and (4), we can express v_{ab} as

$$v_{\text{ab}}(\xi) = \frac{RV_w^\infty}{2\lambda V_h^\infty} e^{-(t-\tau)/(2\lambda)}. \tag{6}$$

The momentum equation for the water flow in the axial direction is obtained from the balance between the capillary force and the viscous resistance. The capillary force is generated by the water-air interface at the imbibition front, and the viscous resistance is caused by shear force developed along the channel wall. Owing to the water absorption and hydrogel swelling, the axial flow speed through the tube varies along the flow direction, and the momentum equation is given in an integral form, $\sigma \cos \theta (2\pi R) = \int_0^l \gamma (2\pi r) \, dx$, where $\gamma = 4\mu u/r$ is the local shear stress on the inner surface of the hydrogel layer. One can rearrange the momentum equation as

$$\int_0^l u \, dx = \frac{1}{2}k^2, \tag{7}$$

where $k = [\sigma R \cos \theta /(2\mu)]^{1/2}$, which is the same parameter that describes $l/t^{1/2}$ in the Washburn equation.

Substituting (3) into (7), we obtain a single equation for $l(t)$:

$$l(t)l'(t) + \int_0^l \int_x^l \frac{2(v_{\text{ab}} - v_{\text{wall}})}{r} \, dx \, dx = \frac{1}{2}k^2. \tag{8}$$

After plugging (4), (5), and (6) for r, v_{wall}, and v_{ab}, respectively, we obtain

$$l(t)l'(t) + \frac{1}{2\lambda}\left(\frac{V_w^\infty}{V_h^\infty} - 1\right)[l(t)]^2 = \frac{1}{2}k^2. \tag{9}$$

Dropping the second term on the left-hand side of (9) restores the Washburn equation, thus suggesting that the effects of water absorption and swelling of the hydrogel layer are included in the second term. We nondimensionalize (9) using the timescale λ and length scale $k\sqrt{\lambda}$, and the governing equation in terms of dimensionless parameters $\tilde{t} = t/\lambda$ and $\tilde{l} = l/(k\sqrt{\lambda})$ is given by

$$\tilde{l}(\tilde{t})\tilde{l}'(\tilde{t}) + \frac{1}{2}(\varepsilon - 1)[\tilde{l}(\tilde{t})]^2 = \frac{1}{2}, \tag{10}$$

where $\varepsilon = V_w^\infty/V_h^\infty$ represents the volume ratio of water ultimately absorbed by the hydrogel layer to the initial void space in the hydrogel-coated tube.

In (10), the relative importance of the term added to the Washburn equation, $\frac{1}{2}(\varepsilon - 1)[\tilde{l}(\tilde{t})]^2$, is determined by ε. For $\varepsilon = 1$, the hydrogel layer locally expands by the volume of water absorbed in the hydrogel layer, so that $v_{\text{ab}} = v_{\text{wall}}$. Accordingly, the hydrogel expansion and water absorption do not affect the water flow in the axial direction, and (10) is restored to the Washburn equation, $\tilde{l}(\tilde{t})\tilde{l}'(\tilde{t}) = \frac{1}{2}$. When $\varepsilon < 1$, the increased volume of the hydrogel layer is greater than the water volume absorbed in the hydrogel. This means that the hydrogel layer locally expands more than it absorbs water, driving water forward in the tube. As a result, the imbibition speed is accelerated by the action of the hydrogel layer. In contrast, when $\varepsilon > 1$, the hydrogel layer locally absorbs a larger volume of water than it expands, and thus the action of the hydrogel layer delays imbibition.

One can solve (10) with the initial condition $\tilde{l}(0) = 0$, and the solution is given by

$$\tilde{l}(\tilde{t}) = \left[\frac{1}{\varepsilon - 1}(1 - e^{-(\varepsilon-1)\tilde{t}})\right]^{1/2}. \tag{11}$$

In (11), $\tilde{l}(\tilde{t})$ converges to $\tilde{t}^{1/2}$ as $\varepsilon \to 1$. The results of (11) are shown in Fig. 5(a), where $\tilde{l}^2$ is plotted with respect to $\tilde{t}$ for $\varepsilon = 0.8$, 1, 1.2, and 1.5. Note that the solid line for $\varepsilon = 1$ coincides with the Washburn equation ($\tilde{l}^2 = \tilde{t}$). In the early stage ($\tilde{t} \ll 1$), all curves collapse to a single straight line, $\tilde{l}^2 = \tilde{t}$ regardless of ε. This result implies that when the absorption and swelling of hydrogel are sufficiently slow ($\lambda \to \infty$), they have little impact on the imbibition dynamics because the solution curves are restored to the Washburn equation (solid line) as $\tilde{t} \ll 1$. For $\varepsilon < 1$, the present model (11) is curved upward and predicts a faster imbibition speed than that of the flow described by the Washburn equation. For $\varepsilon > 1$, the result is the opposite.
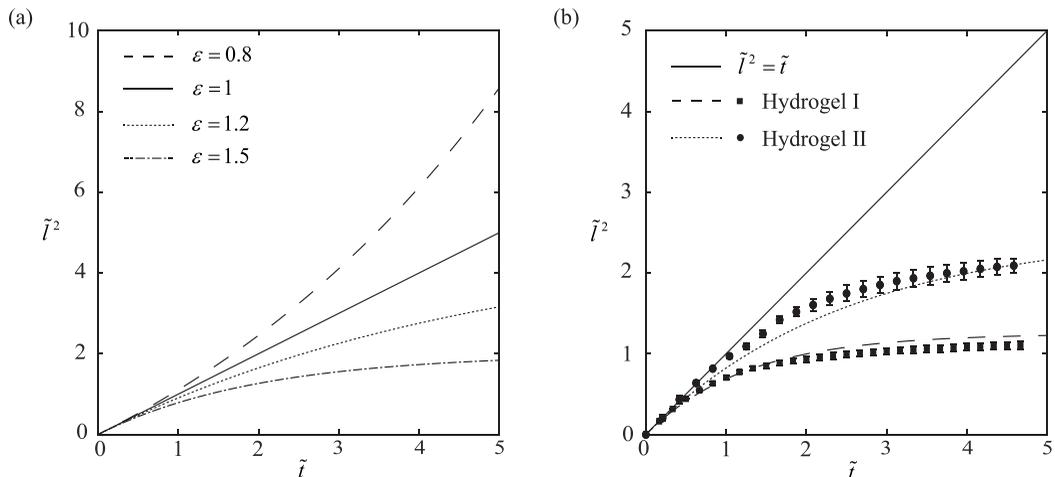
FIG. 5. (a) Prediction of dependence of $\tilde{l}^2$ on $\tilde{t}$ for various ε values by (11). (b) Comparison between theoretical predictions and experimental measurements of imbibition length of water through the capillary tubes coated with hydrogel I or II. The measurements of t_e, λ, and ε for each hydrogel are listed in Table I.

V. COMPARISON OF MODEL PREDICTIONS AND EXPERIMENTAL RESULTS

We compare model predictions to experimental measurements. To predict the imbibition length with (11), we need to determine the parameters listed in Table I. According to the definition of t_e, the time taken to complete 99% of the maximum expansion of the hydrogel [see Fig. 3(a)], we know that the volume of water absorbed by the hydrogel layer $V_w \approx 0.99V_w^\infty$ at $t = t_e$. This is plugged into (1), and the time constant for water uptake by hydrogel is expressed as $\lambda \approx t_e/4.6$. We measured t_e for the capillary tube coated with hydrogels I and II to be 14 s and 11 s, and so $\lambda \approx 3.0$ s and 2.4 s, respectively. We also measured the volume of water absorbed in the fully expanded hydrogel (V_w^∞) for 5 mm cut hydrogel-coated tube pieces as 0.58 and 0.47 mm^3 for hydrogels I and II, respectively. One can estimate the final increased volume of the expanded hydrogel (V_h^∞) for a cut tube piece as the volume of the initial void space formed in the hydrogel layer with an inner diameter of 290 μm, so that $V_h^\infty \approx 0.33$ mm^3. From the ratio of V_w^∞ to V_h^∞, we estimate the values of ε as 1.76 and 1.42 for hydrogel I and II, respectively. In general, t_e, λ, and ε depend on the hydrogel properties, the diameter of the capillary tube, and the thickness of the hydrogel layer. However, hydrogel-coated capillary tubes with the same geometry were used for hydrogels I and II in our experiments, so that the differences in measurements of these parameters are entirely due to the hydrogel properties. Specifically, the distance between two adjacent crosslinks of a hydrogel depends on the cross-linker concentration, so the swelling time and the volume of the absorbed water are different for the two hydrogels.

Figure 5(b) displays a comparison between the experimental measurements and the model predictions of water imbibition through the hydrogel-coated capillary tubes before the swollen

TABLE I. Experimental estimations of parameters of capillary tubes made of hydrogels with different cross-linker concentration C.

Hydrogel	C [g/ml]	t_e [s]	λ [s]	ε
I	0.01	14.0 ± 1.0	3.0 ± 0.2	1.76 ± 0.03
II	0.02	11.0 ± 1.0	2.4 ± 0.2	1.42 ± 0.10

hydrogel layer completely occupies the void ($0 < \tilde{t} < t_e/\lambda$). The model predictions are in good agreement with experimental measurements. It is clearly shown that the imbibition speeds measured for both hydrogels are smaller than those predicted by the Washburn equation (solid line). The poly (acrylic acid) hydrogel layers used in our experiments absorb more water than they expand, thus $\varepsilon > 1$. In particular, hydrogel I with less cross-linker concentration absorbs more water than hydrogel II. The results thus show that the imbibition speed through the channel made of hydrogel I with $\varepsilon = 1.76$ is less than that made of hydrogel II with $\varepsilon = 1.42$. This impacts the imbibition pattern, consistent with theory [Fig. 5(b) and Eq. (11)].

VI. OTHER DEFORMABLE, ABSORBENT CHANNEL FLOWS

We here attempt to extend the present physical understanding of capillary flow through hydrogel-coated channels to flows through other deformable, absorbent channels. Granular hydrogels are used to absorb water in a variety of applications, such as hygienic products, drug delivery systems, and microfluidic pumps, where absorbing large amounts of water at a high rate is often desirable. Because granular hydrogels allow advection flow through interconnected pores by the capillary action, which is characterized by parameter k, they are markedly superior to bulk hydrogels in terms of water absorption flux. Nevertheless, when hydrogel granules expand to fill the void space for advection, the rapid capillary flow is no longer available, so that delayed pore clogging of hydrogel granules at the inlet for water influx is advantageous. Consequently, it is inferred that the hydrogel granules with larger t_e and k can be effectively used for rapid water absorption, and hydrogel granules farther from the inlet than the final water infiltration length, $l(t = t_e)$, do not participate significantly in rapid water absorption.

For applications such as hygienic products in the form of planar absorption pads, the amount of water absorbed per unit area in contact with water is of primary interest. This quantity corresponds to $V_{\text{tot}}/(\pi (R + a)^2)$ in our study, where a is the initial thickness of the hydrogel layer, and V_{tot} is the volume of water absorption for $0 < t < t_e$, which can be calculated as $V_{\text{tot}} = \int_0^{t_e} \pi [r(0, t)]^2 u(0, t) \, dt$. Expressing $V_{\text{tot}}/[\pi (R + a)^2]$ using (3) and (4) yields

$$\frac{V_{\text{tot}}}{\pi (R + a)^2} = \frac{k\sqrt{\lambda}}{1 + (a/R)^2} \int_0^{\tilde{t}_e} e^{-\tilde{t}} [\tilde{l}'(\tilde{t}) + (\varepsilon - 1)\tilde{l}(\tilde{t})] \, d\tilde{t}. \tag{12}$$

Noting that (12) has a unit of length, a dimensionless parameter can be defined as $\tilde{V}_{\text{tot}} = V_{\text{tot}}/[\pi (R + a)^2 l(t_e)]$, which is given by

$$\tilde{V}_{\text{tot}} = \frac{\int_0^{\tilde{t}_e} e^{-\tilde{t}} [\tilde{l}'(\tilde{t}) + (\varepsilon - 1)\tilde{l}(\tilde{t})] \, d\tilde{t}}{[1 + (a/R)^2]\tilde{l}(\tilde{t}_e)}. \tag{13}$$

Here the denominator, $\pi (R + a)^2 l(t_e)$, represents the total volume occupied by water and hydrogels, so that $0 < \tilde{V}_{\text{tot}} < 1$. For a given set of t_e and k, (13) shows that $\tilde{V}_{\text{tot}}$ depends on two dimensionless parameters, ε and a/R. Figure 6 presents the result of numerical calculation of the dependence of $\tilde{V}_{\text{tot}}$ on ε for a/R values of 0.1, 1, 1.5, and 2. It is expected that a/R primarily depends on the porosity of granular materials. Once one experimentally estimates ε and a/R of a granular hydrogel material with a confined volume, the volume of water absorbed by the material before pore clogging at the inlet can be directly predicted from Fig. 6.

In some lab-on-a-chip systems, rapid water transport through channels is of importance for reducing the operation time. In microfluidic devices relying on capillary flow, increasing the flow speed by changing the channel radius is highly limited. In this case, our study proposes design parameters to enhance flow speed. Specifically, the flow speed can be improved compared to flows through stiff channels using materials with $\varepsilon < 1$, as shown in Fig. 5(a). This suggests that channels constructed with materials for which $\varepsilon < 1$ can be advantageously used for rapid transport in lab-on-a-chip systems.
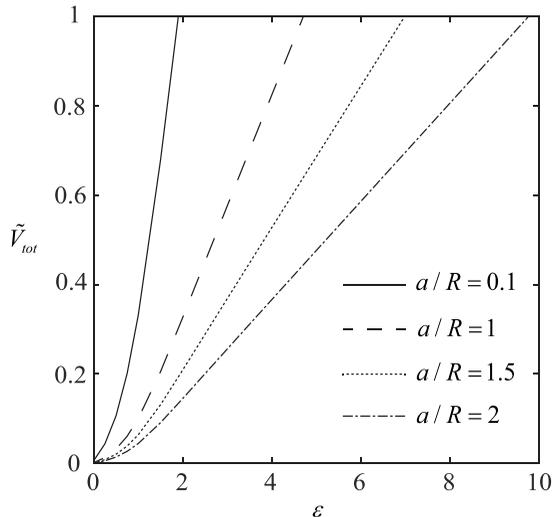
FIG. 6. The dependence of $\tilde{V}_{\text{tot}}$ on ε for various a/R.

VII. CONCLUSIONS

We have studied the dynamics of capillary imbibition of water through hydrogel-coated capillary tubes. In Fig. 3 we have experimentally demonstrated that hydrogel layers cause water uptake and deformation of flow channels and thus affect the dynamics of water flow. By introducing a dimensionless parameter ε that quantifies the volume ratio of water uptake and expansion of the hydrogel layer, we have developed a mathematical model, expressed as (11), for water imbibition through hydrogel-coated capillary tubes. Our model shows a good agreement with experimental results, as demonstrated in Fig. 5(b), and elucidates the effects of the water absorption and expansion of hydrogel on the water flow. Our study provides a theoretical framework for understanding the water flow through hydrogel granular materials, and we expect that it can be extended to capillary flow through various hygroscopic soft porous materials.

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