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Electrocapillary Rise in Nanoporous Media

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Abstract

Capillary flow has been extensively studied due to its broad applications in both engineering and sciences. Detailed studies have shown that capillary rise in hydrophilic microchannels obeys the Lacus-Washburn law, which illustrates a square-root relationship between the imbibition height and the time. The imbibition kinetics depends on the medium structures, liquid viscosity as well as the wetting behavior. These properties are hardly tunable for common porous materials such as rock and paper so as to achieve active control on the imbibition process. In this work, we investigate controllable capillary rise in novel conductive nanoporous materials, i.e., nanoporous gold (NPG) and carbon nanotube (CNT) sponges, by employing the electrocapillary technique to manipulate the wetting property of water on the material surface. Reversible on-off switchable control on the capillary flow in both materials is realized using ultra-low electric voltages. In comparison with NPG, more pronounced flow rate has been obtained in CNT sponges, and, moreover, direct oil recovery can also be realized with the assistance of water-soluble surfactant.

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Keywords: electrocapillary, control, switch behavior, voltage, nanoporous media

1. Introduction

Nomenclature	
h	capillary rise height
r	pore size

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t	imbibition time
σ_{lv}	liquid-vapor interfacial tension
σ_{sl}	solid-liquid interfacial tension
σ_{sv}	solid-vapor interfacial tension
θ	contact angle
η	viscosity of imbibed liquid
τ	tortuosity of the porous media
U	electrode potential
U_{pzc}	potential of zero charge
q	charge density
L	sample length

1.1. Capillary rise

Water spontaneously penetrates hydrophilic porous media such as soil, rock and paper. The invasion process is susceptible to many factors, for instance, temperature, humidity and other environmental conditions[1]. Experiments have shown that the invasion rate is proportional to the pressure gradient, as described by the classical Darcy's law. It is simplified to the Lacus-Washburn law when the capillary pumping pressure is the only driving force[2]. The Lacus-Washburn law shows that the capillary rising height (h) is proportional to the square-root of time (t), that is,

$$h = \sqrt{\frac{\sigma_{lv}\cos\theta}{2\eta}\frac{r}{\tau}t} \tag{1}$$

Equation 1 well describes the case when the inertial and gravitational forces are negligible[3]. On the other hand, at the initial filling phase, the flow is dominated by the inertial force, and the rising height follows a linear relation with time. The validation of equation 1 in microscale has been demonstrated by various experiments. Recently, researchers have shown great interest in the capillary rise behavior in nanometer scale. In such scale, size-dependent line tension and liquid slip effects may deviate the imbibition behavior from the classic 1/2-power law. Molecular dynamics (MD) simulations and experiments have been dedicated to a better understanding of the phenomenon. It is shown by MD simulations that the dependence of the contact angle on the filling velocity slows down the overall invasion rate[4, 5]. However, the experiment using nanoporous Vycor glass with pore size around 3 to 5 nanometers demonstrates the Lacus-Washburn law by measuring the imbibed liquid mass with time. They derived an effective pore size that is smaller than the hydrodynamic one. It is resulted from the strong adsorption of pre-wetting water layer on the glass surface[6], and such immobile layers of molecules also exist in the flow of linear alkanes[7] and liquid crystal[8]. Thus the size effect does exhibit great influence on the capillary flow, and the application of the Lacus-Washburn law may need to consider ingenious modification of the no-slip boundary condition[9].

1.2. Electrocapillary rise

From equation 1, it is easy to see that the imbibition kinetics is related to the pore structures (pore size and tortuosity), liquid properties (viscosity and surface tension) and wetting behavior (contact angle). The intrinsic properties of the solid skeleton and the liquid can hardly be manipulated during the capillary rise process. Thus, it is quite challenging to achieve an active control of the flow, which is needed for micro/nano fluidic and bio-system applications. On the other hand, electrowetting provides an opportunity to tune the wetting behavior between the liquid and the electrode surface[10]. Thus, we can explore the electrocapillary effect to control the wetting property of a conductive porous media so as to realize an active control of the capillary flow.

Based on the generalized Gibbs interfacial thermodynamic theories, the electrocapillary effect is described by the Lippmann's equation, which is expressed as,