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Micro-model experiments and pore network simulations of liquid imbibition in porous media

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HIGHLIGHTS

Wetting experiments with an etched micro model for mixtures of ethanol and water.

- Imbibition kinetics of water mixtures can be described by the Lucas–Washburn.
- Development of a pore network model for spontaneous capillary imbibition.
- Comparison of optical experimental results with pore network simulations.
- Influence of pore structure on the imbibition dynamics by network simulations.

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ABSTRACT

In this study, spontaneous capillary imbibition into an air-filled transparent etched silicon-glass micromodel is investigated by optical imaging under ambient conditions for mixtures of ethanol and water. Images of the micro-model are acquired by a high-speed CCD camera. The binarized images allow us to obtain the overall imbibition kinetics and the time evolution of the phase distribution. A pore network with the structure parameters of the physical micro-model is generated, and a wetting algorithm that combines several pore-level liquid transport rules is developed to simulate the spontaneous imbibition of the liquid mixture into the network. The pore network simulations are able to reproduce the effects observed in the micro-model experiments. The influence of spatially correlated structural features on the imbibition dynamics is studied by further pore network simulations, and pore structures that cause weak or strong capillary imbibition are identified. The results of wetting simulations are shown for situations which cannot be treated by the Lucas–Washburn equation.

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1. Introduction

The capillary imbibition of water and other liquids into gasfilled porous media occurs in various fields of research, such as soil science [\(Blunt et al., 2002](#page--1-0)), oil recovery ([Piri and Blunt, 2004;](#page--1-0) [Yadali Jamaloei and Kharrat, 2010\)](#page--1-0), food industry [\(Saguy et al.,](#page--1-0) [2005\)](#page--1-0), construction [\(Leventis et al., 2000](#page--1-0)), and paper coating ([Ghassemzadeh et al., 2001;](#page--1-0) [Ghassemzadeh and Sahimi, 2004](#page--1-0)), to name only a few. The ongoing issue in the study of capillary flow in porous media is to identify how both the physical properties of the penetrating fluid and the structural characteristics of the porous media affect the imbibition kinetics and the properties of the advancing liquid-gas interface. The detailed description of capillary imbibition is usually complicated due to the intricate

* Corresponding author. E-mail address: abdolreza.kharaghani@ovgu.de (A. Kharaghani). pore structure of hosting media and the local interaction of interfaces. The quasi-steady state capillary flow in a porous medium made of cylindrical tubes with constant cross-section was quan-titatively described more than a century ago ([Bell and Cameron,](#page--1-0) [1906](#page--1-0); [Lucas, 1918](#page--1-0); [Washburn, 1921](#page--1-0)): in a capillary tube, the distance which the liquid has traveled in time t is proportional to the square root of time $(t^{1/2})$ – this relationship is known as the Lucas-Washburn equation. It is derived by combining the Hagen–Poiseuille equation for the viscous flow of a fluid with the Young– Laplace equation for the capillary pressure across the interface between two static fluids. The validity of Lucas–Washburn scaling law has been assessed for capillary imbibition of liquid water into porous media with pore sizes ranging from nanometers [\(Gruener](#page--1-0) [and Huber, 2009](#page--1-0), [2011;](#page--1-0) [Gruener et al., 2012](#page--1-0)) up to centimeters ([Dullien, 1979](#page--1-0); [Sahimi, 1993,](#page--1-0) [2011](#page--1-0)). The same power law has also been observed during the imbibition of water/ethanol droplet mixtures in packed beds of glass beads ([Yang et al., 1988;](#page--1-0) [Jazia](#page--1-0) [et al., 2013\)](#page--1-0). Note that the credibility of this scaling law depends on the viscosity of fluids involved during the imbibition process. It is valid for co-current imbibition when the displaced fluid has low viscosity (i.e. the so-called Washburn situation). It is also valid for counter-current imbibition for two viscous fluids since the imbibition proceeds by a self-similar front which is formed with the resistance behind the front being proportional to the distance the front has propagated into the medium. However, it fails for cocurrent imbibition with two viscous fluids. It also fails for spherical and radial co- and counter-current imbibition, particularly when the front advances outwards ([Mason and Morrow, 2013](#page--1-0)).

Much research work has been done in the development of mathematical models for capillary-driven imbibition (spontaneous imbibition) that take into account the geometrical complexity of pore structures, i.e., pores with noncircular cross-section ([Van der](#page--1-0) [Marck et al., 1997;](#page--1-0) [Polzin and Choueiri, 2003;](#page--1-0) [Cai et al., 2014\)](#page--1-0), capillaries with a monotonically varying radius [\(Young, 2004;](#page--1-0) [Reyssat et al., 2008](#page--1-0)), and tortuous capillaries with various aperture shapes [\(Cai et al., 2014\)](#page--1-0), for example. The research publications cited up to here deal with the modeling of imbibition in capillary tubes or in porous structures which are homogeneous at the macroscopic scale. In such porous media, the wetting front propagates uniformly; hence, the problem can be reduced to one space dimension [\(Kang et al., 2013\)](#page--1-0).

[Reyssat et al. \(2009\)](#page--1-0) developed a continuum model based on Darcy's law to interpret experimental results of spontaneous liquid imbibition in composite structures made of both fine and coarse porous regions. These regions are arranged side-by-side – either with a positive or a negative porosity gradient in the flow direction – which gives rise to a heterogeneous pore structure at the macroscopic scale. Following previous work, [Shou et al. \(2014\)](#page--1-0) developed a physics-based analytical model to study the macroscopic behavior of liquid imbibition into a porous structure made of two layers with distinct heights and porosities. [Cheng et al. \(2015\)](#page--1-0) developed a one-dimensional continuum model to study the spontaneous imbibition of water into air-filled fractured porous rocks. A rock sample with a fracture represents a heterogeneous medium, where the fracture can be considered as a long macro channel inside the porous matrix. Although the analytical results of Cheng et al. reflect fairly accurately the imbibition kinetics in these composite structures, a more detailed model is indispensable to explain the pore-level imbibition dynamics in a physically and numerically sound way. Also note that this continuumscale approach assumes that both pore structure and liquid distribution can be homogenized for a description with constant or smoothly varying parameters and variables. Therefore, these models are neither suited for percolation phenomena with fractal and discontinuous liquid clusters, nor can porous media be properly described whose sample size is not much larger than pore size (as for thin porous layers), because in both cases the two scales cannot be separated.

In order to account for the structural details of porous media and for the pore-level physics of transport phenomena therein, 2D and 3D pore network models (PNMs) have advanced considerably since the pioneering work of [Fatt \(1956\)](#page--1-0). A 2D pore network consists of horizontal and vertical throats with statistically distributed cross-sectional areas; the nodes between the throats act as a spatial grid for the computation of fluid pressure fields. With this simple pore structure, the transport dynamics are determined on the pore level. The flow in each throat is of Poiseuille type, and the advancement of the overall imbibition front is driven by the global pressure difference as well as by local fluid mass conservation at each node of the network. The results of PN simulations are reliable only if the real pore space morphology is represented accurately by the pore network. Established 2D and 3D image acquisition and reconstruction techniques are often used to characterize the pore space topology and geometry of real porous media samples, such as serial sectioning imaging [\(Vogel, 1997\)](#page--1-0), laser confocal microscopy ([Mon](#page--1-0)[toto et al., 1995](#page--1-0)), and synchrotron X-ray tomography or microtomography [\(Al-Raoush and Willson, 2005](#page--1-0); [Wildenschild and](#page--1-0) [Sheppard, 2012;](#page--1-0) [Wang et al., 2012;](#page--1-0) [Beckingham et al., 2013](#page--1-0)). The structural information obtained from these imaging techniques can be used to generate a pore network model. Recent improvements in the fabrication of transparent micro-models and in high-speed imaging provide a spatial and temporal resolution high enough to track the imbibition process at the pore scale and to obtain quantitative information on the time evolution of the interface. Micro-models are artificial porous media consisting of a microfluidic pore network with prescribed geometry. They have contributed significantly to identifying the fluid flow mechanisms [\(Lenormand et al., 1983](#page--1-0); [Yadali Jamaloei and Kharrat, 2010\)](#page--1-0) as well as the transport properties at the pore level ([Cheng et al.,](#page--1-0) [2004](#page--1-0); [Perrin et al., 2006](#page--1-0); [Karadimitriou and Hassanizadeh, 2012\)](#page--1-0). These mechanisms and properties were subsequently incorporated into pore network models resulting in a more accurate representation of the pore-scale physics ([Blunt, 2001](#page--1-0); [Mah](#page--1-0)[mud and Nguyen, 2006;](#page--1-0) [Joekar-Niasar et al., 2009;](#page--1-0) [Joekar-Niasar](#page--1-0) [and Hassanizadeh, 2012a,](#page--1-0) [2012b;](#page--1-0) [Lux and Anguy, 2012](#page--1-0)).

Thompson ([Thompson, 2002](#page--1-0)) demonstrated the great potential of pore network modeling for solving capillary flow problems in disordered fibrous materials. Various prototype fiber networks with, e.g., different network structure, pore spatial correlation, or solid volume fraction, were constructed based on Voronoi diagrams [\(Voronoi, 1908](#page--1-0)). Water invasion simulations with a spatially heterogeneous structure were performed under both spontaneous, due solely to capillary forces, and forced at some volume rate of flow conditions. [Bazylak et al. \(2008\)](#page--1-0) studied numerically and experimentally liquid invasion into heterogeneous pore networks with radial or diagonal pore size gradient. Although similar flow patterns are obtained from simulations and experiments, the numerical model employed ignores the dynamic effects of the water transport. Hence, the structural effects on the imbibition rate cannot be explained by their models. For a comprehensive review on recent developments in spontaneous imbibition refer to [\(Alava](#page--1-0) [et al., 2004](#page--1-0); [Mason and Morrow, 2013](#page--1-0)).

Note that the exact position and shape of the fluid-fluid interface in the pores is often ignored by PNMs, yet it is known to strongly influence the fluid cluster properties during capillary force driven fluid displacement. Direct numerical simulations using level set ([Prodanovi](#page--1-0)[ć](#page--1-0) [and Bryant, 2009\)](#page--1-0), lattice-Boltzmann ([Porter et al., 2009\)](#page--1-0), and volume-of-fluid [\(Ferrari and Lunati, 2013;](#page--1-0) [Kharaghani et al., 2013](#page--1-0)) methods have been able to resolve the sub-pore scale physics and can serve as benchmarks to validate both microscopic pore network and macroscopic continuum models.

In this paper, optical measurements of the spontaneous imbibition of different liquid mixtures of ethanol and water into an etched glass micro-model are presented. The time evolutions of the imbibed liquid volume and of the liquid distribution are determined from two-dimensional images acquired by a high speed CCD camera. A pore network wetting model is briefly recalled ([Sun, 2014](#page--1-0)). The experimental findings are compared to the pore network simulation results. The influence of structural features on the liquid imbibition dynamics is explored by pore network simulations. Finally, prospects for further work, both theoretical and experimental, are discussed.

2. Experimental setup and procedure

The experimental setup consists of a microfabricated silicon network, a synchronized data acquisition system, and an imaging Download English Version:

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