



Short communication

Prediction of capillary air-liquid interfacial area vs. saturation function from relationship between capillary pressure and water saturation

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ABSTRACT

This short communication investigates if the capillary air-liquid interfacial area vs. saturation relationship $A_{lv}(S)$ can be predicted from the capillary pressure vs. saturation relationship $S(h)$, using the theoretical sample scale model of Diamantopoulos and Durner (2013, 2015). We selected three published experimental datasets, where $S(h)$ and $A_{lv}(S)$ relationships had been measured for the same porous media. The sample scale model was fitted to the retention curve $S(h)$ of each porous medium and then used to predict the air-liquid interfacial area $A_{lv}(S)$. We also included in the analysis the thermodynamic models of Leverett (1941) and Grant and Gerhard (2007). For two sandy materials and especially for high saturation values, the model predicted the capillary $A_{lv}(S)$ successfully, which was in one case given by a pore-network model simulation (Kibbey and Chen, 2012) and in the other case experimentally determined (Brusseu et al., 2006). For glass bead experiments, the contact angle needed to be fitted to properly describe the experimental $A_{lv}(S)$ curve.

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1. Introduction

The air-water interfacial area (A_{lv}) is an important parameter for characterizing phase distribution in unsaturated porous media. Hassanizadeh and Gray (1993), based on theoretical considerations, pointed out that different pore-scale air-water configurations may appear for the same porous medium at a given water content or saturation value. For this reason, an accurate description of hydraulic properties and multiphase flow in porous media must account for the geometry of the air-water interfaces (Chen et al., 2007a,b). This highlights the importance of the A_{lv} vs. liquid saturation (S) relationship, which is ignored within the more commonly applied single-phase unsaturated flow models (Niessner et al., 2005).

Determination of A_{lv} is a non-trivial task. Generally, two types of methods are applied to determine $A_{lv}(S)$: tracer-based methods (Brusseu et al. 1997; Kim et al. 1997; Costanza-Robinson and Brusseu 2002) and imaging-based methods (Cheng et al., 2004; Culligan et al. 2004; Brusseu et al. 2006; Chen et al. 2007a; Porter et al. 2009). Both techniques have been widely used for determination of the $A_{lv}(S)$ relationship. However, recent discussions debate

the type of interfaces that are measured by the two methods, respectively. It is generally agreed that tracer methods measure both, the capillary air-water interface and the film air-water interface, whereas the imaging techniques measure only capillary air-water interface area, A_{lv} (Brusseu et al. 2006).

An alternative method to investigate the relationship between interfacial area and saturation is the use of synthetic porous media (Karadimitriou and Hassanizadeh, 2012). A few studies exist, which investigate the $A_{lv}(S)$ relationship in synthetic micro-models (Chen et al., 2007; Pyrak-Nolte et al. 2008).

Diamantopoulos and Durner (2013) developed a pore scale analysis for liquid retention and unsaturated hydraulic conductivity in angular capillary tubes as a function of the contact angle. Their analysis was based on the work of Tuller et al. (1999) and Or and Tuller (1999) without the consideration of films. For upscaling from the pore scale to the sample scale they assumed that the pore-size distribution can be represented by a special Gamma density function. Diamantopoulos and Durner (2015) increased the flexibility of their model by alternatively assuming a lognormal pore-size distribution. Their work provides analytical solutions for liquid retention, saturated/unsaturated hydraulic conductivity, liquid-air interfacial area, and specific surface area at the sample scale.

The objective of this short communication is to test the model of Diamantopoulos and Durner (2013, 2015) by predicting the capillary air-water interfacial area vs. saturation relationship from experimental data of $S(h)$ during primary drainage (h : pressure

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Table 1
Porosity, measured grain size and estimated retention curve parameters $S(h)$ data for all materials using the lognormal model of Diamantopoulos and Durner (2015). The contact angle was assumed to be zero for Chen and Kibbey (2006) and Brusseau et al. (2006). For Culligan et al. (2004), the first case also assumes that the contact angle is zero, while the second case used $A_{lv}(S)$ data to fit the contact angle (see Fig. 3).

Citation	Material	Porosity (φ)	Weight percent grain size distribution (mm)	L_m (cm/-)	σ (-)	θ ($^\circ$)
Chen and Kibbey (2006) Kibbey and Chen (2012)	natural sand	0.362	1%: 0.425–0.300 4%: 0.300–0.212 18%: 0.212–0.150 44%: 0.150–0.106 25%: 0.106–0.075 8%: 0.075–0.053	0.006	0.19	–
Brusseau et al. (2006) Culligan et al. (2004)	natural sand glass beads	0.390 0.340	mean d : 0.234 mm 30%: 1–1.4 35%: 0.850 35%: 0.600	0.009 0.07 0.05	0.9 0.16 0.18	– – 47

head). Published data of Kibbey and Chen (2012), Brusseau et al. (2006) and Culligan et al. (2004) were used to predict $A_{lv}(S)$ to compare it with those measured in these works. For comparison, the analysis also applies the thermodynamic models of Leverett (1941) and Grant and Gerhard (2007).

2. Materials and methods

2.1. Experimental data

Chen and Kibbey (2006) measured $S(h)$ and $A_{lv}(S)$ relationships for a fine quartz sand (F-110, Chen et al., 2007b) by using five different concentrations of sodium octybenzene sulfonate (SOBS) as the interfacial tracer. In a later study, Kibbey and Chen (2012) showed that this method calculates both capillary and film (total) interfacial area. To do this, Kibbey and Chen (2012) calibrated a pore-network model (PNM) against experimental $S(h)$ relationship for the same fine quartz sand. Afterwards, based on their model, they calculated total and capillary $A_{lv}(S)$ relationship. The former was compared against the experimentally measured $A_{lv}(S)$.

Brusseau et al. (2006) measured $S(h)$ and $A_{lv}(S)$ relationships for a natural sandy material by using two methods: gas phase partitioning tracer test and synchrotron X-ray microtomography. The first method measures both capillary and film (total) interfacial area whereas the second method can be used for the estimation of the capillary-associated $A_{lv}(S)$ relationship. Similarly, Culligan et al. (2004) used X-ray microtomography to estimate the $S(h)$ and $A_{lv}(S)$ relationships for a glass bead column. We digitized the experimental data describing the primary drainage experiments by using the Web Plot Digitizer (Rohatgi 2012) on the published figures. Table 1 shows the experimental details for all three studies.

2.2. Diamantopoulos and Durner (2015) model

The essentials of the model which describes both sample scale (as opposed to pore scale) $S(h)$ and $A_{lv}(S)$ are presented by Diamantopoulos and Durner (2013). Briefly, the porous medium can be described as a bundle of angular tubes. In this study the tubes are assumed to have a shape of a equilateral triangular with a side length L . Diamantopoulos and Durner (2015) presented an upscaling scheme that assumed that the sample's pore-size distribution, expressed by the probability distribution of the sample scale pore side-length, L , can be described by a log-normal distribution, $f(L)$:

$$f(L) = \frac{1}{L\sigma\sqrt{2\pi}} e^{-\frac{(\ln(\frac{L}{L_m}))^2}{2\sigma^2}}, \quad L > 0. \quad (1)$$

Parameter L_m is the median of the variable L , and σ is the standard deviation of the natural logarithm of L . Their values depend

on porous medium characteristics. Each of the triangular pores can be either fully saturated or partially saturated, dependent on the pressure head h . The macroscopic water saturation of a sample $S(h)$ is calculated by summing up the water saturations in fully saturated and partially saturated pores:

$$S(h) = S_1(h) + S_2(h) \quad (2)$$

The equations for S_1 and S_2 with lognormal distributed pore-side lengths (Eq. (1)) are:

$$S_1(h) = \left[\frac{1}{2} \left(1 + \operatorname{erf} \left(\frac{\ln(\frac{L}{L_m})}{\sigma\sqrt{2}} \right) \right) \right]_{L_{min}}^{L_1(h)} \quad (3)$$

$$S_2(h) = B_n \left[\frac{1}{2} L_m^{-2} e^{2\sigma^2} \left(1 + \operatorname{erf} \left(\frac{\ln(\frac{L}{L_m})}{\sigma\sqrt{2}} + \frac{2\sigma}{\sqrt{2}} \right) \right) \right]_{L_1(h)}^{L_{max}} \quad (4)$$

where

$$B_n = \frac{4r(h)^2 S_n}{\cot^2\left(\frac{180}{n}\right)}. \quad (5)$$

$S_1(h)$ is the sample saturation of all fully saturated pores with a side length L greater than the minimum side length L_{min} and smaller than $L_1(h)$. $S_2(h)$ is the sample saturation of the partially filled pores with $L_1(h) < L < L_{max}$. L_{max} stands for the pore side-length of the biggest pore, which can be calculated from the soil's air entry value or fixed to a relatively high value.

The sample-scale liquid-vapor interfacial area is given by:

$$A_{lv}(h) = \frac{n(180 - 2(\alpha + \theta))\pi r(h)}{180A_n} \times \left[\frac{1}{2} L_m^{-2} e^{2\sigma^2} \left(1 + \operatorname{erf} \left(\frac{\ln(\frac{L}{L_m})}{\sigma\sqrt{2}} + \frac{2\sigma}{\sqrt{2}} \right) \right) \right]_{L_1(h)}^{L_{max}} \quad (6)$$

The variable definition for Eqs. (1)–(6) is given in Appendix A. Note that in our model only the interfaces between the corner fluid and the air phase contribute to the interfacial area, and that $A_{lv}(S)$ depends on the contact angle, θ , between the air-liquid interface and the solid surface at the pore wall.

2.3. Thermodynamic interfacial area model

According to the thermodynamic approach, changes in interfacial area reflect the mechanical work done on the system, the magnitude of which is proportional to the area under the capillary pressure (P_c [M T⁻² L⁻¹]) - saturation relationship (Grant and Gerhard, 2007). Leverett (1941) defined the total interfacial area per unit volume of porous media a^n [L²L⁻³] as:

$$a^n = -\varphi \frac{1}{\sigma_{wa}} \int_1^S P_c(S) dS \quad (7)$$

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