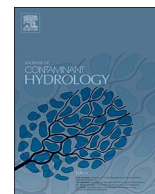




Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

journal homepage: www.elsevier.com/locate/jconhyd

New concept to describe three-phase capillary pressure–degree of saturation relationship in porous media

Keita Nakamura, Mamoru Kikumoto*

Department of Civil Engineering, Yokohama National University, Tokiwadai 79-5, Hodogaya, Yokohama, Kanagawa 240-8501, Japan

ARTICLE INFO

Keywords:

Capillary pressure
Degree of saturation
Nonspreading nonaqueous phase liquid (nonspreading NAPL)
Porous media
Water–nonaqueous phase liquid (NAPL)–air three-phase system

ABSTRACT

The Leverett concept is used conventionally to model the relationship between the capillary pressures and the degrees of saturation in the water–nonaqueous phase liquid (NAPL)–air three-phase system in porous media. In this paper, the limitation of the Leverett concept that the concept is not applicable in the case of nonspreading NAPLs is discussed through microscopic consideration. A new concept that can be applied in the case of nonspreading NAPLs as well as spreading NAPLs is then proposed. The validity of the proposed concept is confirmed by comparing with past experimental data and simulation results obtained using the conventional model based on the Leverett concept. It is confirmed that the proposed concept can correctly predict the observed distributions of NAPLs, including those of nonspreading ones.

1. Introduction

Being able to simulate the seepage behavior of water–nonaqueous phase liquid (NAPL)–air three-phase systems in porous media such as soils is of great importance in geoenvironmental engineering, especially when predicting the ground contamination resulting from the leakage of NAPLs as well as when selecting an efficient remediation method. To predict the seepage flow of NAPLs in a three-phase system, a rational model for the permeability coefficients of the three void fluids is crucial. As the permeability coefficients are primarily determined by the degrees of saturation of the three fluids, the relationship between the capillary pressures and the degrees of saturation of the void fluids needs to be described properly.

In the case of the capillary pressure–saturation relationship in three-phase systems, Leverett (1941) assumed that the NAPL spreads across the water–air interface in a water-wet porous medium and separates the water and air phases. This assumption leads to the following conclusions: (a) the water saturation degree depends only on the capillary pressure between the water and the NAPL; and (b) the liquid saturation degree (which is the sum of the degrees of water saturation and NAPL saturation) depends on the capillary pressure between the NAPL and air phases. Based on this assumption, a number of models for the relationship between the capillary pressure and the degree of saturation for three-phase systems (van Genuchten, 1980; Lenhard and

Parker, 1987a, b, 1988, 1990; Parker and Lenhard, 1987, 1990; Parker et al., 1987; Farr et al., 1990; Blunt et al., 1995; Zhou and Blunt, 1997) have been proposed.

Meanwhile, it is usually believed that the configuration of the three fluids in a void space will necessarily be influenced by their wettability. A few NAPLs such as hexane will spread over the surface of the water phase, while others such as perchloroethylene (PCE) and decane will form a lens-like structure at the water–air interface (McBride et al., 1992; Hofstee et al., 1997). Thus, the Leverett assumption that the NAPL spreads across the water–air interface will not necessarily hold. Hofstee et al. (1997) determined the PCE–air and water–PCE–air retention curves and suggested that the Leverett concept is not applicable in the case of nonspreading NAPLs. In addition, experimental data (Kalaydjian et al., 1993; Vizika and Lombard, 1996; Zhou and Blunt, 1997) have shown that the residual saturation degree of a nonspreading NAPL in water–NAPL–air systems increases with a decrease in the spreading coefficient.

In this study, we first highlighted the limitations of the classical Leverett concept by proving the existence of a critical ratio of the capillary pressures at which the NAPL layer covering the water–air interface cannot exist stably at the microscale (Keller et al., 1997; Fenwick and Blunt, 1998a, 1998b). Next, we propose a new concept for the capillary pressure–degree of saturation relation for three-phase systems in porous media while considering the effects of the spreading coefficient.

* Corresponding author.

E-mail addresses: nakamura-keita-kn@ynu.jp (K. Nakamura), kikumoto@ynu.ac.jp (M. Kikumoto).<https://doi.org/10.1016/j.jconhyd.2018.03.008>Received 25 August 2017; Received in revised form 7 March 2018; Accepted 9 March 2018
0169-7722/ © 2018 Published by Elsevier B.V.

cient of the NAPL. The proposed concept employs the relative magnitude of the NAPL pressure with respect to the water and air pressures as defined by Nakamura and Kikumoto (2014) and should be applicable to any capillary pressure–degree of saturation model (e.g., van Genuchten (1980) and Brooks and Corey (1964)). We also propose a method for taking into account the residual NAPL saturation degree in the three-phase system by using the irreducible (or minimum) degree of saturation of NAPL in the same way as is the case for the residual water in the water–air two-phase system. Finally, the performance of the proposed concept is evaluated based on comparisons with experimental data subject to a monotonic drainage path (Zhou and Blunt, 1997).

2. Overview of existing theories for three-phase systems in porous media

In order to propose a new concept for the capillary pressure–degree of saturation relationship that overcomes the limitations of the classical Leverett concept, we first outline the existing theories for three-phase systems in porous media and their limitations.

2.1. Capillary pressure–degree of saturation relationships for two-phase systems in porous media

Using Laplace's equation, the capillary pressure, P_{cij} , between two fluid phases i and j is given by

$$P_{cij} = P_i - P_j = 2\gamma_{ij}/r_{ij} \quad (1)$$

where P_i and P_j are the pressures of the fluid phases i and j , respectively; γ_{ij} is the interfacial tension between the fluid phases i and j ; and r_{ij} is the radius of curvature of the i – j phase interface. As r_{ij} can be regarded as being a characteristic of the porous medium, Eq. (1) means that the relationship for each combination of phases i and j (i.e., for each i – j system) can be represented by P_{cij}/γ_{ij} (Leverett, 1941 Miller and Miller, 1956).

On substituting the radius of the capillary tube, a , and the contact angle of the fluid phases i and j , θ_{ij} , for r_{ij} in Eq. (1), we obtain the following relationship:

$$\frac{2}{a} = \frac{P_{cij}}{\gamma_{ij} \cos \theta_{ij}} \quad (2)$$

This suggests that the radius, a , is uniquely defined by P_{cij} , γ_{ij} , and θ_{ij} . The degree of saturation is generally determined by the cumulative pore-size distribution, that is, by the pores saturated with the wetting fluid and having an opening radius smaller than a threshold value, a (e.g., Nimmo (2004)). Thus, as a is a function of the effective degree of saturation, \bar{S}_j , we can define a function J using Eq. (2) as follows:

$$J(\bar{S}_j) = \frac{2}{a} = \frac{P_{cij}}{\gamma_{ij} \cos \theta_{ij}} \quad (3)$$

where the subscripts i and j denote the nonwetting and wetting phases, respectively. The inverse function, J^{-1} , also exists:

$$J^{-1}\left(\frac{P_{cij}}{\gamma_{ij} \cos \theta_{ij}}\right) = \bar{S}_j \quad (4)$$

For the water–air two-phase system, the effective degree of saturation of water, \bar{S}_w , is thus given as

$$\bar{S}_w = \frac{S_w - S_w^{\min}}{1 - S_w^{\min} - S_a^{\min}} = J^{-1}\left(\frac{P_{caw}}{\gamma_{aw} \cos \theta_{aw}}\right) \quad (5)$$

where S is the actual degree of saturation, the subscripts w and a denote water and air, respectively, and the superscript min denotes the

minimum (irreducible) degree of saturation. Several equations have been proposed for \bar{S}_w . For instance, van Genuchten (1980) proposed the following one:

$$\bar{S}_w = J_{VG}^{-1}\left(\frac{P_{caw}}{\gamma_{aw} \cos \theta_{aw}}\right) = \{1 + (\alpha P_{caw})^n\}^{\frac{1-n}{n}} \quad (6)$$

with two parameters, α and n . As Eq. (6) states that \bar{S}_w is a function of $\frac{P_{caw}}{\gamma_{aw} \cos \theta_{aw}}$, α is actually a parameter specific to the water–air two phase system and can be represented by

$$\alpha = \frac{a_{VG}}{2\gamma_{aw} \cos \theta_{aw}} \quad (7)$$

where a_{VG} is a constant that is specific to the porous media. Meanwhile, Brooks and Corey (1964) proposed that

$$\bar{S}_w = J_{BC}^{-1}\left(\frac{P_{caw}}{\gamma_{aw} \cos \theta_{aw}}\right) = \left(\frac{P_{caw}}{P_{caw}^d}\right)^{-\lambda} \quad (8)$$

where λ is a material parameter and P_{caw}^d is the entry pressure of air in the water–air two-phase system and given by the following equation:

$$P_{caw}^d = \frac{2\gamma_{aw} \cos \theta_{aw}}{a_{BC}} \quad (9)$$

where a_{BC} is a parameter specific to the porous media.

2.2. Leverett concept and classical capillary pressure–degree of saturation relationships for water–NAPL–air three-phase system in porous media

The Leverett concept (Leverett, 1941) has been usually used to predict the capillary pressure–degree of saturation relationship for three-phase systems based on the relationships for the water–NAPL and NAPL–air two-phase systems. Leverett assumed that, in a water-wet porous medium, the NAPL spreads over the water–air interface, and the water and air phases do not come in contact with each other. This leads to the following: (a) the effective water saturation degree can be determined as a function of the capillary pressure between the water and NAPL phases alone, and (b) the effective degree of total liquid saturation (which is the sum of the degrees of water saturation and NAPL saturation) can be determined as a function of the capillary pressure between the NAPL and air phases alone. Based on Eq. (3) and Leverett's assumption stated above, the effective degree of water saturation, \bar{S}_w , and the effective degree of total liquid saturation, \bar{S}_t , in the three-phase system can be written as

$$J(\bar{S}_w) = \frac{P_{cnw}}{\gamma_{nw} \cos \theta_{nw}} \quad (10)$$

and

$$J(\bar{S}_t) = J(\bar{S}_w + \bar{S}_n) = \frac{P_{can}}{\gamma_{an} \cos \theta_{an}} \quad (11)$$

where J is a function of the effective degree of saturation and describes the capillary pressure for the two-phase fluid, while the subscripts w, n, and a denote water, the NAPL, and air, respectively. Here, the effective degree of saturation, \bar{S}_j , is the effective degree of saturation of the j phase defined by the minimum or irreducible degree of saturation (e.g., Lenhard and Parker (1987a, 1987b)). Eqs. (10) and (11) indicate that the three-phase capillary pressure–degree of saturation relationship can be simply represented by scaling the two-phase relationships (i.e., the water–NAPL and NAPL–air relationships) in the direction of the capillary pressure.

A number of researchers (van Genuchten, 1980 Lenhard and Parker, 1987a, 1987b, 1988, 1990 Lenhard and Parker, 1987a, b, 1990 Parker et al., 1987 Farr et al., 1990 Blunt et al., 1995 Zhou and Blunt, 1997)

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