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Evaluation of the use of capillary numbers for quantifying the removal of DNAPL trapped in a porous medium by surfactant and surfactant foam floods

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Abstract

The capillary number is used to quantify the mobilization potential of organic phases trapped within porous media. The capillary number has been defined in three different forms, according to types of flow velocity and viscosity used in its definition. This study evaluated the suitability of the capillary number definitions representing surfactant and surfactant foam floods by constructing capillary number–TCE saturation relationships. The results implied that the capillary number should be correctly employed, according to scale and fluid flow behavior. This study suggests that the pore-scale capillary number should be used only for investigating the organic-phase mobilization at the pore scale because it is defined by the pore velocity and the dynamic viscosity. The Newtonian-fluid capillary number using the Darcy velocity and the dynamic viscosity may be suitable for quantifying flood systems representing Newtonian fluid behavior. For viscous-force modified flood systems such as surfactant-foam floods, the apparent capillary number definition employing macroscopic properties (permeability and potential gradient) may be used to appropriately represent the desaturation of organic phases from porous media. © 2004 Elsevier Inc. All rights reserved.

Keywords: Capillary number; Foam; Surfactant; Trichloroethylene; Remediation; DNAPL; Porous media

1. Introduction

The remediation of aquifers contaminated by dense nonaqueous-phase liquid (DNAPL) such as trichloroethylene (TCE) and perchloroethylene (PCE) is a national priority. Remediation techniques using surfactants have been used to enhance DNAPL solubility and/or to physically displace entrapped DNAPL ganglia by reducing the interfacial tension (IFT) between the organic phase and the aqueous phase [1,2].

Viscous, capillary, and gravitational forces around a NAPL blob affect the mobilization of NAPL within a porous medium. While the capillary forces act to retain organic phases between the solid grains, the viscous and gravitational forces contribute to mobilize the NAPL blobs.

* Fax: +82-2-380-7744. *E-mail address:* superjeong@yahoo.com. A DNAPL removal pattern by surfactant solution is represented by a relationship between DNAPL saturation and the total trapping number. The total trapping number (N_t) is the sum of the capillary number (N_{ca}) and the bond number (N_b), as shown in Eq. (1) [3]. Here, α is the angle of dip. The capillary number is a dimensionless magnitude and represents the ratio of viscous force to capillary force. The bond number represents the ratio of gravitational force to capillary force:

$$N_{\rm t} = \sqrt{N_{\rm ca}^2 + N_{\rm b}^2 - 2N_{\rm ca}N_{\rm b}\sin\alpha},$$
 (1)

$$N_{\rm b} = \frac{k \kappa_{\rm rw} \Delta \rho_{\rm g}}{\sigma \cos \theta},\tag{2}$$

$$N_{\rm ca} = \frac{\mathbf{v}\mu}{\sigma\cos\theta}, \quad \mathbf{v} = \mathbf{u}/\phi, \tag{3}$$

$$V_{ca}^* = \frac{\mathbf{u}\mu}{\sigma\cos\theta},\tag{4}$$

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$$N_{\rm ca}^{+} = \frac{\mathbf{u}\mu_{\rm ap}}{\sigma\cos\theta},\tag{5}$$

$$N_{\rm ca}^{+} = \frac{\mathbf{u}\mu_{\rm ap}}{\sigma\cos\theta} = \frac{kk_{\rm rw}\nabla\Phi}{\sigma\cos\theta}, \quad \Phi = p + \rho gz, \tag{6}$$

$$\mu_{\rm ap} = \frac{kk_{\rm rw} \nabla P}{\mathbf{u}}.\tag{7}$$

For a homogeneous isotropic porous medium, the bond and capillary numbers have been described as shown in Eqs. (2), (3), (4), and (5). Here, *k* is the permeability, k_{rw} is the relative permeability of the aqueous phase, $\Delta \rho$ is the difference in density between organic and water phases, σ is the interfacial tension, θ is the contact angle between organic and water phases, **v** is the pore velocity, ϕ is the porosity, **u** is the superficial or Darcy velocity, μ is the dynamic viscosity, and μ_{ap} is the apparent viscosity. In Eqs. (6) and (7), $\nabla \Phi$ is the gravitational constant, and *z* is the elevation.

Although the bond number is consistently defined by Eq. (2), the capillary number has been used in several forms, Eqs. (3), (4), and (5), by researchers [4,5]. However, the difference in using the capillary numbers has not received attention from researchers. The objectives of this study were to evaluate the capillary number definitions for quantifying the removal of DNAPL trapped in porous media and to determine an appropriate capillary number definition for surfactant-related remediation system analysis. The relationship between capillary number and DNAPL removal helps to design efficient remediation techniques because the capillary number is dimensionless and gives us important information on flow and interfacial properties.

2. Definition of capillary numbers

The capillary number is usually defined in terms of viscosity, interfacial tension, velocity, and contact angle. However, three capillary number definitions, Eqs. (3), (4), and (5), use different viscosity and flow velocity definitions.

Equation (3) was originally suggested for quantifying the mobilization of an organic phase trapped in a pore [6,7]. Thus, the pore velocity and the dynamic viscosity were used for calculating the pore-scale capillary number. This study calls the capillary number of Eq. (3) as the pore-scale capillary number (N_{ca}). The pore velocity is defined as Darcy velocity divided by the porosity. The dynamic viscosity is determined from the shear rate of capillarity or the pore velocity [8]. Therefore, the upper part of the pore-scale capillary number (Eq. (3)) is defined with pore-scale properties.

In Eq. (4), a macroscopic property, the Darcy velocity, replaces the pore velocity of Eq. (3), but the dynamic viscosity, μ , is still used for the viscosity definition [3,9]. This study calls Eq. (4) the Newtonian-fluid capillary number. The upper part of the Newtonian-fluid capillary number consists of two different scale properties, macroscopic scale (for flow velocity) and pore scale (for viscosity) properties.

Equation (5) uses Darcy velocity for the flow velocity definition and the apparent viscosity for the viscosity definition. The apparent viscosity is the viscosity determined for a non-Newtonian fluid without reference to a particular shear rate. The apparent viscosity would be more compatible with the Darcy velocity than the dynamic viscosity because the apparent viscosity is determined by Eq. (7) using Darcy's law [10,11]. This study calls Eq. (5) the apparent capillary number. Equation (5) can be rewritten using the apparent viscosity definition, Eq. (7), to produce Eq. (6) [12]. In other words, Eqs. (5) and (6) have the same meaning.

It is important to note here that the capillary number of systems representing Newtonian-fluid behavior can be described either by Eq. (4) or by Eqs. (5) or (6) because the apparent viscosity of a Newtonian fluid equals the dynamic viscosity of the fluid. However, if a fluid exhibits non-Newtonian behavior during flow through a porous medium, the values of capillary numbers obtained using Eqs. (4) and (5) could be different because the apparent viscosity and the dynamic viscosity of the fluid are different.

3. Materials and methods

A conventional mobilization scheme is normally designed with a Winsor Type III system which has a middlemicroemulsion phase between an aqueous phase and an oil phase [13]. Some modifications for enhancing mobilization of organic phases have been made to remedial agents. Surfactant foam and polymers can be used to increase the viscosity of the displacing fluid flowing through porous media [14,15]. This study conducted surfactant and surfactant-foam floods for DNAPL removal from a glass porous medium (a porous-pattern-etched glass model). The results were depicted to construct a relationship between DNAPL saturation and capillary number. Three different capillary numbers calculated by three different definitions, Eqs. (3), (4), and (5), were used to construct the relationships. The relationship results were compared each other to evaluate the suitability of the capillary number definitions for representing surfactant and surfactant foam floods.

A porous-pattern glass model would be an excellent tool for DNAPL removal system analysis because the physical properties of the porous medium can easily be quantified and relatively accurate DNAPL saturation can be directly measured by an image analysis technique. The DNAPL saturation was quantified with residual PCE trapped in thousands of pores, representing macroscale DNAPL saturation.

3.1. Experiment procedures

The same homogeneous glass model and experimental setup described in Jeong et al. [16] were used in this study. Therefore, we briefly mention the procedures in this section. TCE (>99.8% purity, Fisher Scientific Inc.) was dyed with 0.5 g Oil-Red-O (Aldrich Chemical)/L of TCE to identify

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