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Journal of Colloid and Interface Science 293 (2006) 455–463

Journal of Colloid and Interface Science

www.elsevier.com/locate/jcis

Existence of fluid layers in the corners of a capillary with non-uniform wettability

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Received 9 December 2004; accepted 19 June 2005

Available online 14 July 2005

Abstract

Based on free energy variation we derive the criterion for displacement during water invasion of oil layers, sandwiched between water in corners and in the centre of a capillary with partly altered wettability. This displacement may arise in combination with a piston-like displacement in which the layers are formed, or, alternatively, these two displacements do not occur and a single piston-like displacement arises removing all oil from the pore cross-section at once. The free energy differentials associated with the three displacements determine exactly which displacement(s) happen during water invasion. Depending on the area and the (advancing) contact angle on the surface of altered wettability, as well as on the half-angles of the pore corners, layers may or may not exist. We compare the criterion for the displacement of oil layers with the existing geometrical criterion. The latter always allows a larger range of contact angles and pressure combination for which layers may exist than the presently derived criterion, hence the geometrical criterion is insufficient and is now superceded.

Keywords: Oil layer; Water invasion; Non-circular cross-section; Non-uniform wettability; Two-phase; Free energy; Capillary entry pressure

1. Introduction

Pore-scale network modelling of multi-phase flow crucially depends on displacement events in individual pores. When pores are modelled as straight tubes of circular cross-section, a given cross-section can be filled by only one phase. Displacement of this phase by another can only occur in a piston-like manner, for which the so-called capillary entry pressure or threshold pressure is described by the well known Young–Laplace equation. However, when non-circular cross-sections are assumed, a given cross-section may be filled by more than one phase, for example in a water-wet pore oil may be present in the centre of the cross-section, while water wetting layers occupy the corners. These layers may swell or shrink when the pressure of its phase is increasing or decreasing, respectively. Furthermore, the capillary entry pressures for the corresponding piston-like displacements are more complicated. For two-phase flow, the so-called MS-P theory for calculating these entry pressures (named after Mayer, Stowe and Princen, the researchers who proposed it) has been known for nearly 40 years and an overview of the literature corresponding to this theory is given by Lago and Araujo [1]. Recently, van Dijke and Sorbie [2] have extended this theory to capillary entry pressures for displacements in three-phase flow.

The above references have only considered capillaries of uniform, though arbitrary, wettability. However, it is well known that part of the pore surface may change wettability and a scenario for this wettability alteration is described by Kovscek et al. [3]. It basically says that in a uniformly water-wet capillary after primary drainage the oil in the centre of the cross-section may render the pore surface oil-wet, while the surface of the pore corners, filled with water wetting layers, remains water-wet. This is described by saying that the intrinsic oil–water contact angle (measured through the water phase) has significantly increased during the transition from water-wet to oil-wet. Additionally, on a given surface the contact angle during a displacement depends on the di-

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0021-9797/$ – see front matter
doi:10.1016/j.jcis.2005.06.059

rection in which this displacement takes place, i.e., contact angle hysteresis, and the above mentioned intrinsic angle is an average of the possible angles. Below, we make our terminology of contact angles more precise for the processes that we are dealing with here.

To explain experimental findings that after water flooding oil could drain to low residual saturations, Kovscek et al. proposed that, based on the envisaged wettability alteration, oil layers are sandwiched between water in corners and in the centre of pores. The key phenomenon in the formation of such layers is that the contacts with the solid of oil–water menisci in the pore corners remain pinned, while the radii of curvature of the menisci adjust to the changing oil–water pressure differences accompanied by changing contact angles. Following this idea, many more cross-sectional fluid configurations have been hypothesised, even including more than one layer, in particular in three-phase flow [4,5]. Notice that in three-phase flow layers of, for example, oil sandwiched between gas and water may arise, even when the surface is uniformly wetted [6–8], because the oil–water and gas–oil contact angles are essentially different.

Ma et al. [9] have described two-phase drainage (oil invasion) and subsequent water flooding in capillaries with non-uniform wettability. In particular, applying the MS-P theory, they derived the capillary entry pressure for piston-like displacement of oil by water, when the corner menisci are pinned to the solid, but their contact angles are allowed to vary. Interestingly, Ma et al. did not comment on the possibility of the sandwiched oil layers. Blunt [10] actually described the formation and collapse of these layers in a pore with square cross-section, which requires that the surface of altered wettability has become relatively strongly oil-wet (large advancing contact angle). Formation of the layers requires a piston-like displacement of oil by water, for which the capillary entry pressure follows straightforwardly from MS-P theory, as no hinging angles are involved. Blunt assumed that the layers would collapse when the surrounding oil–water interfaces met and derived the corresponding geometrical criterion. Many authors have used these criteria for formation and collapse of layers in capillary bundle and network models (e.g., [4,5,10–12]). However, it should be emphasised that the collapse criterion is purely geometrical and is not based on a firm thermodynamic argument. Recently, Piri and Blunt [13] have extended the approach of [2] for three-phase capillary entry pressures to pores of non-uniform wettability, although still using the geometrical criterion for the existence of layers.

Direct observation of oil layers sandwiched between water in the corner and in the centre of a pore has not been reported in the literature, probably because of experimental difficulties. However, indirect measurements of their effects have been made through incorporation of layer criteria in pore network models [10,11] and subsequent comparison of simulated residual oil saturations and oil relative permeabilities at low oil saturations. As expected, these simulations showed oil drainage down to very low oil saturations and corresponding small but non-zero relative permeabilities. However, quantification of the effect has not been possible due to insufficient knowledge of the wettability state of the porous medium and the conductance of the layers. Interestingly, oil layers sandwiched between gas and water in a three-phase system have been studied more extensively, in particular in micromodel experiments by Dong et al. [6] and in single angular tube experiments by Firincioglu et al. [14]. They found reasonable agreement with the corresponding layer criterion.

In this paper we derive a thermodynamic criterion, based on MS-P theory, for the collapse or displacement of the sandwiched oil layers in capillaries with polygonal cross-section and non-uniform wettability, which is more accurate than the geometrical criterion. The idea is to use MS-P theory for the displacement of these layers during water invasion, in fact similar to the displacement of oil layers during three-phase flow in a pore of uniform wettability [8]. In the framework of water invasion, we also revisit the piston-like displacement that leads to the formation of the layers and the alternative piston-like displacement, in which no layers are formed. In Section 2, we describe more accurately the three possible displacements and derive the corresponding capillary (entry) criteria, based on free energy balances. In Section 3, we present some example calculations to investigate when layers may or may not arise and how the various displacements are distinguished. Additionally, we compare the newly derived criterion with the existing geometrical criterion.

2. Capillary entry pressures during water invasion

2.1. Fluid configurations and displacements

We consider a pore of polygonal cross-section, of which the central part has been rendered oil-wet after drainage, such as for the star-shaped pore shown in Fig. 1a. During primary drainage the pore is assumed to have been sufficiently strongly water-wet, such that after oil invasion water wetting layers remained in the pore corners, creating an arc meniscus (AM1), such as shown in configuration a of Fig. 1b. Drainage is supposed to have taken place at a receding contact angle θ_r. After drainage, water is allowed to invade again and any movement on the rendered surface occurs at the (increased) advancing angle θ_a. The difference between θ_r and θ_a forces the contact of AM1 with the solid to be temporarily fixed during water invasion, while its curvature changes.

Let corner configuration a in Fig. 1b represent a cross-sectional fluid distribution after drainage has taken place and the wettability of the central parts of the pore walls has changed. Additionally, water invasion has started, causing the water wetting layer to swell, but no displacement in the centre of the pore has taken place. On the other hand, corner configurations b and c represent the possible distributions after distinct displacements in the pore centre have occurred. The three configurations are distinguished, because either