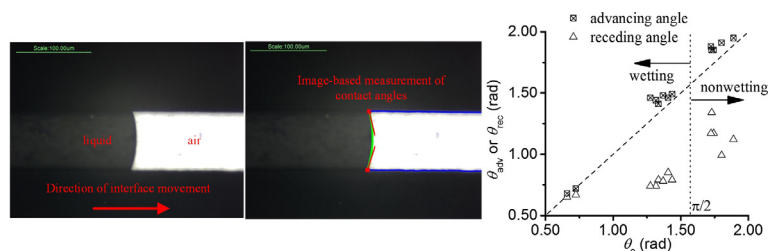


## Regular Article

## A two-angle model of dynamic wetting in microscale capillaries under low capillary numbers with experiments

Da Lei<sup>a</sup>, Mian Lin<sup>b,c,\*</sup>, Yun Li<sup>a,\*</sup>, Wenbin Jiang<sup>b</sup><sup>a</sup> School of Chemical Engineering and Technology, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China<sup>b</sup> Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, China<sup>c</sup> School of Engineering Science, University of Chinese Academy of Sciences, Beijing 100049, China

## GRAPHICAL ABSTRACT



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## ABSTRACT

**Objectives:** An accurate model of the dynamic contact angle  $\theta_d$  is critical for the calculation of capillary force in applications like enhanced oil recovery, where the capillary number  $Ca$  ranges from  $10^{-10}$  to  $10^{-5}$  and the Bond number  $Bo$  is less than  $10^{-4}$ . The rate-dependence of the dynamic contact angle under such conditions remains blurred, and is the main target of this study.

**Experiments:** Featuring with pressure control and interface tracking, the innovative experimental system presented in this work achieves the desired ranges of  $Ca$  and  $Bo$ , and enables the direct optical measurement of dynamic contact angles in capillaries as tiny as  $40 \times 20$  (width  $\times$  height)  $\mu\text{m}$  and  $80 \times 20 \mu\text{m}$ . The advancing and receding processes of wetting and nonwetting liquids were tested.

**Findings:** The dynamic contact angle was confirmed velocity-independent with  $10^{-9} < Ca < 10^{-5}$  (contact line velocity  $V = 0.135\text{--}490 \mu\text{m/s}$ ) and it can be described by a two-angle model with desirable accuracy. A modified two-angle model was developed and an empirical form was obtained from experiments. For different liquids contacting the same surface, the advancing angle  $\theta_{adv}$  approximately equals the static contact angle  $\theta_0$ . The receding angle  $\theta_{rec}$  was found to be a linear function of  $\theta_{adv}$ , in good agreement with our and other experiments from the literature.

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\* Corresponding authors at: Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190, China (M. Lin), School of Chemical Engineering and Technology, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China (Y. Li).

E-mail addresses: [linmian@imech.ac.cn](mailto:linmian@imech.ac.cn) (M. Lin), [yunli@mail.xjtu.edu.cn](mailto:yunli@mail.xjtu.edu.cn) (Y. Li).

## 1. Introduction

The liquid-gas displacement takes place in extracting oil from porous-type reservoirs by solution gas drive, water alternating gas injection or other enhanced oil recovery (EOR) schemes [1–4]. Understanding the mechanism of two-phase flow at pore scales is decisive for optimizing the EOR process, thus has attracted many theoretical and experimental researches during the past four dec-

ades [5–9]. For most pore-level displacements, the capillary number  $Ca$  ( $Ca = \mu V / \gamma$ , where  $\mu$  is the viscosity of the liquid,  $V$  the interface velocity and  $\gamma$  the interfacial tension) in oil reservoirs ranges from  $10^{-10}$  to  $10^{-5}$ , and the Bond number  $Bo$  ( $\rho g h^2 / \gamma$ , where  $\rho$  is the density of the liquid,  $g$  the gravitational constant and  $h$  the characteristic length of the flow) is less than  $10^{-4}$ , implying the capillarity dominates the movement of the liquid-gas interface [3,10]. This paper focuses on the wetting process in the mentioned scopes of  $Ca$  and  $Bo$ .

Precise prediction of capillarity-dominated multiphase flow in porous systems relies on an accurate model of the contact angle in dynamic conditions [11]. Possible models of the dynamic contact angle  $\theta_d$  can be categorized into two types, velocity-dependent and two-angle models, as depicted in Fig. 1. The hydrodynamic model (HD) and molecular kinetic theory (MKT) are typical velocity-dependent models.

### 1.1. Velocity-dependent models

A simplification of the HD for a liquid-gas system was made by Voinov, given by Eq. (1) [12,13]:

$$\begin{cases} \theta_d^3 = \theta_{adv,min}^3 + 9Ca \ln\left(\frac{L_H}{L_S,adv}\right) & Ca > 0 \\ \theta_d^3 = \theta_{rec,max}^3 + 9Ca \ln\left(\frac{L_H}{L_S,rec}\right) & Ca < 0 \end{cases} \quad (1)$$

where the sign of  $Ca$  represents the movement direction, positive  $Ca$  being advancing and negative  $Ca$  being receding.  $\theta_{adv,min}$  and  $\theta_{rec,max}$  are the minimum contact angle during advancing and the maximum contact angle during receding, respectively. A slip length  $L_S$  and a characteristic length  $L_H$  are introduced to characterize the three-phase contact zone, where the no-slip boundary condition fails [13,14].

Another model describing the effect of velocity is the MKT, which assumes that energy dissipates at the contact line where liquid, gas and solid meet. And it is given by Eq. (2) [15,16]:

$$\cos\theta_d = \frac{\cos\theta_{adv,max} + \cos\theta_{rec,min}}{2} - \frac{2k_B T}{\gamma \lambda^2} \operatorname{arsinh}\left(\frac{\gamma Ca}{2K_0 \lambda \mu}\right) \quad (2)$$

where  $T$  stands for the absolute temperature,  $k_B$  the Boltzmann constant,  $\lambda$  the average distance of molecular displacement and  $K_0$  the equilibrium frequency at which the molecules oscillates in the three-phase zone.  $\theta_{adv,max}$  and  $\theta_{rec,min}$  are respectively the maximum and minimum angles for advancing and receding.

### 1.2. Two-angle model

When the velocity-dependence is negligible, the two angle model assumes that the dynamic contact angle  $\theta_d$  can be repre-

sented by  $\theta_{adv}$  and  $\theta_{rec}$  respectively for advancing and receding, given by Eq. (3) [11],

$$\begin{cases} \theta_d = \theta_{adv} & Ca > 0 \\ \theta_d = \theta_{rec} & Ca < 0 \end{cases} \quad (3)$$

Whether or not the  $\theta_d$  is velocity-dependent remains unclear and none of the models have been confirmed to properly describe the dynamic contact angle with  $10^{-10} < Ca < 10^{-5}$  and  $Bo < 10^{-4}$ . Despite a great deal of experiments have been reported, some of them confirm the dependence of velocity [13,17–21], while another portion reports the opposite conclusion [22–25]. Table 1 gives a brief summary of dynamic wetting experiments. Among various methods, the plate immersion method, also known as Wilhelmy plate method, measures the force exerted on a plate when the plate is vertically immersed in the liquid, and the contact angle can be calculated from the force [22]. For filament immersion and sessile drop method, the contact angle can be directly observed and measured [13,24]. In capillary flow method, the contact angle can either be directly observed or calculated from pressure data. Capillary rise experiments, however, measure the height of wetting liquids surface during imbibition and calculate the contact angle through flow dynamics [26]. Moreover, the mentioned models have been used in comparison with various wetting dynamics experiments rather than prediction of wetting behaviors in specific systems [11]. The reason is that large discrepancies lie in the HD and MKT parameters found in the literature, and unreasonable values are frequently obtained [13,27]. Simple as it may be, the two-angle model is not a correlation with the static contact angle  $\theta_0$ , thus has no prediction capability either. Further development of the two-angle model is required. In our work, the static contact angle is measured by the sessile drop method with a droplet volume of 2–3  $\mu\text{L}$ , when the droplet is under static condition.

On one hand, the Bond number directly indicates the gravity effect. On the other hand, it implies the diameter of a capillary. To achieve  $Bo < 10^{-4}$ , the capillary diameter should be less than 0.03 mm (take water at room temperature as an example). Due to the limitation of observation methods, most capillary flow experiments were carried out in capillaries with a diameter near 1 mm [19,20,28]. Without direct measurement of the contact angle, capillary rise experiments can utilize capillaries with a diameter ranging from 0.6 to 0.2 mm, but the  $Ca$  cannot be controlled [23,26,29,30]. These shortcomings should be overcome to achieve expected ranges of  $Ca$  and  $Bo$ .

This paper aims to clarify: 1. Is the dynamic contact angle velocity-dependent under the condition of  $10^{-10} < Ca < 10^{-5}$  and  $Bo < 10^{-4}$ ? 2. What's the best model for the description of the dynamic contact angle under low capillary numbers.

In this work, a system was designed to enable the microscopic observation and tracking of a moving interface in a microscale capillary. Dynamic contact angles of a liquid/gas interface in channels as tiny as  $40 \times 20$  (width  $\times$  height)  $\mu\text{m}$  and  $80 \times 20 \mu\text{m}$  were measured by optical method for the first time. The advancing and receding processes of nonwetting and wetting liquids (deionized water, glycerol, etc.) were tested under  $10^{-10} \leq Ca < 10^{-3}$  and  $Bo < 10^{-4}$ . The system, experiment procedures and the error of measurement will be introduced in the first place. The dynamic contact angle was confirmed to be velocity-independent by experiments, and on this basis a modified two-angle model will be discussed which correlates the advancing and receding angles to the static angle. The model will be compared with our experiments along with others performed in different methods. The two-angle model could be a new solution to the calculation of capillary force in two-phase flow in porous media.

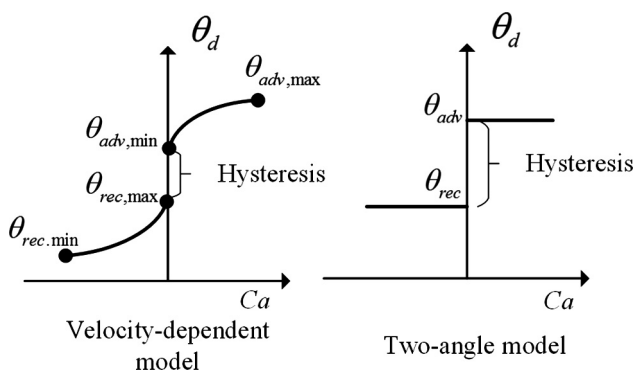


Fig. 1. Possible models of the dynamic contact angle.

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