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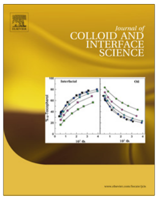

# Study of colloids transport during two-phase flow using a novel polydimethylsiloxane micro-model

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ABSTRACT

As a representation of a porous medium, a closed micro-fluidic device made of polydimethylsiloxane (PDMS), with uniform wettability and stable hydrophobic properties, was designed and fabricated. A flow network, with a mean pore size of 30 μm, was formed in a PDMS slab, covering an area of 1 mm × 10 mm. The PDMS slab was covered and bonded with a 120-μm-thick glass plate to seal the model. The glass plate was first spin-coated with a thin layer, roughly 10 μm, of PDMS. The micro-model was treated with silane in order to make it uniformly and stably hydrophobic. Fluorescent particles of 300 μm in diameter were used as colloids.

It is known that more removal of colloids occurs under unsaturated conditions, compared to saturated flow in soil. At the same time, the change of saturation has been observed to cause remobilization of attached colloids. The mechanisms for these phenomena are not well understood. This is the first time that a closed micro-model, made of PDMS with uniform and stable wettability, has been used in combination with confocal microscopy to study colloid transport under transient two-phase flow conditions. With confocal microscopy, the movement of fluorescent particles and flow of two liquids within the pores can be studied. One can focus at different depths within the pores and thus determine where the particles exactly are. Thus, remobilization of attached colloids by moving fluid–fluid interfaces was visualized. In order to allow for the deposition and subsequent remobilization of colloids during two-phase flow, three micro-channels for the injection of liquids with and without colloids were constructed. An outlet channel was designed where effluent concentration breakthrough curves can be quantified by measuring the fluorescence intensity. A peak concentration also indicated in the breakthrough curve with the drainage event. The acquired images and breakthrough curve successfully confirmed the utility of the combination of such a PDMS micro-model and confocal microscopy for the visualization of colloid transport in a flow network filled with two fluids, and in particular, the colloids retention, mobilization, and transport under transient flow conditions.

## 1. Introduction

As a representation of a porous medium, micro-models are commonly employed to study and visualize physical, chemical, and biological processes at the pore scale. During the last few decades, micro-models have proven to be a valuable tool for the study and observation of flow of fluids and transport of solutes within the pore space. They have been increasingly used to study diverse applications, such as energy-related multiphase transport in porous media [1], reservoir engineering [2], and two-phase flow experiments [3–11]. An extensive review of micro-models and their use in two-phase flow research can be found in Karadimitriou and Hassanizadeh [12], including fabrication methods, materials used, and visualization techniques.

The use of micro-chips in colloids transport is a relatively recent development. So far, mostly, an open micro-channel combined with microscopy has been used to study colloids transport [13–17], where visualization is relatively simple. But, micro-channels are too simple and in particular, various two-phase phenomena occurring in a porous medium will not occur there. One needs to have a network containing a large number of pores. Pioneering visualization experiments on colloid retention in unsaturated media were performed by Wan and Wilson [18,19] in glass micro-models under optical microscopy with fluorescent lighting system. In their system, the air phase was stagnant and water phase flowed

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*Abbreviations:* PDMS, polydimethylsiloxane; FWI, fluorinert–water interface; SWI, PDMS–water interface; FWSC, fluorinert–water–solid contact line.

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at steady state. Later on, etched glass micro-models were used by other research groups [20,21] to study colloids movement at pore scale with optical microscopy. They injected air bubbles into micro-models and visualized the interaction of colloids and a single air bubble under steady-state. Other materials have been also used to make micro-models for colloids transport. For example, silicon micro-models have been used to study colloids hydrodynamic behaviors in a saturated medium [22]. PDMS micro-fluidics has been used in biochemical engineering applications [23], blood micro-particles distribution [24], and multiphase/two phase flow in porous media. Compared to glass and silicon, a soft material like PDMS is more suitable for making inexpensive micro-models by rapid prototyping [25].

One of the open questions in the transport of colloids in unsaturated porous media and/or in two-phase flow is the role of fluid–fluid interfaces, as well as fluids–solid contact line, in the attachment and remobilization of colloids. Optical microscopy provides a lumped image of the whole channel depth. But, in order to investigate the interactions between colloids and fluid–fluid interfaces and/or contact lines in a two-fluid system, one needs to focus and get images at various depths within the pore space. This can be achieved with a confocal microscope. Confocal microscopy is a point-by-point visualization method. One of the fluids should either be dyed with fluorescent dyes or contain some fluorescent particles. With confocal microscopy, the sample can be tracked spatially in three dimensions by superposing two dimensional images taken at sequential z stacks [26] (z being the coordinate in the depth). Furthermore, it allows real-time information about the complex mechanisms under dynamic conditions. However, confocal microscopes have a limited depth of view. Generally, they give good results for a depth of view up to 250 μm [27]. That is the reason that mostly open micro-channels have been used in studies involving confocal microscopy; a cover plate takes up much of the depth of view and thus a very limited depth of the channel can be visualized.

In this work, we describe the design and fabrication of a closed PDMS micro-model, following a procedure described by Xia and Whitesides [28]. PDMS micro-models are very cheap to produce, and can be made under normal laboratory conditions (so, for example, a clean room is not needed). Moreover, the PDMS micro-model that we designed and manufactured had some important properties that are usually lacking in other PDMS models. First, the micro-model was made uniformly and stably hydrophobic. Second, the model was sealed with a very thin glass plate, coated with a film of PDMS. This made it possible to focus at locations throughout the whole depth of the model (30 μm). Third, the system of injection of fluids, with and without colloids, was constructed such as to avoid mixing of fluid phases at the entrance. Fourth, the inner surfaces of the pores were all PDMS. This made sure that the fluids would experience the same wetting properties everywhere; there was no mixed wettability.

To the best of our knowledge, this is the first application of PDMS micro-model combined with confocal laser scan microscopy to study the movement of fluorescent particles in a flow network. It is also the first study of colloids transport in a porous network with transient flow two immiscible liquids. Acquired images were used to analyze colloids interaction with liquid phases, water–fluorinert and liquid–solid interfaces, as well as with the fluids–solid contact lines.

## 2. Experimental setup, materials, and methods

### 2.1. Liquids and particles

As mentioned above, we were interested in generic studies of the fate of adsorbing colloidal particles under transient two-phase flow conditions. So, it is not crucial which phase is the wetting phase and which one is the non-wetting. As the two immiscible fluids, we selected deionized water and fluorinert FC-43(3M). Given the fact that our PDMS micro-model was hydrophobic, water was the non-wetting phase and fluorinert was the wetting phase. The fluorinert liquid type was chosen such that it had nearly the same index of refraction as water (1.291 and 1.332, respectively, at 20 °C). Carboxylated fluorescent microspheres (Polysciences Inc. GmbH), with an average diameter of 300 nm were used as model colloids. They were hydrophilic and weakly negatively charged at neutral pH and were labeled with fluorescein. Because we worked with large concentration of colloids (up to $10^{12}$ particles per liter), we had to make sure no coagulation in the colloids suspension occurred. Therefore, before injection, the colloids-water suspension was immersed in an ultrasound bath for 30 min to prevent coagulation. We were interested in colloid transport and mobilization under transient hydraulic conditions.

### 2.2. The flow network

A two-dimensional pore network was designed based on Delaunay triangulation. Delaunay triangulation is considered to be a good representation of a natural porous medium [29]. The network comprised an assembly of pore bodies (large pores) connected to each other by smaller pores, called pore throats. In Delaunay triangulation, points are connected to their neighbors by non-intersecting bonds. Connected points form triangles that are as equilateral as possible. The coordinates of the triangulation points were generated in MatLab. These points were considered to be the centers of the pore bodies. The network was then exported to AutoCAD sketch for further processing. An outlet channel, along with three inlet channels for the introduction of the fluids and the colloids, were added to the same sketch and connected to the network.

As can be seen in Fig. 1, the final design of the micro-model had five parts: (1) three inlet reservoirs (each 0.5 mL in volume); (2) three inlet channels (6 mm in length, 0.5 mm in width and 30 μm in depth) for introducing fluorinert, water, and water with colloid suspensions; (3) the flow network; (4) the outlet channel; (5) the outlet reservoir. The overall network dimensions were 1 mm by 10 mm, with 90 pore bodies and roughly 200 pore throats. The mean diameter of pore bodies was 30 μm. This was also the depth of all parts of the network, which was measured and found to be constant within a margin of 0.5% throughout the micro-model. The width of pore throats was between 25 μm and 30 μm. The outlet channel, as well as the outlet reservoir had some pillars (white strips) added for structural strength. Given the aspect ratio between the width and the depth of the channel and the reservoir, the top surface would collapse without the pillars [30].

### 2.3. Fabrication of the micro-model

Here, the fabrication of the micro-model is briefly explained. A detailed description of the procedure is given by Karadimitriou

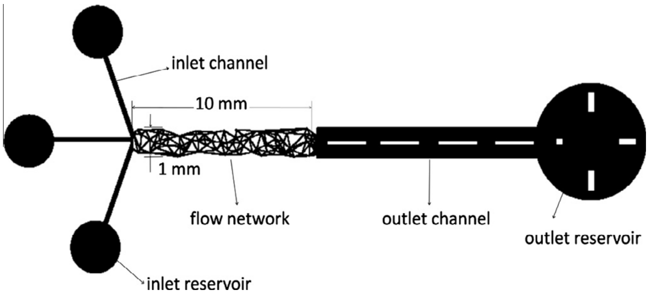

**Fig. 1.** Schematic representation of the micro-model (not to scale).