Microporous and Mesoporous Materials 178 (2013) 64–68

Contents lists available at SciVerse ScienceDirect

Microporous and Mesoporous Materials

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

Magnetic resonance imaging of reaction-driven viscous fingering in a packed bed

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ARTICLE INFO

Article history:
Available online 14 March 2013

Keywords:
Magnetic resonance imaging
MRI
Viscous fingering
Packed bed
Reactive flow

ABSTRACT

The formation of 3-dimensional viscous fingers has been investigated in a packed bed using magnetic resonance imaging. Fingering patterns are produced as a result of the formation of a highly viscous wormlike micelle solution formed at the interface between solutions of cetyltrimethylammonium bromide (CTAB) and sodium salicylate (NaSal), which have similar viscosities, but that are significantly lower to the wormlike micelle solution. In this system, fingers are driven by an interfacial instability, rather than a Saffman–Taylor instability. The structure of the fingering patterns, which were found to be sensitive to flow rate, where analysed using histogram plots characterizing the distribution of pores containing the CTAB solution. This system shares similar characteristics to the growth of biofilms.

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1. Introduction

Viscous fingering, a hydrodynamic instability also commonly known as the Saffman–Taylor instability [1], forms when a more viscous fluid is displaced by a less viscous fluid in a porous medium. This phenomenon is observed across a variety of disciplines including oil recovery [2], hydrology [3,4] and separation science [5,6]. Of the many experimental [2,6–8] and theoretical [3,9] investigations of this phenomenon, most have been focussed on non-reactive systems. However, in recent years, there has been increasing interest concerning the role reaction plays on the formation and development of these hydrodynamic instabilities [10–15]. It has been found that reaction can destabilise the otherwise stable displacement of a less viscous fluid by a more viscous fluid [16]. It has also been observed that reaction can produce fingering when a rheological change occurs at the interface between two fluids of identical viscosity [15]. In this later situation, fingering has been produced when solutions of the cationic surfactant cetyltrimethylammonium bromide (CTAB) and the organic salt sodium salicylate (NaSal) were brought into contact, producing highly viscoelastic wormlike micelles at the interface. These ordinarily miscible, aqueous surfactant and co-surfactant solutions are of the same viscosity, and so should not normally produce viscous fingers. However, when mixed, CTAB and NaSal, which acts as a co-surfactant, organise to form long wormlike micelles, which become entangled and dramatically increase the viscosity of the solution. The interface between the two solutions thickens over time, as surfactant and co-surfactant molecules diffuse and aggregate. As only the region containing wormlike micelles has increased viscosity, the phenomenon is an interfacial instability and occurs irrespective of which fluid displaces which. Thus it is not a Saffman–Taylor instability. The fingering produced in this system, which shares many of the characteristics of a biofilm [15], has been investigated as a function of solution concentration, flow rate and which solution is injected or displaced [15]. A variety of fingering patterns have been observed, and characterized by their shape, such as tentacle, fan and mushroom fingers. The fingers develop from an initially stable interface into these structures as a function of time and flow rate. Belmonte and co-workers [15] only found slight differences in the patterns depending on which solution was displaced by which, and explained this by differences in the diffusivity between the different molecules and micelles [15].

Much of the experimental research investigating fingering instabilities has employed Hele–Shaw cells, which are composed of parallel transparent glass plates, separated by a narrow gap (typically <1 mm) [2]. The cell is filled with one fluid, with the second injected into the first. Fluids are frequently distinguished visually with the aid of a dye. The advantages with Hele–Shaw cells are that they are simple to set up and fluids can be easily observed. The disadvantages with this system are that it is only, at best, a 2-dimensional system and a rather crude model for a real porous medium. To date only experiments of fingering in reactive systems have been performed in Hele–Shaw cells. However, with the use of magnetic resonance imaging (MRI) it is possible to investigate these systems in a 3-dimensional porous medium. This technique has been successfully employed by Fernandez and co-workers to investigate non-reactive viscous fingering in chromatography columns [5,17]. In these experiments a glycerol solution was displaced by saline solution and the fingers were observed by doping the glycerol solution with Gd^{3+} and using T_2 relaxation time contrast to observe only the saline solution phase.

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1387-1811/$ - see front matter © 2013 Elsevier Inc. All rights reserved.
http://dx.doi.org/10.1016/j.micromeso.2013.02.039

In this paper, we report the first experimental investigation of fingering instabilities in a reactive system in a 3-dimensional (3D) porous medium. Fingers were visualised in packed beds using MRI, exploiting the inherent variation in T_2 magnetic resonance relaxation time of water in the different solutions as the means of producing image contrast. One challenge with studying these fingering patterns in a 3D packed bed is associated with how these structures are characterised. Hele–Shaw cells enable visual description of fingers, as patterns are formed in a plane. In a 3D porous medium, the tortuosity of the pore space allows fingers to develop with greater freedom to meander. We, therefore, applied an alternative method for characterizing fingering patterns in packed beds, using statistical analysis of the imaging data.

2. Experimental

2.1. Materials

Cetyltrimethylammonium bromide (CTAB) (Sigma Aldrich, ⩾98%) and sodium salicylate (NaSal) (Sigma Aldrich, ⩾99.5%) were used without further purification. Solutions of 50 mM CTAB and 100 mM NaSal were prepared using distilled water. These solutions were used to fill a packed bed comprising a cylindrical glass tube of length 30 cm and internal diameter 11 mm, packed with 1 mm diameter borosilicate glass beads (Sigma). The packed bed was held vertically in a 7T magnet with the NaSal solution above the CTAB solution and the interface between the two solutions positioned at the bottom of the homogeneous region of a 25 mm birdcage radiofrequency resonator. The CTAB solution was pumped from below using a syringe pump (Harvard pump 22) at flow rates of 1–4 ml h^{-1}.

2.2. Magnetic resonance experiments

All magnetic resonance (MR) experiments were performed using a Bruker DMX 300 spectrometer, operating at a ^1H resonance frequency of 300.13 MHz, at a temperature of 294.0 ± 0.3 K. Relaxation experiments [18] were performed to measure the spin-lattice, T_1, and spin–spin, T_2, MR relaxation times of the 50 mM CTAB, 100 mM NaSal and 50 mM/100 mM CTAB/NaSal wormlike micelle solutions. Inversion-recovery (IR) experiments were performed to measure the T_1 times for solutions, using 16 experiments with IR delays logarithmically spaced between 5 μs and 15 s. The T_1 relaxation times for the solutions were 2.3 ± 0.1 s (50 mM CTAB), 2.4 ± 0.1 s (100 mM NaSal) and 2.3 ± 0.1 s (50 mM/100 mM CTAB/NaSal). Carr–Purcell–Meiboom–Gill (CPMG) experiments were performed to measure the T_2 for the solutions, acquiring a maximum of 512 echoes with an echo spacing of 20 ms. The T_2 relaxation times for the solutions were 1660 ± 10 ms (50 mM CTAB), 240 ± 12 ms (100 mM NaSal) and 664 ± 8 ms (50 mM/100 mM CTAB/NaSal).

Three-dimensional images were acquired using the fast spin-echo imaging technique RARE [19], with a field of view of 40 mm × 20 mm × 12 mm and a pixel array of 256 (z) × 128 (x) × 16 (y), respectively. A RARE factor of 128 was used, giving an effective echo time of T_{eff} = 575 ms resulting in high signal intensity (SI) for the water in the CTAB solution and low SI for the NaSal and micelle solutions. The repetition time was T_R = 1 s, with a time resolution between images of 120 or 60 s for flow rates of 1 ml h^{-1} or 4 ml h^{-1}, respectively. The orientation of images with respect to the tube and magnet is shown in Fig. 1.

2.3. Image analysis

All MR data was analysed using Prospa [20]. Images were Fourier transformed and compressed from 256 pixels to 32 along the z direction, so that each pixel shown was integrated over 8 pixels along this direction. The displacement of the CTAB and wormlike micelle solutions into the NaSal solution was detected by monitoring the position of pixels above a threshold signal intensity (SI), determined by the average of the SI of the CTAB (high SI) and NaSal (low SI) solutions. As the relaxation time for the wormlike micelle solution was in between that of the CTAB and NaSal solutions, it was, in principle, possible to distinguish between all three regions. However, as the T_{eff} was comparable with the relaxation time of the wormlike micelle solution, it was not possible to distinguish between the wormlike micelle and NaSal solutions in the packed bed, hence, the NaSal and micelle solutions are considered together. For each uncompressed 3D data set, the distribution of the high SI fluid was characterized using three histogram plots, giving the number of pixels, N(**r**), above the SI threshold, at a distance r along the x, y and z *axes* from a root mean square position for the high SI fluid. The root mean square position for pixels filled with the high SI fluid, which defines their centre, was determined using a plug-flow model for the filling of the packed bed. In the plug-flow model, the distribution of high SI fluid was allowed to develop as the packed bed filled, as a plug, at a rate dependent on the volume flow rate and porosity of the packed bed (determined from a spin density image of the packed bed).

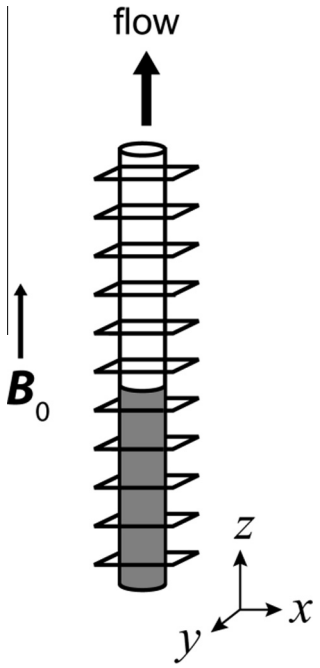

Fig. 1. Schematic diagram showing the orientation of images with respect to the tube, magnet and flow direction. The 50 mM CTAB solution (grey) is positioned at the bottom of the tube and the 100 mM sodium salicylate solution (white) above.

The histogram plots for each image were then compared with those associated with four simple models, where a spin-density image of the packed bed was allowed to fill with high SI fluid as a plug, single central-finger, single side-finger or pair of side-fingers. Each model system was created by selecting pores in certain regions to be filled with the displacing fluid. The distribution of the filled pores were restricted to one of the four shapes used – plug, side finger, central finger or double finger. The cross-sectional area of each finger was 33% of the width of the tube. The length of the finger in each model system was allowed to increase as a function of time and flow rate and was controlled by the volume flow rate and the pore space volume found in the experimental packed bed. By using these factors and the porosity of the packed bed, it was possible to determine the number and distribution of the

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