Creation of a dual-porosity and dual-depth micromodel for the study of multiphase flow in complex porous media†‡

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Silicon-based microfluidic devices, so-called micromodels in this application, are particularly useful laboratory tools for the direct visualization of fluid flow revealing pore-scale mechanisms controlling flow and transport phenomena in natural porous media. Current microfluidic devices with uniform etched depths, however, are limited when representing complex geometries such as the multiple-scale pore sizes common in carbonate rocks. In this study, we successfully developed optimized sequential photolithography to etch micropores (1.5 to 21 μm width) less deeply than the depth of wider macropores (>21 μm width) to improve the structural realism of an existing single-depth micromodel with a carbonate-derived pore structure. Surface profilimetry illustrates the configuration of the dual-depth dual-porosity micromodel and is used to estimate the corresponding pore volume change for the dual-depth micromodel compared to the equivalent uniform- or single-depth model. The flow characteristics of the dual-depth dual-porosity micromodel were characterized using micro-particle image velocimetry (μ-PIV), relative permeability measurements, and pore-scale observations during imbibition and drainage processes. The μ-PIV technique provides insights into the fluid dynamics within microfluidic channels and relevant fluid velocities controlled predominantly by changes in etching depth. In addition, the reduction of end-point relative permeability for both oil and water in the new dual-depth dual-porosity micromodel compared to the equivalent single-depth micromodel implies more realistic capillary forces occurring in the new dual-depth micromodel. Throughout the imbibition and drainage experiments, the flow behaviors of single- and dual-depth micromodels are further differentiated using direct visualization of the trapped non-wetting phase and the preferential mobilization of the wetting phase in the dual-depth micromodel. The visual observations agree with the relative permeability results. These findings indicate that dual-porosity and dual-depth micromodels have enhanced physical realism that is pertinent to oil recovery processes in complex porous media.

1. Introduction

Carbonate reservoirs hold substantial petroleum resources; yet, understanding the flow dynamics in these reservoirs is challenging due to their propensity toward multi-scale heterogeneity. For instance, the Ghawar Field of Saudi Arabia is the world’s largest oil field in terms of production (5.8 million barrels of oil per day) and total remaining proven oil reserves of 75 billion barrels as of January 1, 2014. Production in the Ghawar Field as well as other Saudi Arabian fields is predominantly from carbonate reservoir rocks including the prolific Upper Jurassic Arab Formation. Characterizing carbonate reservoirs is often difficult due to multi-scale variability in pore structure and physical properties such as porosity and permeability. The heterogeneity of the pore structure as well as mixed surface wettability, typical of many carbonate reservoirs, results in significant residual oil saturation compared to most siliciclastic reservoirs. This phenomenon has been well documented.2,3

Microporosity, as well as large aspect ratios (that is, the ratio of pore-body size to throat size), contributes to the large residual oil saturations in carbonates.2,4 Cantrell and Hagerty (1999) report that microporosity (pores 10 μm or less in diameter for their study) ranges from 0 to 100% and typically comprise 25% to 50% of the total porosity in the most productive reservoir intervals of the Arab Formation.4 Alternatively, large aspect ratios (values >3) inhibit flow and contribute to snap-off in which fluid phases become discontinuous or isolated.5,6

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2 Time-resolved digital particle image velocimetry tool for MATLAB (version: 1.4) is an open source software and available from https://dx.doi.org/10.6084/m9.figshare.1092508.v6
3 Electronic supplementary information (ESI) available. See DOI: 10.1039/c6lc01343k
The abundance of microporosity and the complexity of the pore structure coupled with large aspect ratios provide an ongoing challenge to understanding the distribution of fluids and the fluid flow behavior in carbonate reservoirs. This knowledge enables optimal field development and recovery efficiencies. In light of this, a focused approach is needed to better understand the heterogeneous nature of carbonates containing multiple fluid phases and the flow properties within these porus formations. This involves detailed characterization (including spatial distribution) of the pore structure (pore-body and throat sizes, surface roughness, and connectivity), fluids (properties and saturations), permeability (absolute and relative), and capillarity.

2. Micromodels

Micromodels are 2D representations of porous media that allow direct visualization of fluid flow behavior at the pore scale. Pore-scale observations made using silicon-based micromodels have been widely used for understanding multiphase flow and oil–water–solid interaction in enhanced oil recovery (EOR) studies. Micromodels have been used as a tool for the visualization of behaviors such as capillary trapping/fingering, observation of micro-emulsion phases, pore-level hydrate formation, foam coalescence, and fluid analysis such as the measurement of minimum miscibility pressure of CO₂ in crude oils.

Rock wettability is a key determining factor in the recoverability of oil from the subsurface. Silicon-based photolithography creates strongly water-wet surfaces representative of a clean, unaltered sandstone rock surface. Reservoir wettability converts from strongly water-wet toward either oil-wet or mixed-wettability due to the interaction between mineral surfaces, connate water, and specific components in the hydrocarbons. To be able to replicate realistic surface interactions between reservoir fluids and reservoir rocks, previous studies presented methods to modify the strongly water-wet surfaces in silicon-based micromodels. Alternatively, a simple method has been introduced to fabricate synthetic CaCO₃ reservoir micromodels that replicate the surface geochemistry of carbonate in microfluidic channels with relative simple and homogenously aligned posts.

In order to represent geometrically the complex pore structure of the Arab-D carbonate reservoir in a micromodel, Buchgraber et al. (2012) used silicon-based photolithography with a mask derived from images of a reservoir sample to create a dual-porosity micromodel with a carbonate pore network pattern and a single etch depth. The dual-porosity micromodel simulates typical Arab-D pore-size heterogeneity including both macro-pores (>21 μm) and microporosity (1.5 to ≤21 μm). Using the realistic dual-porosity micromodel, the researchers observed pore-level quantification of multiphase flow and provided pore-scale quantification of fluid phase dynamics by measuring porosity, permeability, fluid desaturation patterns, and recovery factors.

During two-phase flow, the capillary entry pressure ($ΔP_{c,e}$) must be overcome for the non-wetting fluid (oil) to invade wetting-phase-filled pore throats as shown in Fig. 1. The required capillary entry pressure ($ΔP_{c,e}$) for a circular cross section is written as:

$$ΔP_{c,e} = 2σ_{ow} \cos θ \left( \frac{1}{R_{th}} \right)$$

where $R_{th}$ is pore throat diameter, and $σ_{ow}$ is the interfacial tension (IFT) between wetting and non-wetting fluids. Thus, the non-wetting fluid intrudes preferentially into pore bodies accessed by larger pore-throat diameters unless the driving force ($ΔP_{c,e}$) is sufficient enough to penetrate smaller wetting-phase-filled pore throats.

Single-depth microfluidics devices use microchannels where variability in pore size is created by changes in channel width. As presented by Wan et al. (1996), the etching depth ($H$) is constant regardless of the pore-size transition and may cause either over- or underestimation of capillarity effects. To overcome this limitation in micromodels, Trygstad et al. (1986) bonded two etched glass plates with all of the porosity etched onto one surface and only the wider pores onto the opposing surface to fabricate a glass micromodel representing microscopic rock pore heterogeneity. Wan et al. (1996) attempted to vary both width and etching height using a sequential-step, hydrofluoric (HF) acid etching technique for relatively simple fractures and a homogeneously patterned matrix structure. More recently, Xu et al. (2017) demonstrated a standard single-step lithography process using hydrofluoric (HF) acid to etch multilayered glass microchips with a homogeneous pattern.

As yet, there has been no complete and rigorous silicon-based micromodel fabrication technique reported for a complex dual-porosity pore network system that mitigates the capillarity limitation arising from single- or uniform-etching depths; hence, the contrast in capillary characteristics between macro- and microporosity of dual-porosity pore networks have not been fully realized within a micromodel.

![Fig. 1 Schematic diagram of oil-phase invasion into water-filled pore throats. (a) Oil ganglia remain trapped when unable to surmount capillary entry pressure exerted by the pore throat size ($R_{th}$). (b) The driving force (left to right) overcomes capillary entry pressure and non-wetting phase (oil) flows into the water-filled pore throat. Adapted from Rossen.](image-url)
Evidently, fluid flow behavior is more realistically represented using contrasting pore sizes where certain 3D characteristics, namely wider pores, are etched more deeply than smaller pores. Accordingly, we developed an optimized sequential-etching approach to add 3D aspects to a 2D dual-porosity micromodel. Multiple etch depths corresponding with macro- and micro-pore sizes are integral to the approach. Characterization methods include pore-scale observation, permeability measurements, and real-time velocity profiles within the multi-depth dual-porosity micromodel. This newly developed microfluidic device further improves the study of pore-scale single- and two-phase fluid dynamics as well as fluid desaturation.

3. Methodology

This section discusses the detailed procedure to prepare two masks based on the pore structures of real reservoir rocks and their usage in the sequential micromodel fabrication process. Furthermore, micromodel characterization methods including μ-PIV, absolute and relative permeability measurements, and fluid saturation calculation are demonstrated.

3.1 Micromodel fabrication: complete- and partial-pore masks

Mask preparation for the dual-depth dual-porosity micromodel requires first constructing a mosaic consisting of overlapping high-resolution images from a polished epoxy-impregnated thin section of reservoir rock as described in Buchgraber et al. (2012). Images were collected with a pixel resolution of 0.235 μm using a JEOL JSM-5600LV scanning electron microscope (SEM) in back-scattered mode (BSE). The grayscale mosaic compiled from these images was then segmented into pore and non-pore pixels. The resulting binary image was modified to fill pores with incomplete epoxy impregnation, reconnect pores in the 2D mosaic using throat sizes measured via mercury injection (Fig. 2), and alter the edges of the mosaic to ensure seamless connectivity between repeated base images in the 3-by-3 arrayed mask in Fig. 3. The resulting base image (Fig. 2) has a porosity (\( \phi_{H} \)) of 45.3% area. Due to hardware limitations, the base image was printed on the mask with a pixel size of 1.5 μm. This upscaling preserves the heterogeneity and relative pore sizes compared to the alternative of resampling the base image. The throat size distribution shown in Fig. 2 was rescaled to match. Distribution channels, injection ports, and registration marks were added (Fig. 3). The resulting base image and mask is called “complete-pore”, because the mask contains both micro- and macro-sized pore structures. This mask is used for the shallower etch depth.

A second mask for the deeper etch depth was then prepared by subjecting the “complete-pore” base image (Fig. 2) to seven cycles of erosion-dilation (E-D; 2-D Petrographic Image Analysis System v. 7.0). E-D strips off the outermost layer of pixels (erosion) and then adds a layer of pixels to the remaining surface (dilation). This process is repeated for two layers, three layers, and so forth until seven layers have been eroded and dilated. This process removes pores, throats, and surface roughness features that are, in this particular case, less than or equal to 21 μm in diameter. This resulting base image has a porosity (\( \phi_{L} \)) of 34.1% area and is denoted as the “partial-pore” image, because it contains only macro-porosity, that is pores and throats wider than 21 μm. Changes in the pore structure are observed in close-up examples of the original and eroded base images (Fig. 2). The difference in porosity between “complete-pore” and “partial-pore” base images is 11.2% and is hereafter referred to as microporosity (\( \phi_{\text{micro}} \)). Similar to the “complete-pore” mask, the “partial-pore” base image is arrayed as shown in Fig. 3. Registration marks identical to those on the “complete-pore” mask were added for alignment. Injection ports and distribution channels were not included in the “partial-pore” mask.
μ spin-coated with 1.6 tween the wafer and photoresist. The primed wafer was then methyldisilazane) allowing better coverage and adhesion between the wafer and photoresist. The wafer was then dehydrated at 150 °C for 30 minutes using HMDS (hexamethyldisilazane) for 50 s and then washing with deionized water was repeated across the micromodel was monitored using pressure transducer (C), and the back pressure regulator (E) minimizes possible pressure drop fluctuation. A syringe pump drives solutions into the micromodel through the injection port (I-A). A flow distribution fracture or channel along the matrix connects to the other inlet port (I-B); a similar distribution channel connects the production ports (I-C and I-D). In the matrix, the rock-based pore network was repeated in 3 by 3 array. Matrix size (grey area) is 6 by 4.5 cm.

3.2 Micromodel: sequential photolithography

Once the “complete-pore” and “partial-pore” masks were prepared, the masks were then used for sequential (two-step) photolithography. A detailed two-step fabrication procedure was developed to generate precision dual-depth micromodels beginning with “partial-pore” single-depth etching and subsequent “complete-pore” dual-depth etching as demonstrated in Fig. 4. During the “partial-pore” first- or single-etch depth process, the fresh 4 inch silicon wafers were primed and dehydrated at 150 °C for 30 minutes using HMDS (hexamethyldisilazane) allowing better coverage and adhesion between the wafer and photoresist. The primed wafer was then spin-coated with 1.6 μm thick photoresist using a SVG (Silicon Valley Group) spin coater with an automated track system for dispensing photoresist onto the silicon wafers. The uniformity of spun photoresists is typically ±100 Å using the SVG spin coater on the flat surface of a fresh silicon wafer. After this coating process depicted in Fig. 4a, the wafers were exposed to ultraviolet light through the “partial-pore” carbonate mask containing only the larger macro-pore network (Fig. 4b) using Karl Suss MA-6 Contact Aligner system.

After exposure, the wafers were processed through an automated developing track system (SVG Developer) for developing and post-baking the exposed photoresist-coated wafers. In order to finalize the pore network on the developed wafers, wafers were etched to about 7 μm using an inductively charged plasma deep reactive ion etcher with 1.8–4 μm min⁻¹ etching rate (Fig. 4c). The remaining photoresist on the wafer was then removed by soaking the wafers in a chemical bath of “piranha” (90% sulfuric acid/hydrogen peroxide) for 20 minutes (Fig. 4d).

For the “complete-pore” etching process, the etched wafer from the “partial-pore” or single-etch depth process was coated with photoresist using an EVG101 Spray coater (Fig. 4e). Spray coating was used to cover the uneven, previously etched wafer surfaces produced during the “partial-pore” procedure. Spray coating distributes photoresist more evenly on the arbitrarily shaped and textured substrates whereas spin coating proved inadequate with respect to film thickness homogeneity and edge coverage.

Photoresist for spray coating was prepared by mixing conventional positive photoresist SPR 220-7 (Rohm and Haas Electronic Materials LLC, Marlborough, MA) with solvents such as MEK (methyl-ethyl ketone) and PGMEA (metoxy-propyl acetate) to adjust the evaporation rate and the solid content of the photoresist.¹ The photoresist mixture consists of photoresist and solvents combined in various proportions such as 6.5 wt% SPR220-7, 68 wt% MEK, and 25.5 wt% PGMEA. A 1.6 μm thick layer of photoresist was spray-coated on the etched silicon wafer using six passes at 80 °C followed by post baking at 90 °C for 200 s. Once the photoresist mixture was uniformly coated on the wafer by the spray-coating technique and the “complete-pore” mask is aligned with previous pattern, the photoresist-coated wafer was exposed to ultraviolet radiation for six seconds at vacuum contact mode by following the exposure protocol: 3 s exposure followed by 10 s pause and another 3 s exposure. After the 6 s exposure process, a developing process of immersing the exposed wafer in conventional MF26A developer (mixture of >95% water, 2.3% tetramethylammonium hydroxide, and <1% polyglycol; Rohm and Haas Electronic Materials LLC, Marlborough, MA) for 50 s and then washing with deionized water was repeated three times (see ES†).

The “complete-pore” overlay mask pattern alignment during the exposure step (Fig. 4f) is critical to expose the previously etched area in Fig. 4c. Once the “complete-pore” dual-depth wafer shown in Fig. 4g was etched up to H₁, 1 mm diameter holes were drilled at the production and injection ports (I-A, I-B, I-C, and I-D in Fig. 3). After the ports were drilled, the wafers went through a thorough cleaning process (Fig. 4h) to remove any remaining photoresist and
precipitants that might block the microchannels (pore bodies and throats) on the wafers. Finally, a Schott Borofloat 33 glass wafer (S.I. Howard Glass, Worcester, MA) was anodically bonded to the top of the clean, twice-etched wafers at 1200 volts after a thermal oxidation process at 300 °C for 45 minutes to produce a water-wet silicon dioxide (SiO₂) surface on the silicon wafer.

3.3 Micro-particle image velocimetry (μ-PIV)

Particle Image Velocimetry (PIV) is a technique widely used in hydrodynamics. The fluid is seeded with tracer particles that are assumed to follow faithfully the flow. PIV results in the accurate quantitative measurement of fluid velocity vectors. In μ-PIV, the ability of the objective lens of the microscope to focus only on one plane at a time is used.32 The application of μ-PIV techniques for the study of flow in porous media has been reported previously.33–35 Flows may be steady or transient.35 More importantly, for more complex and realistic pore geometries, validation exercises of μ-PIV techniques are necessary. Roman et al. (2015) have successfully applied the technique to the analysis of realistic sandstone geometries where converging and diverging pore structures exist.35

Our setup to perform μ-PIV is the same as that in Roman et al. (2015). Our optical velocimetry technique uses conventional microscopy and digital imaging methods for the quantitative determination of two-component velocity data. In this work, μ-PIV was used to observe the fluid velocity field within the dual-porosity micromodels under steady-state flow conditions. Movies (640 by 480 pixels) recorded the movement of particles as the water phase flowed through the water-saturated pore space of both single- and dual-depth dual porosity micromodels. The particles in the water phase are carboxylate modified latex (CML) micro-particles (polybead carboxylate microsphere 1 μm diameter, Polysciences) that have a hydrophilic surface due to carboxyl functional groups and a negative charge at pH values greater than 5. The density (1.05 g ml⁻¹) and hydrophilic properties of the particles minimizes sedimentation and adsorption on the micromodel walls that are water-wet. The particle seeding concentration in the water phase is set at approximately 0.06% by volume.

The principle of PIV is to record two images of the flow of particles separated by a short time interval. Images are subdivided into many small interrogation windows. The displacements of interrogation windows between the two images are determined through spatial cross-correlation. Velocity is merely found by dividing the particle displacements by the time between images. The quality of μ-PIV measurements is closely connected to the implementation, post-experiment processing, and data analysis. As in Roman et al. (2015),35 image processing was conducted in order to obtain a sequence of images that contains information related to the moving particles only. This final image sequence was used for μ-PIV analysis. An example is shown in Fig. 5. Images extracted from the video file (Fig. 5a) were processed using noise filtration, grayscale image transformation, and correction of light intensity fluctuations (Fig. 5b). Each pixel of every three images was averaged over the duration of the sequence to obtain a "reference image" that corresponds to the image background (Fig. 5c). Grain edge detection from the reference image was conducted using the Canny method (Fig. 5d).36 The reference image was subtracted from each image of the sequence, (Fig. 5e) producing a binary image of particles only, in white (Fig. 5f).

In this work, more than 600 images were extracted from the original video and post-processed prior to use in a PIV
algorithm. Velocities of particles in the final images were calculated by PIVLab. The initial movie was recorded at 30 fps. To run PIVLab, one image for every three frames was used in order to have enough particle displacement from one frame to the other such that there was at least a displacement of one to two particle sizes per frame. This leads to a frame rate of 10 images per second. For running PIVlab, three passes with windows of 64 × 64, 32 × 32 and 16 × 16 pixels were selected and arranged at 50% overlap. Based on the image spatial resolution (0.4 μm per pixels), the smallest window contains at least three particle images. Micro-PIV measurements usually suffer from low particle image density, thus to ensure fully converged measurements, time averaging over the 600 image pairs was performed to increase the number of particle images contributing to a micro-PIV evaluation. Moreover, the thickness over which moving particles are imaged is on the order of the thickness of our micro-models (≈14 μm). Therefore, the velocities are measured for only one focal plane that corresponds to the whole thickness. Thus, the velocities measured are assumed to be an average velocity for each depth.

3.4 Permeability and fluid saturation measurements

The micromodel was placed under a Nikon ME600 microscope. The objective lens used to track the motion of the fluids in the micromodel has a magnification of 10 and a numerical aperture of 0.3. The light source was a Metal Halide lamp, whereas a CCD camera (Nikon Coolpix 5100, 640 × 480 pixels at 30 fps, 8-bits) was used to acquire image sequences. The recorded images have a spatial resolution of 0.4 μm per pixels. All the experiments are conducted at room temperature.

Imbibition and drainage processes are crucial to oil recovery. Imbibition signifies non-wetting fluid removal from porous media by wetting fluid and drainage is the displacement of wetting fluid from pore space by non-wetting fluid. Hence, absolute and end point relative permeability measurements throughout the drainage and imbibition processes were conducted sequentially to investigate the meaningful change in fluid dynamics in both the single- and dual-depth micromodels.

First, the fluorescent deionized water was injected using an 8 mL syringe (Harvard Stainless Steel Syringes) and a syringe pump (Harvard Apparatus, Holliston, MA) setup, as shown in Fig. 3B. The pore space in the micromodel was fully saturated with water in order to maintain micromodel surface wettability as strongly water-wet. A 10−3 M fluorescein dye solution gave a green fluorescence to the water phase for better color-based oil and water phase identification. During the initial water injection, the absolute permeability (k) of the micromodel was calculated using Darcy’s law:

\[ k = -\left(\frac{q\mu_w}{A}\right)\left(\frac{L}{\Delta P}\right) \]

where A is the cross sectional area (product of width, 4.5 cm, and etching depth (H2)), \( \Delta P \) is the pressure drop in atm, \( \mu_w \) is the viscosity of water in cp, q is the volumetric flow rate in ml s⁻¹, L is the length (6 cm), and k is the absolute permeability in Darcys. Several steady-state pressure drops across the micromodel were measured at different flow rates (0.001–
0.02 ml min\(^{-1}\)) as shown in Fig. 3(C) and (E). The capillary number (\(N_{ca}\)), the ratio of viscous force to capillary force, is defined as:

\[
N_{ca} = \frac{V \mu}{\sigma_{ov}} \tag{3}
\]

\(V\) is the interstitial velocity, \(\mu\) is the displacing phase viscosity, and \(\sigma_{ov}\) is the interfacial tension of \(n\)-decane and water phase, 52 mN m\(^{-1}\). The range of capillary numbers is from \(1.3 \times 10^{-6}\) to \(2.6 \times 10^{-5}\) indicating capillary-dominated flow conditions in the micromodel. Using the dimensions of the micromodel, known constants, and measured variables, the product of \(q_1\), \(\mu_{n-o}\) and \(L\) versus the product of \(A\) and \(\Delta P\) was plotted, and the slope, that is equal to \(k\), was determined (plots are shown in Fig. S-10 of the ESI\(^\dagger\)).

For forced water drainage experiments, injection port (I-B) in Fig. 3 is closed when injected oil from I-A filled the flow distribution channel. Similarly, the \(n\)-decane solution with 1 vol% of crude oil (\(\mu_{o} = 0.859\)) was continuously injected to displace the water phase at constant pressure, 100 psi (maximum pressure limit of these micromodels), for 100 PVI (pore volume injected) until the residual water saturation (\(S_{wr}\)) is reached. The residual water saturation was calculated by averaging the saturations of images at nine locations (Fig. 3) within the micromodel using image segmentation by the RGB-based threshold method in ImageJ.\(^{21}\) Phase saturations are calculated based on the number of pixels corresponding to either the water phase (\(n_w\)) or oil phase (\(n_o\)).

\[
\begin{align*}
S_{water} &= \frac{n_w}{n_w + n_o}, \quad S_{oil} = \frac{n_o}{n_w + n_o} \tag{4}
\end{align*}
\]

At the residual water saturation (\(S_{wr}\)), the end point oil permeability (\(k_o\)) was calculated by monitoring the pressure difference. Lastly, a forced water imbibition experiment was conducted to flush out \(n\)-decane from the micromodel at 100 psi for 100 PVI until the residual oil saturation (\(S_{or}\)) is reached. When steady state was obtained at \(S_{or}\), the end point water permeability (\(k_w\)) was measured. Image segmentation was used to obtain the oil saturation corresponding to the \(k_w\). A schematic diagram for the entire process of permeability measurement is presented in Fig. S-9 of the ESI.\(^\dagger\)

4. Results and discussion

Single- and dual-depth micromodels were fabricated as described above. Their topological and flow properties were then compared.

4.1 Characterization of micromodel

A portion of the etched dual-depth wafer was processed using a CCI HD-3D surface and film thickness optical profiler (Tay-lor Hobson USA, West Chicago, IL). The profiler is used to measure the surface topography of etched wafers. Fig. 6 shows the surface topology for corresponding regions of the single- and dual-depth wafers. The figure illustrates the successful alignment and execution of the sequential-etching fabrication. Using the CCI HD step height profile in Fig. 6, etching depths were measured as \(H_1 = 7\ \mu m\) and \(H_2 = 14\ \mu m\) for the micro- and macro-porous regions, respectively. The etched depth of the single-depth wafer is the same as the maximum depth (\(H_2\)) of dual-depth case, 14 \(\mu m\). Microporous regions (i.e., etch depth = \(H_1\)) are apparent in fluid-filled micromodels having lesser luminescence compared to more deeply etched (\(H_2\)) pores (Fig. 6a and b).

The porosity, including the single- and dual-etched depths, is calculated using a geometrical interpretation expressed as (see ESI\(^\dagger\)):

\[
\left(\frac{a \times \phi_1 + \phi_{\text{micromodel}}}{a}\right) = \phi_{\text{dual-depth}} \tag{5}
\]

where \(\phi_{\text{dual-depth}}\) is the porosity of the dual-depth carbonate micromodel and \(a\) is the ratio \(H_2/H_1\). The dual-depth micromodel porosity was calculated as 39.7% volume whereas the porosity of the single-depth micromodel is 45.3% volume. The porosity of the dual-depth micromodel is 87.6% that of the single-depth micromodel.

4.2 Velocity profile from \(\mu\)-PIV

The existing pressure versus flow rate relationship in the microfluidic channels (rectangular conduits) of micromodels, as shown in Fig. 7, has been studied. Assuming the upper limit of the width to height ratio is one and using a Fourier sum representation, the approximate solution (within 10% error) for the Navier–Stokes equation for a fully developed, steady-state flow with a Newtonian fluid between parallel rigid plates was expressed by Bruus (2008) and Cheung et al. (2012).\(^{39,40}\) The pressure drop (\(\Delta P\)) across microchannel length (\(L\)) is represented as:

\[
\Delta P = \frac{12q \mu L}{H^4(W/H - 0.63)} \tag{6}
\]

where \(q\) is the imposed flow rate, \(H\) is the height of the microchannel, and \(\mu\) is the viscosity of the Newtonian fluid. The pressure drop is a function of flow rate, height, and width of the cross sectional area. More importantly, given 0.001 ml min\(^{-1}\) injection of water into a 6 cm long channel with 500 \(\mu m\) width and 20 \(\mu m\) height, a reduction of both channel width (20 \(\mu m\)) and height (10 \(\mu m\)) increases the pressure drop roughly four times greater than the case in which only the width changes. Regardless of the importance of the height and width on the pressure drop across a microchannel, typical microfluidic studies use devices with microchannels where the etching depth (\(H\)) is constant. Hence, the
The pressure drop in a microchannel is controlled only by the change in channel width.

The μ-PIV technique provided velocity profiles for both single- and dual-depth dual-porosity micromodels (Fig. 8) under single-phase flow condition. The velocity values are normalized by the maximum velocity in the area under study. Accordingly, velocity profiles are used to evaluate the impact of change in the aspect ratio (height/width) of the channel on the fluid flow velocity. Specifically, in the local region of interest shown in Fig. 8, the matrix with a microchannel is located in between two large pore spaces in the single-depth micromodel. The aspect ratio of the fluid channel increases due to the reduction of width as the fluid meets the entrance throat to the microchannel.

For the dual-depth carbonate micromodel, the increase in the aspect ratio is less than the change observed in the single-depth micromodel, because both the height and width of the same region are reduced. Dashed-circles in Fig. 8 indicate three transition zones where water phase transports from the large pore to microchannel for both micromodels.

In practice, the limitation of using uniform-depth porous media is most obvious at pore-size transition zones. The μ-PIV results demonstrate the sudden jump of fluid horizontal velocity at constrictions or pore throats within the single-depth micromodel (indicated by yellow and red regions within dashed circles; Fig. 8). On the other hand, as expected, these large flow velocities are not observed in the corresponding regions of the dual-depth micromodels. The dual-etched
depths serve to reduce the aspect ratio change compared to the single-etched depth model. The dual-depth micromodel has the smaller cross sectional areas at the pore throat imposing the greater resistance against the flow than the resistance of single-depth. In addition, depth differences (reduction by half) in the dual-etch case significantly impact the flow at constrictions or pore throats more than only width reduction of the single-etch depth case by two orders of magnitude.

### 4.3 Permeability comparison

Absolute permeability ($k$) is used to quantify the capacity for flow through porous material with dimensions of length squared, often expressed as Darcy ($1 \text{ D} = 10^{-12} \text{ m}^2$). Absolute permeability for the single- and dual-depth micromodels is 0.298 D and 0.178 D, respectively. The permeability ($k_{\text{dual}}$) with $H_2/H_1$ (=2) of the dual-depth micromodel is 60% of the single-depth micromodel permeability ($k_{\text{single}}$). The reduction in dual-depth absolute permeability is related to the reduction in cross-sectional area for flow. More importantly, this result closely mimics the observation of flow behavior from the $\mu$-PIV velocity profile where water showed preferential flow through macro-pores and experienced a barrier at the entrance to micropores in the dual-depth micromodel.

To further compare the single- and dual-depth micromodels, end point relative permeabilities were measured. End point relative permeability values are important parameters to quantify the modified transport through porous media due to the presence of trapped oil and water. The end point permeabilities of water ($k_w$) and oil ($k_o$) at residual phase saturations were evaluated using the best linear fit of data points. The value for $k_w$ is 0.155 D for the dual-depth micromodel and 0.069 D for the single-depth micromodels. $k_o$ at the residual oil saturation was determined to be 0.131 D for the dual-depth micromodel and 0.060 D for the single-depth micromodel. The values for the relative permeability, $k_{\text{rim}} = k_{\text{im}}/k_{\text{m}}$ where i is either water or oil phase and m is denoted as single- (s) or dual- (d) depth micromodel, were calculated and tabulated in Table 1. For the dual-depth water-wet micromodel, a drainage experiment (oil injection) left 7.6% more connate water in the porous medium than the single-depth case (Table 1). This additional water was predominantly spread over microporous regions and became an obstacle to
oil flow. It was difficult to develop sufficient capillary entry pressure for the oil phase to enter these microporous water-saturated zones. Thus, the end point oil relative permeability \( k_{ro\text{d}} = 0.39 \) for the dual-depth micromodel is 0.13 lower than \( k_{ro\text{s}} = 0.52 \) for the single-depth case.

The water end point relative permeability was measured at \( S_{or} \), which were 33.2% and 34.3% for single- and dual-depth micromodel, respectively. Even though there is a relatively insignificant difference (±1.1%) of \( S_{or} \) between single- and dual-depth micromodels, the end point water permeability \( k_{rw\text{d}} = 0.34 \) for the dual-depth case is 0.1 less than \( k_{rw\text{s}} = 0.43 \) of the single-depth micromodel. This is mainly attributed to residual oil trapping in macro pores for the dual-etched depth case and micro pores for the single-etched depth case. To be more specific, the dual-depth micromodel may trap more oil in macro-pores, because the pore-throat aspect ratio increases the capillary pressure threshold that is needed to drive the oil phase into narrow water-wet micro-channels or throats.

The single-depth micromodel, however, has an insufficient capillary pressure threshold across the pore throat to prevent the non-wetting fluid from entering microchannels. Eventually for the single-depth case, water bypassed the residual non-wetting oil phase trapped in microporous zones (with narrower widths) and flowed through macro-pores and along the pore walls due to capillary force and the adverse mobility ratio between the oil and water phase. For dual-depth micromodels, the existence of the trapped oil ganglia in macrochannels may cause the wetting fluid to flow around the ganglia or bypass the microchannels.41 Hence, compared to the single-depth micromodel, the reduction of available channel sizes for water transport is more significant and thus represented as a decrease in relative permeability for the dual-depth case.

In the next section, visualization of oil and water distributions further confirms this interpretation of the relative permeability reduction, oil trapping, and preferential flow of water.

### 4.4 Visualization of fluid desaturation

The pore structure in many carbonate rocks has disordered channel networks unlike the relatively homogeneous pore geometries with high pore-to-pore connectivity found in many sandstones. Consequently, high permeability and porosity values associated with these disordered channel networks do not guarantee an increase in oil recovery.4 Understanding oil recovery processes for complex pore networks, therefore, requires further investigation beyond porosity and relative permeability analyses.

Throughout the experimental steps of the permeability measurements, water drainage and imbibition processes provide evidence of significantly different capillary forces in the single- and dual-depth micromodels. The capillary force dominating immiscible displacement was expected to be larger in the dual-depth micromodel, and the experimental result for oil recovery agreed with this preliminary estimation.

As noted in previous sections, visual observation of the pattern of phase saturation in the micromodel reveals the residual oil phase trapping mechanisms and preferential flow of water. During the water drainage process by oil injection for dual-depth micromodel, Fig. 9(a) show that the oil

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**Table 1** Summary of end point relative permeability corresponding to the residual water and oil saturations, \( S_{rw} \) and \( S_{ro} \), respectively

<table>
<thead>
<tr>
<th></th>
<th>( S_{rw} )</th>
<th>( k_{ro} )</th>
<th>( S_{ro} )</th>
<th>( k_{rw} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single-depth</td>
<td>32.5</td>
<td>0.52</td>
<td>33.2</td>
<td>0.44</td>
</tr>
<tr>
<td>Dual-depth</td>
<td>40.1</td>
<td>0.39</td>
<td>34.3</td>
<td>0.34</td>
</tr>
</tbody>
</table>

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**Fig. 9** For dual-depth micromodel: (a) water (green) drainage experiment shows the water-saturated shallow-depth (\( H_1 \)) microporosity (darker green) and deeper (\( H_2 \)) macro-porosity (bright green). Oil phase (blue) displaces the water phase in the macro pores. (b) Water imbibition experiment shows oil ganglia (blue) occupying macro pores after 100 PVI water flooding at 100 psi. Oil ganglia meets pore throats with a cross section of 20 \( \mu \)m width by 7 \( \mu \)m depth (X-box in a). Scale bar is 100 \( \mu \)m.
preferentially displaced water in the macro pore space; hence, a larger portion of the microporous space remained water saturated. As expected after the water imbibition experiment, the existence of trapped oil ganglia in macro pores (Fig. 9b) is evidence of wetting fluid either flowing around the oil ganglia or bypassing along the pore wall into microporous regions.

In contrast, the oil phase flushed into the microporous area of the single-depth micromodel during oil flooding, because it had an insufficient capillary entry pressure barrier (Fig. 10a). Water bypassed the residual non-wetting phase trapped in the microporous zone and displaced preferentially the oil remaining in the macro pores during the water imbibition experiment as shown in Fig. 10(b). These results concur with the larger relative permeability values for the single-depth micromodel compared to the dual-depth model. The oil recovery values for single- and dual-depth micromodels were calculated by averaging the oil recovery values from the nine regions of interest. The resulting oil recovery (50.8%) from the single-depth micromodel was 7.9% greater than the oil recovery (42.7%) in the dual-depth micromodel. Preferential displacement of oil from macropores by water in the single-depth case yields greater oil recovery as compared to the dual-depth model. The macropores comprise the majority of the pore structure, by volume, in the micromodel and hence changes in fluid occupancy of macropores dominate recovery.

5. Concluding remarks

For the purpose of investigating multiphase flow through heterogeneous dual-porosity pore networks, a microfluidic device should have certain 3D characteristics to better represent and mimic real rocks. To accomplish this, modification of the conventional mask and fabrication steps for a dual-porosity micromodel generated multiple pore depths with a suitable change in cross-sectional height corresponding to the width of the pores. Using multiple masks, an original base image and an overlay, created using E-D processing of the original base image, provided an easy method of preserving selected pore sizes that are, in this case, macro pores greater than 21 μm in diameter. The size threshold is determined by the number of E-D cycles and the pixel size. We established a reliable and repeatable fabrication routine for dual-depth silicon microfluidic devices using an effective approach to the spray coating, exposure, and developing processes. This methodology could be extended to micromodels with three and four etch depths.

Characterization methods including porosity and permeability measurements serve to compare the single- and dual-depth micromodels. The porosity of the dual-depth micromodel \( (H_2/H_1 = 2) \) was calculated as 39.7% compared to 45.3% porosity of a single-depth micromodel. The absolute permeability values of the single- and dual-depth micromodel with 14 μm maximum depths were 0.298 D and 0.178 D, respectively.

We compared flow within the dual-depth networks to that in the single-depth micromodels using various quantification methods. The observation of the change in velocity field using μ-PIV provided an opportunity to evaluate the dynamics of single-phase flow in the pore space of a dual-porosity micromodel for both single and dual etched depth cases. The μ-PIV measurements demonstrated differences in fluid flow behavior at the pore-size transition zone due to the modification of height-to-width aspect ratio of cross section of channel.

Finally, we investigated the relative permeability of single- and dual-depth micromodels at residual oil and water saturations to quantify the capacity for flow of the wetting and non-wetting phases. The end point relative permeability of oil \( (k_{ro}) \) is 0.39 for dual-depth and 0.52 for single-depth micromodels. The end point permeability of water \( (k_{rw}) \) was calculated as 0.34 for dual-depth and 0.44 for single-depth micromodels. Visual observation of the oil and water phase demonstrated enhanced capillarity whereby residual oil
remains in macropores and prevents preferential water flow through macropores. This causes the reduction of relative permeability for the dual-depth micromodel. Due to the increased capillarity, oil recovery (50.8%) in the single-depth micromodel was 7.9% greater than the oil recovery (42.7%) in the dual-depth micromodel.

The new methodology provides an optimized process to develop and fabricate microfluidic devices with more realistic 3D features and diverse approaches to qualify and quantify the improvements provided by the dual-depth micromodel as a tool for the study of multiphase flow. The use of dual-depth dual-porosity micromodels with carbonate pore networks may further knowledge of the fluid dynamics underlying the complex interactions among oil, water, steam, gas, surfactant, and foam for enhanced oil recovery processes (EOR). In addition, combining dual-depth dual-porosity micromodels with surface treatments for wettability modification may help elucidate the role of wettability and the surface-fluid interaction on the EOR mechanism.

Our fabrication method allows the height ratio ($H_2/H_1$) of a dual-porosity pore network to be tuned; hence, it paves the way for the systematic investigation of the effect of various dimensions in the pore throat on the fluid-flow and oil-desaturation. Especially, a tunable pore throat dimension is beneficial to study effects of interfacial tension reducing agents on extracting hydrocarbon.

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**Notes and references**