Fluorescent Visualization of Oil Displacement in a Microfluidic Device for Enhanced Oil Recovery Applications

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Materials and Methods

Surface Tension Measurements

The surface tension measurements were performed on a Biolin Theta optical tensiometer. A pendant drop tensiometry method was used to measure the surface tension of deionized (DI) water, DI water spiked with 100 μM Rhodamine 6G (Chem-Impex Int'l Inc.), decane, and decane spiked with 100 μM Nile Red (Sigma-Aldrich). A 1 cm × 1 cm × 4.5 cm quartz cuvette filled with the desired solution was placed in the optical path. A pendant drop of 5 μL dispersion was generated on a blunt needle, and the surface tension was measured in 10 s by averaging 140 measurements. An average surface tension value of the dispersion was determined by analyzing the droplet curvature using Young−Laplace as follows. The interfacial tension (γ) was measured from $\gamma = \Delta\rho g R_0^2/\beta$, where $\Delta\rho$ is the density difference between fluid, g is gravitational constant,

R_0 is the radius of drop curvature at apex, and β is a shape factor, calculated from the Young-Laplace equation.[1]

$$d\phi/ds = 2 + \beta z - sin\phi\ x \quad \text{Eqn. S1}$$

$$dx/ds = cos\phi \quad \text{Eqn. S2}$$

$$dz/ds = sin\phi \quad \text{Eqn. S3}$$

Where ϕ is the contact angle, s is the curvature, z is the vertical axis and x is the horizontal axis. The average surface tension values were further analyzed using a single-tailed t-test to ensure similar surface tension between pure and spiked fluids using Origin software. The threshold (p-value) for all experiments was set to 0.05.

Visualization of Different Phases Inside the Microfluidic Channels

To test device functionality, the tubing was placed in the inlet and outlet ports to inject fluids into the device. Fluids were injected by syringe pumps (KD Scientific) using a 5 mL syringe (BD Syringe) and a 23-gauge needle (BD PrecisionGlide Needle). Decane spiked with Nile Red (100 μM) was first injected into the device at a rate of 300 μL/h for 1 min, followed by an injection of DI water spiked with Rhodamine 6G (100 μM) at a rate of 300 μL/h for 1 min. The device was imaged using a fluorescent DMi8 inverted microscope (Leica microsystems) outfitted with a 10x objective (Leica HC PL FL L, 0.4x correction) under brightfield. Images were acquired using the digital CMOS camera C11440 (Hamamatsu Photonics K.K.) with a fixed exposure time of 100 ms for the FITC and Rhodamine filters and 25 ms for brightfield. The following excitation/emission filters (Chroma Tech. Corp) were used to image the device: fluorescein isothiocyanate-FITC (λ_{ex} 440-520 nm and λ_{em} 497-557 nm) for capturing the signal from Nile Red; Rhodamine (λ_{ex} 536-556 nm and λ_{em} 545-625 nm) for capturing the signal from Rhodamine 6G. Air trapped in the device after injection was identified by a lack of fluorescent signal in the overlay image. Image

acquisition was controlled using the Leica Application Suite software (LAS X), where all images were recorded using the same parameters. The imaging was followed by a series of robust quantitative analysis, where 10 manual line scans were drawn across each water/oil channels as the region of interest (ROI) to quantify the fluorescent signals of each phase.

Decane Recovery, Image Processing and Analysis

An additional step was performed for oil recovery experiments, where the device was flushed with air to remove oil from the device. Following the injection of decane and water as previously described, images were taken of the entire device. Air was then injected into the device for 1 min at rates varying from 1000-5000 μL/h and the entire device was imaged again. Oil displacement was quantified by determining the change in the surface area of the decane inside the microfluidic device. All microscope images of the device were processed with ImageJ (NIH). The images from the FITC filter which was used to visualize decane were first converted to the RGB image type. The color threshold of the image was then used to select decane present in the channels for the analysis. Adjusting the brightness setting in the color threshold menu was necessary to ensure only signal from the Nile Red dye was included in the calculation and not bleed-through from the Rhodamine 6G. Once the selected region in the RGB image matched the decane shown in the overlay image, the area of the region could be determined by ImageJ. The total amount of decane was determined by combining the calculated areas for all images of the device at each condition, with each device taking approximately one hour to analyze. Normalized oil recovery was calculated by dividing the total area of decane in the device before and after flushing with air using the following equation:

$$Normalized\ EOR = 1 - \frac{Total\ area\ of\ decane\ after\ air\ injection}{Total\ area\ of\ decane\ before\ air\ injection}. \quad \text{Eqn. S4}$$

Table S1. Quantitative analysis confirmed minimal spectral overlap between the DI water spiked with Rhodamine 6G (100 μM) and decane spiked with Nile Red (100 μM) in Rhod Filter.

Null Hypothesis	**F-value**	**Pr > F**	**Accept/Reject Null**
Mean Intensity (DI Water) = Intensity (Decane)	140.3	6.22E-10	Reject

Table S2. Quantitative analysis confirmed minimal spectral overlap between the DI water spiked with Rhodamine 6G (100 μM) and decane spiked with Nile Red (100 μM) in FITC Filter.

Null Hypothesis	**F-value**	**Pr > F**	**Accept/Reject Null**
Mean Intensity (DI Water) = Intensity (Decane)	44.8	2.83E-06	Reject

References

1. Mobius, D.; Miller, R., Drops and Bubbles in Interfacial Research. Elsevier: 1997; Vol. 6.