# Locally-calibrated light transmission visualization methods to quantify nonaqueous phase liquid mass in porous media

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### ARTICLE INFO

### ABSTRACT

Five locally-calibrated light transmission visualization (LTV) methods were tested to quantify nonaqueous phase liquid (NAPL) mass and mass reduction in porous media. Tetrachloroethylene (PCE) was released into a two-dimensional laboratory flow chamber packed with water-saturated sand which was then flushed with a surfactant solution (2% Tween 80) until all of the PCE had been dissolved. In all the LTV methods employed here, the water phase was dyed, rather than the more common approach of dyeing the NAPL phase, such that the light adsorption characteristics of NAPL did not change as dissolution progressed. Also, none of the methods used here required the use of external calibration chambers. The five visualization approaches evaluated included three methods developed from previously published models, a binary method, and a novel multiple wavelength method that has the advantage of not requiring any assumptions about the intra-pore interface structure between the various phases (sand/water/NAPL). The new multiple wavelength method is also expected to be applicable to any translucent porous media containing two immiscible fluids (e.g., water-air, water-NAPL). Results from the sand-water-PCE system evaluated here showed that the model that assumes wetting media of uniform pore size (Model C of Niemet and Selker, 2001) and the multiple wavelength model with no interface structure assumptions were able to accurately quantify PCE mass reduction during surfactant flushing. The average mass recoveries from these two imaging methods were greater than 95% for domain-average NAPL saturations of approximately $2.6 \times 10^{-2}$, and were approximately 90% during seven cycles of surfactant flushing that sequentially reduced the average NAPL saturation to $7.5 \times 10^{-4}$.

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## 1. Introduction

Laboratory studies of multiphase fluid flow problems are commonly conducted in benchtop two-dimensional flow chambers because the experimental conditions are controllable, measurements are easily conducted, and the capillary, viscous, and buoyancy forces found in the subsurface environment can be reproduced (Chevalier and Petersen, 1999). Such flow chambers have become one of the most important tools for studying multiple-phase fluid migration in homogeneous and heterogeneous porous media (Jones and Smith, 2005; Kechavarzi et al., 2005; Nambi and Powers, 2003; Ramsburg and Pennell, 2002), development and

evaluation of aquifer remediation strategies (Chevalier, 2003; Conrad et al., 2002; Jawitz et al., 1998) and model development and verification (Chrysikopoulos et al., 1994; Clement et al., 2004; Fure et al., 2006).

In laboratory scale experiments, non-intrusive and non-destructive measurement of fluid saturation can directly provide both local and instantaneous information about multi-phase fluid dynamics in porous media. Measurement methods include gamma ray attenuation (Tidwell and Glass, 1994; Tuck et al., 1998), X-ray attenuation (Hilpert et al., 2000; Liu et al., 1993; Moreno-Barbero and Illangasekare, 2006), magnetic resonance imaging (Chu et al., 2004) and light transmission or reflection (Conrad et al., 2002; Fure et al., 2006; Darnault et al., 1998; Kechavarzi et al., 2000; Niemet and Selker, 2001; O'Carroll et al., 2004; Zhang and Smith, 2002). Of these methods, light transmission and reflection

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require the least amount of specialized equipment and do not require hazardous radiation. Furthermore, for laboratory experimental chambers of many different scales (i.e., from centimeter to meter) a single image collected with a digital camera can at any time provide information about multi-phase fluid distributions over the entire chamber area with very high spatial resolution and very high sensitivity to changes in fluid saturation.

Quantitative visualization with a digital imaging system requires either 1) external calibration chambers coupled with empirical relations between light intensity and fluid satura-tion, or 2) locally-calibrated light transmission methods. In the former method, known ratios of solid and fluids are packed into chambers resulting in predetermined fluid saturations. The light transmission or reflection character-istics (such as intensity or hue) of these chambers are measured in order to obtain empirical relations that may be used for quantifying unknown fluid distributions in ot-her systems. For example, Kechavarzi et al. (2000) used 74 calibration chambers to calibrate two linear equations to describe the relationship between reflective light densities and water, air, and nonaqueous phase liquid (NAPL, Soltrol dyed with oil Red O) saturations in five different sands, and applied these equations to quantify NAPL infiltration and redistribution in homogeneous and layered-heterogeneous sands in a two-dimensional flow chamber. Darnault et al. (1998, 2001) used calibration chambers to calibrate multiple relationships between water (dyed with $CuSO_4$) content and transmission light hue as well as multiple relationships between total liquid content (water+Soltrol) and light intensity, and applied them to measure water fingers emanating in homogeneous sand-oil-air systems. Similar methods were recently used to quantify tetrachloroethylene (PCE) saturation distributions following surfactant flushing (Suchomel and Pennell, 2006).

Locally-calibrated methods are directly developed from light transmission models that explicitly account for the effects of both light absorption from the bulk volume of media and light refraction at the interfaces between pha-ses. These light transmission models are described as a combination of Beer's Law and Fresnel's Law where light incidence on the flow chamber is assumed to be normal. Such models have been applied to water–air systems in homogeneous sands (Tidwell and Glass, 1994; Niemet and Selker, 2001).

Light transmission visualization (LTV) methods have received limited application to quantify NAPL dissolution dynamics (Conrad et al., 2002; Fure et al., 2006; Suchomel and Pennell, 2006). Conrad et al. (2002) identified two important problems with current LTV methods for studying NAPL dissolution. First, current quantitative visualization methods have yet to solve the problem of the intrinsically variable structure of the interfaces between phases (solid–fluid, and fluid–fluid). When light propagates through a heterogeneous system comprised of phases with different refractive indices (such as in a sand–water–NAPL system), light is refracted at the interface between two phases resulting in a loss of light transmission intensity. Thus, transmitted light intensity is a function of both fluid saturation (i.e. bulk effect of light adsorption) and interface structure. Most researchers have elected to minimize the uncertainties associated with the

interface effect by decreasing the relative significance of light refraction compared to light absorption. This can be accom-plished by dyeing the NAPL such that the resulting light transmittance is dominated by the bulk effect rather than the interface effect and can then be approximately quanti-fied using the calculation method analogous to that used in X-ray transmission (Glass et al., 2000) or by applying Beer's law only.

This leads to the second major problem identified by Conrad et al. (2002). In LTV methods that rely on dyed NAPL, fluid dye concentrations should remain constant for the experimental duration of interest. This limitation may be unacceptable for quantifying the NAPL distribution during aqueous dissolution or remediation by flushing with surfac-tant or co-solvent (Fure et al., 2006). In order for dye concentrations in the NAPL to remain constant during remediation, the dye must be solubilized from the NAPL to the remedial fluid at the same rate as the NAPL. In practice, achieving this same release rate is difficult to realize, and dyed-NAPL visualization during dissolution has been reported for qualitative rather than quantitative analysis (Conrad et al., 2002; Fure et al., 2006). Suchomel and Pennell (2006) also used dyed NAPL for LTV visualization during surfactant-enhanced dissolution, but these authors did not comment on the relative solubilization rates of dye and NAPL.

Calibration-chamber based LTV methods are also subject to several other limitations that could be overcome through the use of local calibration. Methods based on the former use the mean of pixel-scale observations integrated over the entire calibration chamber to calibrate observations at the pixel scale in the experimental chamber. In practice, such calibration requires that the local porosity in the experimental chamber should be the same as in the ca-libration chamber. This requirement limits flexibility in experimental design by precluding application to arbitrary heterogeneous media. Analysis of heterogeneous domains is limited to homogeneous blocks of one porous media type included within or adjacent to another homogenous porous media type. Calibration chambers and experimental cham-bers must also be carefully packed to ensure congruent properties.

The goal of this work was to develop, test and compare locally-calibrated LTV methods that can be used to quantify NAPL mass and mass reduction by chemical remediation processes in porous media. Five LTV methods were tested, including four methods developed from previously pub-lished light transmission models in porous media, and a novel multiple wavelength method that has the advantage of not requiring any assumptions about the intra-pore interface structure between the various phases (sand/water/ NAPL). In all the LTV methods employed here, the water phase was dyed, rather than the more common approach of dyeing the NAPL phase, such that the light adsorption characteristics of NAPL did not change as dissolution progressed. Also, calibration was conducted locally at the pixel-scale in the experimental chamber, such that none of the methods used here required the use of calibration chambers. The new multiple wavelength method is also expected to be applicable to any translucent porous media containing two immiscible fluids (e.g., water-air, water–NAPL).