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Non-equilibrium partitioning tracer transport in porous media: 2-D physical modelling and imaging using a partitioning fluorescent dye

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Abstract

This paper describes an investigation into non-equilibrium partitioning tracer transport and interaction with non-aqueous-phase liquid (NAPL) contaminated water-saturated porous media using a two-dimensional (2-D) physical modelling methodology. A fluorescent partitioning tracer is employed within a transparent porous model which when imaged by a CCD digital camera can provide full spatial tracer concentrations and tracer breakthrough curves. Quasi one-dimensional (1-D) benchmarking tests in models packed with various combinations of clean quartz sand and NAPL are described. These modelled residual NAPL saturations, S_n, of 0–15%. Results demonstrated that the fluorescent partitioning tracer was able to detect and quantify the presence of NAPL at low flow rates. At larger flow rates and/or higher NAPL saturations, the tracer increasingly underpredicted the NAPL volume as expected and this is attributed primarily to non-equilibrium partitioning. Despite little change in permeability, change in NAPL saturations from 4% to 8% resulted in significant NAPL saturation underestimates at the same flow rates implying coalescence of NAPL into wider separated but larger ganglia. A 2-D investigation of an idealised heterogeneous residual NAPL contaminated flow field indicated little permeability change in the NAPL contaminated zone and thus little flow bypassing, leading to reduced underpredictions of NAPL saturations than for equivalent quasi 1-D cases. This was attributed to increased 'sampling' of the NAPL by the tracer. The process is clearly visually identifiable from the experimental images. This rapid and relatively inexpensive experimental method is of value in laboratory studies of partitioning tracer behaviour in porous media; in particular, the ability to observe full field concentrations makes it valuable for the study of complex heterogeneous systems.

Keywords: Porous media; Fluorescent tracer; Non-invasive technique; Partitioning tracer; NAPL characterisation; Residual saturation; Flow bypass

1. Introduction

The location and characterisation of non-aqueous-phase liquids (NAPLs) in the subsurface have presented significant challenges to scientists and engineers involved

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Nomenclature

b	thickness of porous media
c, c_0	concentration of tracer
l	slug length
m	mass of tracer
t	time
v_s	seepage velocity
x	cartesian coordinate (perpendicular to flow direction)
D_d^*	diffusion coefficient of tracer in porous media
K_{nw}	tracer NAPL–water partitioning coefficient
L	length of contaminated zone
R_d	retardation coefficient
S_n	NAPL saturation

in risk assessment and remediation of contaminated sites. Leakages, spills and improper storage and disposal of organic liquids can lead to large dissolved and non-aqueous-phase plumes in the subsurface.

Inherent difficulties with conventional NAPL characterisation techniques have prompted the recent development of innovative, non-invasive techniques utilising partitioning and interfacial groundwater tracers for NAPL quantification (Rao et al., 2000). Partitioning tracers reversibly partition to different degrees between NAPL and groundwater. By calculating the retardation, R_d, of a partitioning tracer relative to that of a non-partitioning tracer from first moment analysis of the breakthrough curves (BTC), the NAPL saturation, S_n, within the tracer swept region can be theoretically quantified using the following equation (Jin et al., 1995):

$$S_n = \frac{R_d - 1}{R_d - 1 + K_{nw}}, \tag{1}$$

where K_{nw} is the tracer NAPL–water partitioning coefficient, and linear reversible partitioning is assumed. Tracers should be selected to provide adequate retardation to give reliable estimates of NAPL saturation but not so large that the tracer test duration is unreasonable. Jin et al. (1995) recommended a range of $1.2 < R_d < 4$. Equilibrium partitioning is not necessary for Eq. (1) to apply. However, lack of equilibrium does lead to practical difficulties in fully capturing the resulting long tail of a BTC.

The task of locating and characterising subsurface occurrences of NAPL using partitioning tracers is complicated by aquifer heterogeneity, which will increase both the complexity of the original NAPL migration and subsequent entrapment, and tracer migration patterns. The complex spatial distribution of NAPL is controlled by unstable fingering, preferential channelling and both micro- (pore) and macro-scale heterogeneity (layering and soil texture contrasts) of the subsurface formation. The final entrapment of these distributions in complex geologic environments ranges from regions of low (residual ganglia) to high saturation (free-phase pools and macro-scale entrapment zones resulting from the presence of capillary barriers). Such regions will often be separated from each other by zones of zero contamination. Assessing the influence of such heterogeneous NAPL distributions on tracer BTCs requires a combination of numerical and physical modelling. In the context of physical modelling, measurement of the tracer migration patterns through the entire flow field, such as through imaging techniques, is preferable. This paper describes the development of such an approach. The main objectives of this study were to:

1. Develop a non-invasive imaging technique for the quantitative monitoring of partitioning tracer migration behaviour. To the knowledge of the authors, this will allow the partitioning process to be visually observed and quantified in two-dimensional (2-D) flow fields for the first time.
2. Validate its application in simple quasi one-dimensional (1-D) NAPL contamination scenarios.
3. Investigate 2-D partitioning tracer behaviour through an idealised heterogeneous NAPL contaminated flow field consisting of uncontaminated and residual (low saturation) NAPL zones.

2. Non-invasive imaging systems

In recent years, a number of researchers have successfully utilised transparent porous media models coupled with light transmission/reflection and image analysis techniques for the acquisition and analysis of experimental data. A number of different research areas have been addressed, including solute transport, biological clogging, fluid content, surfactant floods and plume biodegradation (Corapcioglu and Fedirchuk, 1999; Huang et al., 2002, 2003; Kildsgaard and Engesgaard, 2002; Darnault et al., 1998, 2001).

Methods based on fluorescent tracers (Huang et al., 2002) are attractive due to their high sensitivity and resolution. They suffer less ambient light noise compared to absorbed/transmitted light methods as the only light captured is that emitted from the fluorescent tracer. Sensitivity is an important issue because the fluorescence signal is proportional to the concentration of the substance investigated. While absorbance measurements can reliably determine concentrations as low as several