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Capillary pressure-saturation relationships for diluted bitumen and water in gravel

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ABSTRACT

Spills of diluted bitumen (dilbit) to rivers by rail or pipeline accidents can have serious long-term impacts on environment and ecology due to the submergence and trapping of oil within the river bed sediment. The extent of this problem is dictated by the amount of immobile oil available for mass transfer into the water flowing through the sediment pores. An understanding of multiphase (oil and water) flow in the sediment, including oil trapping by hysteretic drainage and imbibition, is important for the development of spill response and risk assessment strategies. Therefore, the objective of this study was to measure capillary pressure-saturation (P_c-S_w) relationships for dilbit and water, and air and water in gravel using a custom-made pressure cell. The P_c-S_w relationships obtained using standard procedures in coarse porous media are height-averaged and often require correction. By developing and comparing air-water and dilbit-water P_c-S_w curves, it was found that correction was less important in dilbit-water systems due to the smaller difference in density between the fluids. In both systems, small displacement pressures were needed for the entry of non-wetting fluid in gravel. Approximately 14% of the pore space was occupied by trapped dilbit after imbibition, which can serve as a source of long-term contamination. While air-water data can be scaled to reasonably predict dilbit-water behaviour, it cannot be used to determine the trapped amount.

1. Introduction

Spills and accidental releases of crude oil during its overland transportation, for example by pipeline or rail, are potential sources of environmental contamination. When released to freshwater bodies, oil spills can be more damaging than marine spills because dilution is limited in confined freshwater systems (Trett, 1989). Typically, response to an oil spill is intended to address short-term adverse effects and includes limiting access to the spill site as well as containment and removal of the oil slick, for example using booms, suction or mechanical skimming (Vandermeulen and Ross, 1995; Brown, 1989). However, longer-term effects are also possible due to the persistence of oil under some conditions. For example, a spill of diluted bitumen to the Kalamazoo river in Michigan, USA in 2010 (Dollhopf et al., 2014) resulted in sinking of oil and oil contamination of the river bed. Removal of oil from the bed called for extensive remedial measures resulting in a total cost of US$1.2 billion (Fitzpatrick et al., 2015), and 20–30% of the spilled oil remained in the Kalamazoo river sediment three years after the spill occurred (King et al., 2014).

The Alberta oil sands in western Canada represent a significant oil reserve in the form of recoverable bitumen. Because bitumen is highly viscous, its transportation to ports and refineries through pipelines requires that it be diluted with low viscosity products (Government of Canada, 2013). The resulting mixture is referred to as diluted bitumen or dilbit. When released to freshwater, dilbit has the potential to sink and the immersed oil can settle onto the sediments, due to an increase in density caused by environmental weathering or by binding of oil droplets to suspended solids (Dew et al., 2015). River bed sediments, including gravels and coarse sands, are crucial for the reproduction of various fish species, including salmon and trout (Kondolf and Wolman, 1993). There is the potential for oil transported to gravel sediment to enter the large pores, become trapped in the pore spaces and act as a persistent source of contamination (Lee et al., 2015). To successfully manage the contamination of freshwater systems following an oil spill, remediation and risk management must address both the oil in the sediments as well as the oil in the surface water. A strategy that considers both the short- and long-term adverse effects requires an understanding of multiphase (oil and water) flow in the porous media that makes up the bed sediment.

An understanding of oil and water flow in porous media relies on the knowledge of capillary pressure-saturation (P_c-S_w) relationships (also referred to as retention curves), including the trapping

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of oil caused by hysteretic drainage and imbibition pathways. Quantifying the amount of trapped oil in gravel sediments is particularly important to estimate the mass of oil capable of acting as a long-term contaminant as well as the potential for dissolution of oil components to the surrounding pore water. Numerous studies have been done to measure P_c-S_w relationships for air-water and oil-water systems, many of which are related to groundwater contamination by non-aqueous phase liquids (NAPLs), but most studies have been conducted in porous media that are sand-sized or smaller. Few studies of P_c-S_w have been conducted in gravel (e.g., Tokunaga et al., 2002, 2004).

P_c-S_w relationships are typically measured in the laboratory by packing a porous medium into a pressure cell and measuring changes in saturation due to changes in fluid pressures (e.g., Dane et al., 1992; Sakaki and Illangasekare, 2007). Measurements, done in a step-wise manner, typically consist of drainage (a decrease in the saturation of the wetting fluid due to an increase in the capillary pressure) of a porous medium sample initially saturated with the wetting fluid, followed by imbibition (an increase in the saturation of the wetting fluid due to a decrease in capillary pressure). Measurements of displaced fluid volume represent changes in fluid saturation averaged over the sample volume. Calculating saturation by measuring displaced fluid volume (outflow or inflow), however, produces inaccurate results for some combinations of fluids and porous media (Liu and Dane, 1995; Tokunaga et al., 2002; Sakaki and Illangasekare, 2007). A height-averaged saturation is not equal to a local (point) measurement of saturation if capillary forces are weak, as occurs in coarse-grained media and with fluid pairs having small interfacial tensions (Dane et al., 1992). Because fluid pressures are often set (and measured) at the sample boundaries, capillary pressure and saturation values vary with height within the sample (Dane et al., 1992; Liu and Dane, 1995), and this variation is prominent within tall samples (Dane and Hopmans, 2002). Therefore, P_c-S_w curves developed with height-averaged capillary pressure and saturation values cannot represent P_c-S_w behaviour at a single point in systems with weak capillary forces. Local measurements of pressure and saturation must be made (e.g., Dane et al., 1992; Sakaki and Illangasekare, 2007), or height-averaged measurements must be corrected (e.g., Liu and Dane, 1995; Tokunaga et al., 2002; Schroth et al., 1996) to account for these variations.

The objective of this study was to measure P_c-S_w relationships for dilbit and water in gravel, and compare those relationships to those for air and water in gravel. Dilbit was chosen for this study because there is not enough information available on the fate and transport of diluted bitumen following a spill (Government of Canada, 2013), and that lack of knowledge can complicate cleanup efforts in freshwater systems (Dew et al., 2015). Air-water and dilbit-water relationships were compared to determine the potential to scale curves between fluid pairs, and dilbit displacement during imbibition was measured to determine the potential for gravel to trap dilbit, which could subsequently serve as a long-term contaminant source. These are the first such experiments to investigate drainage and imbibition of diluted bitumen and water, and it is expected that these results will help in estimating the possible risk of a dilbit spill and to develop effective remediation programs.

2. Background

2.1. Correction of height-averaged P_c-S_w data

While the standard procedure for measuring P_c-S_w relationships is based on boundary fluid pressures and total fluid volumes, certain combinations of factors such as grain size, sample height, fluid properties (density and interfacial tension) and configuration of the pressure cell can result in significant differences between average and local measurements. For example, capillary pressure varies with sample height when the densities of two immiscible fluids are different (Dane et al., 1992; Liu and Dane, 1995; Perfect et al., 2004). Korteland et al. (2010) emphasized that for samples in which two immiscible fluids are not uniformly distributed over the sample height, the centroid of the phases and the centroid of the sample are not the same. Thus, capillary pressure measured at a single elevation within the sample (e.g., mid-height) cannot represent capillary pressure at different elevations. In addition, during drainage of a coarse porous medium packed in a tall column, the top of the sample starts draining once the displacement pressure is exceeded, while the bottom remains saturated with the wetting fluid. This vertical stratification of saturation is observed in the air-water drainage of gravel, particularly at higher average wetting saturations (Tokunaga et al., 2004). Consequently, the saturation calculated using outflow volume is height-averaged and does not represent the saturation at the top or bottom of the column.

Using sample height and displacement pressure to represent the gravity and capillary forces, respectively, Liu and Dane (1995) showed that as the ratio of sample height to displacement pressure increases, the height-averaged curves are more likely to deviate from the point-measured curves. They also discussed the effect of density difference between the fluids and found that the disagreement between the curves is reduced when the densities of two immiscible fluids are equal. The requirement of shorter column heights must be balanced with the need for the sample to contain multiple material grains to meet the requirement of a representative elementary volume (REV). This presents a challenge for testing gravel samples, where capillary forces are weak due to large pore sizes (suggesting a need for small sample heights) but large grain sizes create large sample heights. For example, Tokunaga et al. (2004) found noisy results after gravity correction was applied to results from a test using 10 mm gravel in a 30 mm tall Tempe cell due to porosity variation caused by fewer grains contained in the sample.

In this study, height-averaged capillary pressure ($\bar{P}_c$) and saturation ($\bar{S}_w$) were corrected to obtain a local capillary pressure-saturation relationship using the method presented by Liu and Dane (1995), and the Brooks and Corey (1964) relationship for P_c-S_w. For drainage:

$$S_w = S_r + (1 - S_r)\left(\frac{P_d}{P_c}\right)^{\lambda} \text{ for } P_c \geqslant P_d \tag{1a}$$

$$S_w = 1 \text{ for } P_c < P_d \tag{1b}$$

where S_r is the residual wetting fluid saturation, P_d is the displacement pressure for the porous medium expressed as an equivalent height of water [L], and λ is the pore size distribution index. It is important to note that in the Brooks-Corey relationship, there is no distinction between a displacement pressure and an entry pressure (Gerhard and Kueper, 2003). Here, the term displacement pressure and the symbol P_d has been used throughout to be consistent with both the traditional expression of the Brooks-Corey relationship and the nomenclature used by Liu and Dane (1995). For imbibition, the Brooks-Corey equations were modified as (Gerhard and Kueper, 2003):

$$S_w = S_r + (S_w^X - S_r)\left(\frac{P_t}{P_c}\right)^{\lambda} \text{for} P_c \geqslant P_t \tag{2a}$$

$$S_w = S_w^X \text{ for } P_c < P_t \tag{2b}$$

where S_w^X is the maximum wetting fluid saturation at the end of imbibition, and P_t is the terminal capillary pressure [L].