DNAPL pool mobilization in fractured rock

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Abstract The stable DNAPL pool length formed in a rough-walled fracture is a function of the DNAPL density, fracture dip, interfacial tension, fracture aperture, and applied hydraulic gradient. The presented calculation procedure demonstrates that downward groundwater flow will lead to a lower probability of forming DNAPL pools, and shorter equilibrium pool lengths. Hydrostatic and/or upward flowing systems promote the formation of longer pools. Long DNAPL pools are more susceptible to mobilization through increases in hydraulic gradient than shorter pools. In a composite heterogeneous system, the majority of aqueous phase head loss can occur through the DNAPL pool, implying that the critical hydraulic gradient required to mobilize a pool is a function of the distance over which the gradient is measured. The general result is that much lower gradients can mobilize DNAPL pools than predicted by expressions that calculate the gradient over the length of the pool alone. The implication of this work is that it is much easier to mobilize DNAPL pools through activities such as pump testing, well purging, and pump-and-treat than previously thought.

INTRODUCTION

Groundwater contamination by dense non-aqueous phase liquids (DNAPLs) such as chlorinated solvents, PCB oils, creosote, and coal tar is a common occurrence throughout many areas of North America, Europe, and Asia. Upon release to the subsurface, these liquids distribute themselves as disconnected blobs and ganglia of organic liquid referred to as residual, and in connected-phase distributions referred to as pools. As groundwater flows past residual and pooled DNAPL, components will partition from the DNAPL phase to the aqueous phase, giving rise to dissolved plumes of contamination. It is currently believed that the lifespan of residual and pooled DNAPL subject to dissolution into flowing groundwater is of the order of decades to a few hundred years, depending on site-specific flow conditions and the particular chemical composition of the DNAPL. This statement is supported by the fact that chlorinated solvent DNAPLs are still today encountered at sites where releases occurred in the 1950s, and by the fact that coal tar and creosote DNAPLs are encountered at sites where it is known that releases occurred in the late 1800s.

Pooled DNAPL is distinguished from residual DNAPL in that it is potentially mobile. While it is difficult to mobilize residual DNAPL with even aggressive groundwater pumping, pooled DNAPL is relatively easy to mobilize in response to increases in the aqueous phase hydraulic gradient. Such increases in hydraulic gradient can be brought about through purging of monitoring wells prior to sampling, conducting pump tests, dramatic changes in infiltration, the operation of pump-and-treat extraction wells, and water flooding. Each of these activities has the potential to mobilize pooled DNAPL in the downgradient direction. When the
mobilization occurs as part of a DNAPL recovery scheme, it may be deemed beneficial. If the mobilization results in an expansion of the spatial extent of the DNAPL source zone, it may be undesirable.

The purpose of this paper is to present a calculation procedure to predict the relationship between stable DNAPL pool length and hydraulic gradient in rock fractures. The calculation procedure is also applicable to porous media through the use of appropriate input parameters. While expressions to predict the critical gradient required to mobilize pooled DNAPL have appeared elsewhere in the literature (e.g. Cohen & Mercer, 1993), these are posed in terms of the hydraulic gradient across the DNAPL pool alone. Of much more practical importance is the hydraulic gradient measured over a larger distance, between two points on either side of the DNAPL pool. These points can be thought of as piezometers used for water level measurement. In such a composite system, the DNAPL pool can be thought of as a local aquitard, experiencing the majority of the head loss in the system. The result is that the hydraulic gradient required to mobilize a pool is a function of the distance over which the gradient is measured, with the general result that much lower gradients can mobilize DNAPL pools than predicted by the expressions that calculate the gradient over the length of the pool alone.

**THEORY**

The following mathematical development will outline the hydraulic gradient required to mobilize a pool of DNAPL in a one-dimensional, flowing system. A one-dimensional analysis lends itself to an exact analytical solution, and is appropriate in geological settings where the deposits of interest are stratified, and/or the DNAPL pool is constrained on one or more sides such as in rock fractures. The analysis which follows assumes that both the liquids and media of interest are incompressible. It is further assumed that DNAPL is non-wetting with respect to water.

The flux of the non-wetting phase in a two-phase system, \( q_{NW} \), is given by:

\[ q_{NW} = -\frac{kk_{NW}}{\mu_{NW}}\left(\frac{dP_{NW}}{dz} - \rho_{NW} g \sin \alpha\right) \]  

where \( k \) is the intrinsic permeability of the medium containing the pool, \( k_{NW} \) is the relative permeability to the non-wetting phase, \( P_{NW} \) is the non-wetting phase pressure, \( \mu_{NW} \) is the non-wetting phase viscosity, \( \rho_{NW} \) is the non-wetting phase density, \( g \) is gravity, \( z \) is the direction of flow, and \( \alpha \) is the dip below horizontal. In order for the non-wetting flux to be equal to zero, the following must hold:

\[ \frac{dP_{NW}}{dz} = \rho_{NW} g \sin \alpha \]  

If the pressure gradient on the left side of equation (2) is greater than \( \rho_{NW} g \sin \alpha \), the pool will be mobilized in the up-dip direction. If the pressure gradient is less than this value, the pool will be mobilized in the down-dip direction. If the pressure gradient is equal to \( \rho_{NW} g \sin \alpha \), the pool will remain stationary.

The flux of the wetting phase through the pool, \( q_w \), is given by:
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\[ q_w = -\frac{k k_{rw}}{\mu_w} \left( \frac{d P_w}{dz} - \rho_w g \sin \alpha \right) \]  

(3)

where \( P_w \) is the wetting phase pressure, \( \mu_w \) is the wetting phase viscosity, \( \rho_w \) is the wetting phase density, and \( k_{rw} \) is the relative permeability to the wetting phase. In this analysis, the wetting phase relative permeability is represented by (Brooks & Corey, 1966):

\[ k_{rw} = S_e^{(2+3\lambda)/\lambda} \]  

(4)

where \( \lambda \) is a pore size distribution index, and \( S_e \) is an effective wetting phase saturation defined as:

\[ S_e = \frac{(S_w - S_r)}{(1 - S_r)} \]  

(5)

where \( S_w \) is the wetting phase saturation, and \( S_r \) is the residual wetting phase saturation.

Given that the capillary pressure, \( P_C \), is defined as \( P_{nw} - P_w \), it follows that:

\[ \frac{d P_w}{dz} = \rho_{nw} g \sin \alpha - \frac{d P_C}{dz} \]  

(6)

This expression holds when the non-wetting phase is immobile. Equation (3) can now be written as:

\[ q_w = \frac{k k_{rw}}{\mu_w} \left( \frac{d P_C}{dz} - \Delta \rho g \sin \alpha \right) \]  

(7)

where \( \Delta \rho \) is defined as \( \rho_{nw} - \rho_w \). The term in parentheses in equation (7) represents the hydraulic gradient that must be maintained across the pool to prevent mobilization. Evaluated in terms of the minimum hydraulic gradient that must be maintained across the pool, this leads to:

\[ \nabla h = \frac{\Delta \rho \sin \alpha}{\rho_w} - \frac{P_C(L) - P_C(0)}{\rho_w g L} \]  

(8)

where \( \nabla h \) is the hydraulic gradient across the pool defined as the hydraulic head at the base of the pool \((z = L)\) minus the hydraulic head at the top of the pool \((z = 0)\) all divided by the length of the pool \((L)\). \( P_C(L) \) is the entry pressure of the fracture at the base of the pool, and \( P_C(0) \) is the capillary pressure at the top of the pool. It can be seen from equation (8) that if the entry pressure of the fracture at the base of the pool is greater than the capillary pressure that can be generated at that location under hydrostatic conditions, flow in the down-dip direction can be tolerated without bringing about pool mobilization. The maximum hydraulic gradient that can be sustained in the down-dip direction without causing mobilization is related to the length of the DNAPL pool. Any pool at its maximum stable length will be mobilized by any increase in hydraulic gradient. A pool having a length less than its maximum stable length can tolerate an increase in hydraulic gradient without being mobilized.

In order to extend the above analysis to a heterogeneous system, an expression is
required between the wetting phase flux through the pool and the pool height. Equation (7) leads to:

\[ \text{dz} = \frac{dP_c}{\left( \frac{q_w u_w}{kk_w} + \Delta \rho g \sin \alpha \right)} \] (9)

The length of non-wetting phase pool that will be in equilibrium with a given wetting phase flux is obtained through integration of (9) as follows:

\[ L = \int_{P_c(0)}^{P_c(L)} \frac{dP_c}{\left( \frac{q_w u_w}{kk_w} + \Delta \rho g \sin \alpha \right)} \] (10)

The upper limit on this integral is set to the entry pressure of the fracture at the base of the pool to yield the maximum pool length that can be sustained for a given wetting phase flux.

The total head drop that can be sustained across a heterogeneous system can be arrived at by noting that the wetting phase flux is constant within any layer. The total head drop, \( \Delta H \), is given by:

\[ \Delta H = \frac{q_w L_1}{K_1} + \frac{q_w L_2}{K_2} + \frac{q_w L_3}{K_3} + \ldots + \frac{q_w L_n}{K_n} + \left( \frac{\Delta \rho \sin \alpha}{\rho_w} L_p - \frac{\Delta P_c}{\rho_w g} \right) \] (11)

where \( L_i \) is the length of the \( i \)th layer in the system, \( K_i \) is the hydraulic conductivity of the \( i \)th layer in the system, \( \Delta P_c \) is the difference in capillary pressure between the base and top of the pool, and \( L_p \) is the length of the non-wetting phase pool. The non-wetting phase pool can be situated anywhere in the sequence of \( n \) layers comprising the heterogeneous system. The overall hydraulic gradient in the \( n \)-layer system is arrived at by dividing \( \Delta H \) by the total length of the system (\( L_1 + L_2 + L_3 + \ldots + L_n \)).

In the above analysis, the hydraulic conductivity of the fracture, \( K \), can be defined as:

\[ K = \frac{e^2 \rho g}{12 \mu} \] (12)

where \( e \) is the hydraulic aperture of the fracture, \( \rho \) is the density of the groundwater, \( g \) is the acceleration due to gravity, and \( \mu \) is the viscosity of the groundwater. The fracture entry pressure can be calculated as (Kueper & McWhorter, 1991):

\[ P_e = \frac{2 \sigma \cos \theta}{e} \] (13)

where \( P_e \) is the fracture entry pressure, \( \sigma \) is the DNAPL-water interfacial tension, \( \theta \) is the contact angle, and \( e \) is the fracture aperture first giving rise to a continuous non-wetting phase along main drainage. The relationship between fracture entry pressure and hydraulic aperture is discussed by Reitsma & Kueper (1994).
EXAMPLE CALCULATIONS

This example examines the relationship between hydraulic gradient and DNAPL pool length in a single fracture. A three-layer system is adopted as illustrated in Fig. 1. Each of the three portions of the fracture are assigned an aperture and a length. The DNAPL pool is situated in the first portion of the fracture which is assigned a length $L_1$ and an aperture $E_1$. The second portion of the fracture has a smaller aperture and a higher entry pressure than the first portion, thereby providing the capillary resistance necessary to support the DNAPL pool.

Figure 2 illustrates the relationship between hydraulic gradient and DNAPL pool length for a variety of values for $E_2$ in a fracture dipping $20^\circ$ below horizontal. A negative gradient corresponds to flow from right to left (A to B) in Fig. 1. The DNAPL is assigned a density of 1230 kg m$^{-3}$, and an interfacial tension with water of 0.010 N m$^{-1}$. The portions of the fracture are assigned lengths $L_1 = 10$ m, $L_2 = 1$ m, and $L_3 = 10$ m. The total distance between measuring points for determining the hydraulic gradient is therefore 21 m (horizontal distance of 19.73 m). The first and third portions of the fracture are assigned apertures of 138 microns. The figure
shows that the aperture of the middle fracture portion is influential in determining the maximum stable pool length for small hydraulic gradients, but not for larger gradients. It is clear from the figure that longer DNAPL pools are more easily mobilized than shorter pools.

Figure 3 examines the influence of fracture dip on the relationship between hydraulic gradient and DNAPL pool length. The middle portion of the fracture is assigned an aperture (E2) of 50 microns. All other fluid and media properties are identical to those in the previous figure. Figure 3 shows that a shallower dip results in a longer pool length for a specified hydraulic gradient, consistent with the fact that

Fig. 3 Hydraulic gradient versus stable DNAPL pool length for various fracture dips.

Fig. 4 Hydraulic gradient versus stable DNAPL pool length for fracture dip of zero. The figure illustrates the gradient across both the DNAPL pool, and the composite system.
gravity provides less of a mobilizing force in a shallower dipping system. For hydraulic gradients in excess of approximately -0.1, the influence of fracture dip is relatively small, however, indicating that the system is governed by hydraulic forces. As in Fig. 2, note that very small increases in the hydraulic gradient will mobilize pools longer than approximately 0.2 m. It is clear that sites subjected to moderate aqueous phase hydraulic gradients may be relatively void of DNAPL pools.

Figure 4 presents a plot of hydraulic gradient versus stable pool length for a horizontal fracture using the same parameters as in the previous figure. The figure presents both the hydraulic gradient across the DNAPL pool (lower curve), and the gradient across the entire 21 m long system (upper curve). It is clear that the local gradient across the pool is significantly higher than that across the composite system, consistent with the fact that there is a low relative permeability to water within the DNAPL pool. The pool can be though of as an aquitard which experiences most of the head loss in the system. For the system shown, the calculated critical gradient will decrease as the distance between the measuring points (L1 + L2 + L3) increases.

The implications of the above example calculations are that well purging activities, pump testing, and pump-and-treat extraction systems may result in an unwanted expansion of the DNAPL source zone. Consideration should be given to carrying out such activities a specified distance away from suspected DNAPL pools such that the critical mobilization gradients will not be generated. This work also demonstrates that DNAPL pools are more likely to form in systems subject to upward or no-flow conditions. DNAPL pools will be shorter, and less likely to form in systems which have experienced high groundwater flow rates and strong downward components to flow.

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