LNAPL Behavior in Fractured Rock: Implications for Characterization and Remediation

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Abstract

Light non-aqueous phase liquids (LNAPLs) behave very differently in fractured rocks than they do in porous media, and differently again than DNAPLs in fractured rocks. Despite this, relatively little is available in the literature on the specific factors governing LNAPL distribution, migration, and behavior in fractured aquifers. Behavior of LNAPL within a fractured rock mass is a function of the properties of the immiscible fluid, the geometry of the fracture network, rock matrix properties, and the groundwater regime. Relatively small volumes of LNAPL within vertical or sub-vertical fractures can produce significant LNAPL pressure heads, resulting in LNAPL penetration into the saturated zone. Penetration can be significantly deeper than predicted by porous medium models. Groundwater surface fluctuations can cause lateral LNAPL migration in directions controlled by the fracture network geometry, rather than the hydraulic gradient. LNAPL on the groundwater surface can be pumped along intersecting fractures as the groundwater surface rises and falls. Vertical and near-vertical fractures play a key role in LNAPL movement and penetration beneath the groundwater surface, and so should be a focus of investigation and remedial activity. The implications for characterization and remediation are significant, illustrated by case histories from three sites where LNAPL is present in discretely fractured aquifers. Measured thicknesses of LNAPL in wells relate more to the triggering of individual pulse-like flow events, triggered by changes in LNAPL connectivity with the fracture network and changes in the pressure regime, than to equilibria between fluids in the well and the adjacent rock. Accordingly, angled holes can play a key role in detection and collection of LNAPL within near-vertical fractures. The presence of potentially mobile LNAPL well below historical groundwater surface lows should be considered, and groundwater surface fluctuations should be carefully controlled during remediation to avoid triggering lateral migration.

Introduction

Contamination of the subsurface by light non-aqueous phase liquids (LNAPL) is common in industrialized and less-developed countries alike, and is well documented. The majority of the literature describing LNAPL in the subsurface and groundwater focuses on porous media, reflecting the widespread occurrence of motor fuel and other LNAPL contamination in shallow unconsolidated sediments. Increasingly however, important and often vulnerable fractured bedrock aquifers are being affected by LNAPL. The problems associated with NAPLs generally (non-aqueous phase liquids) in fractured rock and aquifers have been reviewed by several authors, including Mackay and Cherry (1989), Mercer and Cohen (1990), and Pankow and Cherry (1996). Concerns over the impacts of chlorinated solvents on groundwater have led to a significant body of work examining DNAPLs in the subsurface, including in fractured rocks (Cohen and Mercer, 1993; Kueper and McWhorter, 1991). More recently, the behavior and problems associated with LNAPLs in fractured aquifers have been studied (Hardisty *et al,* 1998a; Wealthall *et al,* 2002). Nevertheless, conventional site investigation techniques developed for porous media tend to be used when characterizing LNAPL spills in fractured rock environments. This is partly due to a lack of established field methods for the investigation of LNAPL-contaminated fractured systems (Mercer and Cohen, 1990). Accordingly, there is little in the literature describing the economic costs and benefits of various remedial options for LNAPL in fractured rocks (Hardisty and Ozdemiroglu, 2004), or specifically on remediation methods for dealing with LNAPL in fractured systems.

Behavior Of LNAPL in Fractured Rocks

Hardisty *et al*. (1998a) presented a model of LNAPL behavior in fractured rock. LNAPL migrating from the surface through unsaturated fractured rock is described as single phase flow. LNAPL flow through a fracture in the unsaturated zone increases as the viscosity of the fluid decreases and as fracture aperture increases. An LNAPL "plate" of unit width and length L produces a hydrostatic driving pressure head which is a function of the fracture dip angle ψ. The steeper the fracture dip, the greater the head, all other factors remaining equal. Vertical migration of LNAPL from surface through unsaturated fractured rock will occur most effectively in larger aperture vertical or near-vertical fractures. As migration continues, connected vertical height of LNAPL ($L\sin\psi$) increases, and LNAPL pressure heads (h_L) driving flow increase (assuming an active source). Significant lateral migration of LNAPL may occur via dipping fractures in the unsaturated zone.

Once LNAPL reaches the groundwater surface, its accumulated weight will begin to depress the LNAPL-water interface within the fracture. In a water wet system, LNAPL will enter a given fracture only if the LNAPL-water capillary pressure (P_c) at the fracture entrance is greater than the fracture entry pressure P_e. Idealizing the fracture as two parallel plates of aperture b, the fracture entry pressure can be described as a capillary phenomenon, and is expressed as (Kueper and McWhorter,1991)

$$P_e = \frac{2\sigma\cos\phi}{b} \tag{1}$$

where, σ is the interfacial tension between LNAPL and water, and ϕ is the interface contact angle through the wetting phase. At the water table, where the water fluid pressure P_w equals zero, the capillary pressure is equal to the LNAPL fluid pressure at the interface, which is proportional to the connected vertical height of LNAPL within the fracture (h_L). This pressure is balanced by the buoyancy of the LNAPL provided by penetration beneath the groundwater surface, and the entry pressure of the fracture, as (Hardisty *et al.* 1998a):

$$h_L\rho_L g = h_p\rho_w g + (\frac{2\sigma\cos\phi}{b}) \tag{2}$$

where ρ_L and ρ_w are the densities of LNAPL and water, respectively, g is gravity, and h_p is the depth of LNAPL penetration below the groundwater surface, and b is the fracture aperture. Other fractures connected to the LNAPL-bearing fracture beneath the groundwater surface could also be invaded by LNAPL, depending on their aperture and orientation (dip angle). This is in contrast to porous media, where capillary effects tend to dominate and LNAPL is not physically constrained as it is in fractures, preventing significant penetration of LNAPL beneath the groundwater surface.

Penetration of LNAPL to significant depths beneath the groundwater surface may lead to circumstances where LNAPL becomes trapped beneath the groundwater surface. In the case of an LNAPL plate trapped in a fracture below the groundwater surface, upward entry of LNAPL into a water-filled fracture can occur if the LNAPL-water capillary pressure at the upper plate edge (provided by buoyancy) exceeds the entry pressure of the fracture. This condition can be expressed as (Hardisty *et al.* 1998a):

$$h_L = \frac{2\sigma\cos\phi}{|\Delta\rho| gb} \tag{3}$$

where $|\Delta\rho|$ is the absolute value of the difference in density of water and LNAPL. The LNAPL pressure at the upper plate interface is:

$$h_L = L\sin\psi \tag{4}$$

Thus, fracture geometry has a direct effect on the behaviour and occurrence of LNAPL. Trapped LNAPL can be remobilized by a fall in the groundwater surface, reducing the hydrostatic pressure at the plate interface, increasing the LNAPL-water capillary pressure. Introduction of fresh LNAPL, or lateral redistribution of existing LNAPL, can reconnect trapped LNAPL with LNAPL in other sub-vertical or vertical fractures, significantly increasing the effective connected vertical height of LNAPL. This could be provide sufficient impetus to allow LNAPL previously trapped beneath the groundwater surface to flow into an observation well. This model explains the often-observed erratic occurrence of measured LNAPL thicknesses in observation wells completed in fractured rock.

The effects of a fluctuating groundwater surface within fractured rock on the entrapment and migration of LNAPL can be pronounced (Hardisty *et al,* 1994). As the groundwater surface drops, LNAPL within individual fractures follows under the influence of gravity. Depending on the capillary pressure characteristics of the matrix, the LNAPL will follow the groundwater surface most immediately through larger aperture vertical fractures. Lateral migration of LNAPL into newly-unsaturated fractures may then occur via less steeply dipping fractures. If the groundwater surface then rises, LNAPL contained in steeply dipping fractures will be most able to follow, and new water-filled fractures may be entered. In this way, a fluctuating groundwater surface can literally “pump” LNAPL laterally, LNAPL entering new fractures with each cycle of rise and fall. In fractured rocks with high transmissivity and comparatively low storage (such as in fractured crystalline rocks), these effects can be pronounced, driven by large and rapid groundwater surface fluctuations.

Characterization of LNAPL In Fractured Rock

Characterization Methods

Because LNAPL behaves differently in fractured rocks, it may not always be enough to directly apply conventional porous medium site investigation techniques at a fractured rock site. For site investigations in fractured rock, we should anticipate the possibility that LNAPL lies *below* the current groundwater surface, and *up-gradient or cross-gradient* from the spill location. Since LNAPL migration can be triggered by groundwater surface fluctuations, data is needed on the range of water table fluctuations, and aquifer tests should be planned carefully to avoid mobilizing LNAPL (Hardisty et al, 2003). During hydraulic testing, the minimum pumped water level should be maintained above the lowest recorded level for the site. Some of the key parameters to be determined during site characterization are listed in Table 1.

Table 1: Parameters to be determined for characterisation of LNAPL in fractured rock

Category	**Parameters**
LNAPL properties	density, viscosity, interfacial tension
	chemical composition
Fracture Network	location of major fracture sets
	orientation of fracture sets
	fracture aperture and length estimates
	fracture network connectivity
	fracture density
Rock Matrix	permeability, porosity
	capillary pressure characteristics
	LNAPL presence in rock matrix
LNAPL occurrence	fractures containing LNAPL
	depth range of LNAPL
	areal distribution of LNAPL
Hydrogeology	groundwater flow regime
	system transients
	hydraulic parameters (K,T,S)
	groundwater chemistry

A combination of site characterization techniques is needed. Outcrop mapping and remote sensing techniques allow inexpensive data to be collected on fracture network characteristics. Geophysical tools can help to build up a conceptual model of the site. Table 2 provides a list of some of the methods which can be used to collect the data required for characterizing LNAPL in fractured systems (Hardisty et al, 2003).

Table 2. Characterization methods for LNAPL in fractured rock

Method	**Data Provided**
aerial-photo and remote sensing fracture lineament studies; (Rouleau, 1994)	regional fracture trends
fracture mapping at outcrop	local fracture data and statistics
surface geophysical techniques: high resolution seismic, electrical resistance tomography (ERT) (Lane *et al*. 2000)	identification of major vertical fractures
Rock coring (Schmelling and Ross, 1989), angled coring	site fracture data and statistics, (density, orientation, location, character) site geology
lab analysis of rock matrix plug samples from core	rock matrix properties
digital borehole imaging (SIPS) (Hardisty *et al*. 2001); other wellbore geophysical logging;	fracture data and statistics, fracture aperture data; presence of fractures and fracture zones
hydraulic packer testing (Novakowski,1988)	fracture aperture, bulk and fracture conductivity estimates
sleeved coring with field analysis of fluorescence, sponge coring and laboratory analysis (Hardisty *et al*. 1994)	LNAPL occurrence and distribution, identification of LNAPL-bearing fractures, matrix Pc and LNAPL saturation measurements
depth-specific short-screened monitoring wells, aqueous phase sampling and monitoring (Mercer and Cohen, 1990)	mobile LNAPL presence and temporal behavior in individual or groups of fractures; aqueous phase plume position and migration; inferred presence of residual LNAPL
flexible absorbent borehole liners system (Guswa *et al.* 2001)	fracture-specific LNAPL identification
laboratory analysis of LNAPL fluid samples	LNAPL density, viscosity, interfacial tension properties, chemical composition
single hole and multi-well tracer tests (Burton *et al*. 2002)	identification of flowing fractures, network connectivity
LNAPL bail-down tests (Aral and Liao, 2000)	LNAPL flow potential and volume

Interpretation of LNAPL thickness data from monitoring wells

One of the most widely used LNAPL site characterization techniques is the installation of monitoring wells within zones of suspected contamination. In porous media, measured thickness of accumulated LNAPL within monitoring wells are used to infer LNAPL thicknesses within the adjacent formation (Farr *et al.* 1990; and Lenhard and Parker 1990). The limitations of this approach are described in Abdul *et al.* (1989). In fractured media, however, LNAPL flow into the well could be the result of connection with LNAPL plates in fractures at a considerable height above or below the screened interval. Hardisty *et al.* (1998b) discuss the interpretation of LNAPL thicknesses in monitoring wells completed in fractured rock. LNAPL flow to a monitoring well completed in fractured rock (assuming no matrix flow) occurs within fractures which intersect the screened interval. These fractures must furthermore contain LNAPL at fluid pressures sufficient for LNAPL flow. The orientation of the fractures intersecting the borehole, and the network with which they are connected beyond the borehole, also help to control LNAPL flow to the well. Thus LNAPL flow to a monitoring well in fractured rock can be described as a dynamic process. LNAPL flow to the well occurs only when conditions are appropriate, and will then stop when conditions change. Relating observed LNAPL thicknesses in monitoring wells to conditions in the formation, as is done in porous media, is extremely difficult.

Implications for Remediation

Whether dealing with LNAPL in fractured rock, significant challenges exist when contemplating remediation. First, characterization of the distribution and behavior of all NAPLs in fractured rock is notoriously difficult (CL:AIRE, 2002; Pankow and cherry, 1996). Rarely in practice is a complete characterization feasible. Proven techniques for NAPL removal from fractures are few. Pump-and-treat methods, while effective for containment, have proven disappointing for NAPL removal, even when coupled with targeted NAPL recovery pumping and skimming (Schmelling and Ross, 1989). Recently, more aggressive in-situ NAPL-removal methods have been field tested, including high vacuum extraction, thermal heating, and surfactant assisted aquifer remediation (Taylor et al, 2001). These relatively expensive methods have shown good results in some cases, but have not yet been rigorously tested in fractured rock environments.

In general, if the remediation strategy depends on mass removal, efforts should focus on high aperture vertical and sub-vertical fractures where the bulk of the most mobile LNAPL is likely to occur. These fractures are likely to contain the highest LNAPL saturations, and will yield LNAPL more readily than smaller aperture horizontal fractures. High vacuum extraction methods, for instance, might be best applied using angled or horizontal wells, which can be placed to intersect vertical and sub-vertical fractures. When placing wells, LNAPL should be expected both below the historical groundwater surface low, and sometimes significantly up-gradient or cross-gradient of the spill source, given the proper conditions.

If remediation is aimed at containment, the role of seasonal groundwater surface fluctuations should be considered. In low porosity fractured systems, seasonal groundwater surface fluctuations may be considerable, and response to recharge events quick. Under these conditions, groundwater level control is important to prevent further lateral migration of LNAPL, especially in rock with complementary sets of dipping fractures. A sound understanding of the recharge mechanisms and responses governing aquifer behavior will allow changes in pumping rates to be planned and implemented.

Conclusions

LNAPL behavior and occurrence with fractured aquifers depends on the characteristics of the fracture network, LNAPL spill history and configuration, the properties of the LNAPL and the rock matrix, and the hydrogeological regime. LNAPL may penetrate beneath the groundwater surface in vertical or sub-vertical fractures, and may become trapped beneath the groundwater surface in sub-horizontal fractures at saturations sufficient for flow, if accessed by a well screen. Lateral LNAPL migration is governed by the fracture network geometry and the frequency and magnitude of groundwater surface fluctuations, and so LNAPL may migrate up-or cross-gradient from the spill location. The complexity of LNAPL behavior in fractured systems requires that detailed site investigation, sometimes using specialized techniques, be undertaken before considering a remedial design. A characterization program must provide information on the fracture network geometry, the occurrence and distribution of LNAPL within the rock matrix and fractures, and the hydrology of the system which governs LNAPL migration. This will greatly aid in the selection of remediation objectives and techniques.

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