



# Incorporating hysteresis in a multi-phase multi-component NAPL modelling framework; a multi-component LNAPL gasoline example



Kaveh Sookhak Lari<sup>a,b,\*</sup>, Greg B. Davis<sup>a,c</sup>, Colin D. Johnston<sup>a,c</sup>

<sup>a</sup> CSIRO Land and Water, Private Bag No. 5, Wembley WA 6913, Australia

<sup>b</sup> School of Engineering, Edith Cowan University, 270 Joondalup Drive, Joondalup, WA 6027, Australia

<sup>c</sup> School of Earth and Environment, The University of Western Australia, 35 Stirling Highway Crawley, WA 6009, Australia

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## ABSTRACT

The longevity of chemicals in subsurface NAPL releases is a function of their partitioning into different phases. Hysteresis can affect distribution and partitioning of compounds in the vadose zone. We separated and modified hysteresis code from NAPL Simulator (which include hysteresis caused by fluid entrapment and capillary effects) and embedded it into TMVOC. For the first time, the resulting framework is used to model multi-component and multi-phase NAPL release, partitioning and transport. We then applied the verified framework to model effects of hysteresis on partitioning of BTEX, TMB and short and long chain alkanes from a typical gasoline spill. Excluding hysteresis resulted in an expanded LNAPL plume and underestimated the compounds longevity. Hysteresis altered the spatial distribution of LNAPL molar fractions as well as gas flow path and contaminants distribution compared to the non-hysteretic case. The amplifying effect of hysteresis on the longevity of mixtures (and associated risks) should be considered if non-hysteretic relationships are applied.

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## 1. Introduction

Accidental release of non aqueous phase liquids (NAPLs) into the subsurface may occur due to a number of causes including leakage and failure in underground storage tanks and pipelines or direct spills onto the soil during refuelling. Hundreds of chemicals, some with high health risk profiles (e.g. carcinogenic compounds), may be present in typical fuels released (Davis et al., 2009; Sookhak Lari and Safavi, 2008). These compounds, inside a multi-component NAPL plume, partition into gaseous and aqueous phases based on their solubilities and vapour pressures (Davis et al., 2005; Lang et al., 2009; Lekmine et al., 2014). In addition to these chemico-physical properties, the rates of partitioning (volatilisation and dissolution of NAPL compounds) are functions of NAPL/water/air saturation distributions and architecture. In particular for light NAPLs (LNAPLs), the NAPL plume resides in the vadose zone and the capillary fringe with a limited tendency to penetrate below the water table. Therefore, both volatilisation and dissolution are of significance during the life of a LNAPL plume (Sookhak Lari et al., 2016).

The architecture of a LNAPL plume is a function of various parameters including depth, rate and duration of the release, intrinsic soil physical properties (such as permeability) and also multi-phase characteristic curves describing the relationship between capillary pressure of different fluid pairs, relative permeabilities and saturations. Extensive experimental studies have been conducted over the past decades to elucidate multi-phase characteristic curve parameters for various soils and fluids during imbibition and drainage phases of flow history. These parameters (e.g., in the form of the surface tensions for various fluid pairs and the constitutive relationships constants which identify capillary pressure-saturation- permeability (K-S-P) curves) have then been applied in various multi-phase numerical modelling platforms to study hysteretic and non-hysteretic dynamics of LNAPL plumes (Pasha et al., 2014).

The current multi-phase modelling platforms include different levels of hysteretic constitutive relationships in the form of built-in functions. For instance, STOMP (White and Oostrom, 2006) considers entrapment of non-wetting fluids (i.e., gas in aqueous, gas in NAPL, and NAPL in aqueous) with imbibing wetting fluid conditions to be the only hysteretic process for the hysteretic three-phase K-S-P functions. The Mualem (1976) and Burdine (1953) relative permeability functions in STOMP are dependent on the capillary pressure/saturation function type and are applicable to the

\* Corresponding author.

E-mail address: [kaveh.sookhaklari@csiro.au](mailto:kaveh.sookhaklari@csiro.au) (K. Sookhak Lari).

van Genuchten (1980) and Brooks and Corey (1964) functions. NAPL Simulator (Guarnaccia et al., 1997) includes a three phase K-S-P relationship and fluid entrapment of any fluid in the system based on the extension of the two phase hysteretic K-S-P model by Luckner et al. (1989) which has been derived from the van Genuchten and Mualem constitutive relationships (Mualem, 1976; van Genuchten, 1980). The constitutive relationships for K-S-P in NAPL Simulator are presented as two inter-related submodels, S-P (van Genuchten, 1980) and K-S (Mualem, 1976). The user needs to specify a reference fluid pair (e.g., NAPL and water) and the code then determines the S-P relation for the other two pairs (in a three phase system) based on the interfacial tension scaling (Leverett, 1941). Improvements and field application have been the subject of investigations for some time (e.g., Steffy et al., 1997; 1998). Two-phase hysteretic capillary pressure functions were used in TOUGH (Pruess, 2004) in the late 1980s (Niemi and Bodvarsson, 1988) and hysteretic capillary pressure and relative permeability functions were included in iTOUGH2 a decade after (Finsterle et al., 1998). Doughty et al. (2007) then modified the iTOUGH2 hysteretic formulation. Patterson and Falta (2012) incorporated a simple model into TMVOC and TOUGH2-ECO2N to consider hysteretic effects of non-wetting phase fluid entrapment. Their model required adjustment of the characteristic curve parameters to match the real S-P curves and its application was reported for single component NAPLs. The current two-phase hysteresis formulation in TOUGH2 is adapted from the van Genuchten (1980) formulation for the primary drainage and primary imbibition capillary pressure curves. The capillary pressure curves during the simulation follow unique paths to residual saturations which vary with initial or maximum non-wetting saturation achieved during drainage. The relative permeability functions in TOUGH2 also consider hysteretic effects due to the trapped component of the gas phase during imbibition (Doughty, 2013).

The discussion above shows that several options are available for multi-phase hysteretic studies assuming a single component NAPL. The main gap in the available modelling platforms is where the NAPL is a multi-component mixture of several chemicals. This case is of particular importance in environmental and public health risk assessments since different chemicals partition differently into gas and aqueous phases. For instance Sookhak Lari et al. (2016) showed that partitioning of BTEX, TMB and short and long chain alkanes during a fresh gasoline release and transport in the vadose zone can form a complex non-homogeneous LNAPL plume with different compounds partitioning into gaseous and aqueous phases. This is of particular importance since some of the components such as benzene have a high risk profile.

Here, for the first time we present and implement a multi-phase modelling framework capable of considering simultaneous hysteresis (caused by fluid entrapment and capillary effects) and multi-component partitioning of NAPLs in a subsurface system. To do so, we separated, modified and adjusted the hysteretic K-S-P constitutive relationships in NAPL Simulator and embedded the modified version into the built-in constitutive relationships in TMVOC. The modification was then verified versus a recent experimental and numerical study involving one NAPL component (Pasha et al., 2014). The model then was used to evaluate the effects of LNAPL plume architecture changes due to hysteresis on partitioning of seven representative groups for aromatics and alkanes in a typical fresh gasoline spill. TMVOC is tailored to model multi-component NAPL transport and partitioning in subsurface systems. NAPL Simulator's hysteretic K-S-P code is also selected since its comprehensive three phase formulation includes fluid entrapment of any fluid in the system. A brief technical discussion on the application of the NAPL Simulator K-S-P code in TMVOC is presented in the Appendix.

## 2. Governing equations

TMVOC (Pruess and Battistelli, 2002) is an integral-finite-difference simulator for three-phase non-isothermal flow of water, soil gas and a multi-component mixture of volatile organic chemicals (VOCs) in multi-dimensional heterogeneous porous media (Chen et al., 2012; Kererat et al., 2013). The basic Darcy-scale mass balance equations solved by TMVOC can be written in the general form (Pruess and Battistelli, 2002)

$$\frac{d}{dt} \int_{V_n} M^{\kappa} dV_n = \int_{\Gamma_n} \mathbf{F}^{\kappa} \cdot \mathbf{n} d\Gamma_n + \int_{V_n} q^{\kappa} dV_n \quad (1)$$

where  $dV_n$  is an arbitrary subdomain,  $\Gamma_n$  is the surface for the subdomain,  $M^{\kappa}$  is the mass component under study,  $\mathbf{F}$  is the mass flux,  $q^{\kappa}$  is the sink/source term and  $\mathbf{n}$  is the normal vector on the surface element. Here,  $\kappa = 1, \dots, NK$ , where  $NK$  is the number of components in the system. The total advective mass flux is

$$\mathbf{F}^{\kappa} = \sum_{\beta} X_{\beta}^{\kappa} \mathbf{F}_{\beta} \quad (2)$$

where

$$\mathbf{F}_{\beta} = -k \frac{k_{r\beta} \rho_{\beta}}{\mu_{\beta}} (\nabla P_{\beta} - \rho_{\beta} \mathbf{g}) \quad (3)$$

and  $X_{\beta}^{\kappa}$  is the molar fraction of  $\kappa$  in phase  $\beta$ . Here,  $k$  and  $k_{r\beta}$  are absolute and relative permeability (to phase  $\beta$ ),  $\mu$  is the viscosity and  $P$  is the sum of the pressure of the reference phase and the capillary pressure.  $\mathbf{g}$  is also the gravitational acceleration. The feasibility of these governing equations for simulation of Darcy-scale interfacial mass transfer and partitioning has been verified (Sookhak Lari et al., 2015) through the approaches presented for determination of the entrance length scales for neutral (van Reeuwijk and Sookhak Lari, 2012a; Sookhak Lari et al., 2010; 2013) and reactive flows (van Reeuwijk and Sookhak Lari, 2012b; Sookhak Lari et al., 2011).

Details of the K-S-P model in NAPL Simulator are provided in the model manual (Guarnaccia et al., 1997). In general, hysteresis in the K-S-P relationship in NAPL Simulator is assumed to be caused by both fluid entrapment effects and capillary effects. The capillary pressure head function versus saturation (for any pair of phases) is defined as

$$h_{c(f)} = [S_{e(f)}^{-1/m} - 1]^{1/n} \alpha_{(f)}^{-1} \quad (4)$$

where  $f$  indicates the hysteretic level of the characteristic curve (from primary (drainage or imbibition) curves to (third order) secondary scanning curves) and  $\alpha_d < \alpha_{(f)} < \alpha_i$  are the drainage, current flow path and imbibition coefficients of the characteristic curve respectively. Indeed,  $\alpha_{(f)}$  varies during the course of sequential (and incomplete) imbibition and drainage and represents the capillary effects on the K-S-P model. The value for  $\alpha_{(f)}$  is a function of the hysteretic level of the characteristic curve, the wetting phase saturation at the beginning of the flow path (or the reversal point) and the scaled trapped fluids saturation. Table 5.2 in Guarnaccia et al. (1997) summarises different possible equations for  $\alpha_{(f)}$ . It is also mentioned that in order to determine  $\alpha_{(f)}$  through these equations, a blending coefficient is also applied to minimise effects of discontinuous slopes (at reversal points) in S-P curve on the numerical performance of the model.

The curve fitting parameters  $m = 1 - 1/n$  reflect the pore size distribution and  $S_{e(f)}$  is the effective wetting phase saturation

$$S_{e(f)} = \frac{S_w - S_{r(f)}}{S_{s(f)} - S_{r(f)}} \quad (5)$$

where  $S_w$  is the wetting phase saturation and  $S_{s(f)}$  and  $S_{r(f)}$  are the maximum and minimum saturations along the flow path. The hysteretic effects of fluid entrapment on the S-P model are mainly

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