Modeling Hysteresis of Water–NAPL–Air Suction–Saturation Relationship in Porous Media

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INTRODUCTION

Each year, approximately 16,000 chemical spills occur, among which 90% are petroleum products. Understanding of the transport mechanism of such contaminant fluids in a subsurface spill is essential in implementing rational remediation techniques. Non–Aqueous Phase Liquids (NAPLs) as oils do not mix readily with water and form an independent phase in the multiphase fluids system in the ground.

The suction–degree of saturation, \( s–S \) relationship is crucial in predicting the movement of NAPLs in porous media. Extensive research has been carried out in describing two–phase \( s–S \) relationship (e.g., van Genuchten, 1980; Fredlund and Xing, 1994). Parker and Lenhard (1987) attempted to explain the \( k–S \) relationship in the three–phase system by the extension of the model for two–phase system. Some attempts were also made to describe the \( s–S \) relationship in a three–phase system (e.g., Lenhard and Parker, 1987; Eckberg and Sunada, 1984). Nakamura and Kikumoto (2014, 2018) proposed a rational model for multi-phase fluids incorporating transition among arbitrary two– and three–phase systems.

Due to a change in the interfacial tension, the fluids exhibit a behavior wherein there is a delay in the variation of saturation, i.e., the \( s–S \) relationship does not remain the same during wetting and drying processes contributing to the phenomenon of hysteresis. While there were numerous attempts (Kaluarachchi and Parker, 1987; Huang et al., 2005; Sharma and Mohamed, 2003; and others) to explain hysteresis in a two–phase system, they could not be extended to a three–phase system. Nevertheless, Lenhard (1992) proposed an \( s–S \) model for the hysteresis in a three–phase system, which did not incorporate the transition among arbitrary two– and three–phase systems. Hence, in this paper, a rational concept for a hysteretic \( s–S \) relationship in a two– and three–phase system is proposed and is applied to the model proposed by Nakamura and Kikumoto (2014, 2018).

OVERVIEW OF THE EXISTING SUCTION–SATURATION MODELS

2.1 Models for two–phase system

Matric suction between non–wetting fluid \( i \) and wetting fluid \( j \) is given as the difference between the pore pressures, \( p_i \) and \( p_j \):

\[
s_{ij} = p_i - p_j
\]  

where \( i \) and \( j \) are \( a, o \), or \( w \) representing air, oil (NAPL), or water phases, respectively, in the increasing order of their wettability. Saturation degree, \( S^j_i \), of the wetting fluid \( j \) in \( i–j \) (air–water or NAPL–water) phase system is given by a
Table 1. State parameter, $\mu$, phase system, and scaling function, $\beta_i$ for arbitrary 2– and 3–phase systems

| Parameter $\mu$ | Pressures $p_w$, $p_o$, $p_a$ | Phase system | Saturations $S_{aw}^w$, $S_{aw}^l$ | Scaling functions $\beta_w^w$, $\beta_l^l$ |
|----------------|------------------|-------------|-------------------------------|------------------|
| $\mu = 0$     | $p_w = p_o < p_a$ | Air–Water   | $S_{aw}^w = S_{aw}^l = S_{aw}^w$ | $\beta_w^w = 1$ |
| $\mu = 1$     | $p_w < p_o = p_a$ | NAPL–Water  | $S_{aw}^w = 1$                | $\beta_w^w = \beta_{ow} = \sigma_{ow}/\sigma_{ow}$ |
| $0 < \mu < 1$ | $p_w < p_o < p_a$ | Air–NAPL–Water | $S_{aw}^w < S_{aw}^l$       | $\beta_w^w(\mu) \geq \beta_l(\mu)$ (Bezier curves) |

A straightforward extension of van Genuchten's empirical relationship:

$$S_{ij} = S(\beta_{ij} S_{ij})$$

$$S_{ij} = S_{min} + (S_{max} - S_{min})\left[1 + (\alpha \beta_{ij} S_{ij})^n\right]^{-m} (2)$$

where $S_{min}$ and $S_{max}$ are minimum and maximum degrees of saturation, respectively. $\alpha$, $n$, and $m$ are material parameters, wherein $m = 1–1/n$. $\beta_{ij}$ is a scaling parameter defined as the ratio of interfacial tension between air and water, $\sigma_{ow}$, to that between the focused two–phase fluids, $\sigma_{ij}$.

2.2 Models for three–phase system

Leveret (1941) hypothesized that a void fluid with higher wettability tends to reside near the contact point of soil particles. Based on this assumption, Parker and Lenhard (1987) estimated saturation degrees, $S_{aw}$, for water–NAPL–air three–phase system as:

$$S_{aw}^w = S(\beta_{aw} S_{aw})$$

$$S_{aw}^l = S(\beta_{al} S_{aw})$$

where superscript $l$ represents the liquid phase. Saturation degrees of oil and air are derived as:

$$S_{ao}^w = S_{aw}^w - S_{aw}^l$$

$$S_{ao}^l = 1 - S_{aw}^l$$

$$(4)$$

However, this approach cannot incorporate the transition from a 2–phase to 3–phase system and vice–versa. Meanwhile, Nakamura and Kikumoto (2014, 2018) proposed a novel approach with the transition between arbitrary 2– and 3–phase systems through a new state parameter, $\mu$:

$$\mu = \frac{p_o - p_w}{p_a - p_w} = \frac{S_{ow}}{S_{sw}}$$

A systematically–summarized $\mu$ in the arbitrary 2– and 3–phase systems is shown in Table 1. Using the scaling functions, $\beta_i^i(\mu)$ ($i = w$, $l$), of the state parameter, $\mu$, given by quadratic Bezier curves shown in Fig. 1, the suction–saturation relationships in equation (3) were replaced as:

$$S_{aw}^w = S(\beta_w^w(\mu) S_{aw})$$

$$S_{aw}^l = S(\beta_l^l(\mu) S_{aw})$$

$$(6)$$

This model successfully predicted arbitrary 2– and 3–phase suction–saturation relationship under the monotonic suction regime. However, hysteretic change under cyclic variation in suction has not been incorporated.

3. MODELLING OF HYSTERESIS IN MULTIPHASE SYSTEM

3.1 Effective suction incorporating hysteresis

Hysteresis in three–phase suction–saturation relationship is primarily controlled by contact angle dynamics of menisci of liquids in the porous media. Hysteresis causes delayed variation in saturation degree. This phenomenon can be relatively easily modeled by a delay in the application of matric suction. We newly introduce an effective suction, $s'(s_{ao}, s_{ow}, s_{aw})$, which incorporates delayed application of matric suction. $s'$ describes saturation degrees uniquely instead of the ordinary matric suction. Replacing matric suction in equations (5) and (6) with the corresponding components of $s'$, we obtain:

$$\mu = \frac{s_{ow}'}{s_{sw}'}$$

$$(7)$$

$$S_{aw}^w = S(\beta_w^w(\mu) s_{aw}')$$

$$S_{aw}^l = S(\beta_l^l(\mu) s_{aw}')$$

$$(8)$$

3.2 Evolution of effective suction

Effective suction incorporates the delayed variation in saturation degree by chasing matric suction in the saturation $s_{ao} - s_{ow} - s_{aw}$ space. As matric suction, $s(s_{ao}, s_{ow}, s_{aw})$ always holds the following equality by its definition:
Effective suction $s'$ in the suction $s$ space

Fig. 2

Direction of incremental effective suction, $ds'$ with respect to the incremental matric suction, $ds$

$$s_{aw} = s_{ao} + s_{ow}$$

$s$ always lies on a linear plane in $s_{ao}$-$s_{ow}$-$s_{aw}$ space as shown in the Fig. 2. Effective suction, $s' (s'_{ao}, s'_{ow}, s'_{aw})$, is assumed to stay on or inside a closed domain around matric suction, $s$, on the same plane ($s'_{aw} = s'_{ao} + s'_{ow}$). The domain is named as “hysteresis region”; which is assumed to be a circle centered at suction, $s$, with a radius, $r$, as shown in the Fig. 2. Hence,

$$|s' - s| \leq r$$

$s'$ is assumed to vary in the same direction as $s$ with some delay. Thus, incremental effective suction, $ds'$, must hold Eqs. (11) and (12).

$$ds' \cdot ds \geq 0$$

$$|ds'| \leq |ds|$$

The equations (10), (11), and (12) form the basic requirements for the evolution of effective suction.

Direction of incremental effective suction

To satisfy the inequality (11), $ds'$ is directed towards the edge of the hysteresis region along with $ds$ as shown in Fig. 3. As the edge is given by $s + r \frac{ds}{|ds|}$, the direction of incremental effective suction, $ds'$, is given as equation (13).

$$\frac{ds'}{|ds'|} = \frac{s + r \frac{ds}{|ds|} - s'}{s + r \frac{ds}{|ds|} - s'}$$

Magnitude of incremental effective suction

The magnitude of incremental effective suction, $|ds'|$, in Eq. (13) must be given to describe the evolution of effective suction. The inequality (12) on the delayed variation of effective suction causing is rewritten using a multiplier, $k$, as:

$$|ds'| = k|ds|$$

where the range of $k$ must be $0 \leq k \leq 1$. The two necessary conditions for the $k$ value are: (a) $k$ must attain 1 when $s'$ stays on the boundary opposite to the direction of the $s$ variation ($s$ tries to move away from $s'$) so that $s'$ can remain inside the hysteresis region. (b) In other cases, $k$ must be less than 1 to manifest a delayed effect of suction.

This is incorporated by an ‘$l$’ defined as the distance from $s'$ to the boundary of the hysteresis domain in the direction of $ds$, as shown in Fig. 4. For simplicity, $k$ is modeled to monotonically increase from 0 to 1 with an increase in the distance $l$. This follows that, when $l$ becomes its maximum length, $L$, $k$ reaches unity to satisfy the requirement (b). This monotonic increase is given by the ratio of $l$ and $L$ as follows.

$$k = \frac{l}{L}$$

This is represented in the model rationally by the resulting contours of the same magnitude in the hysteresis domain that forms equipotential lines
with the flow line ds as shown in Fig. 5. From Eqs. (13) and (15), incremental effective suction finally becomes as follows.

\[
ds' = \frac{L}{ds} \left[ \frac{s + r \frac{ds}{ds} - s'}{s + r \frac{ds}{ds} - s'} \right]
\]

(16)

The evolution of effective suction is explained in Fig. 6 (i), which depicts the evolution of modified suction with a monotonic change in suction, s, from the state (a) to (d) on the plane given by Eq. (9). The tracking of effective suction, s', towards matric suction, s, enables the description of a delayed variation in s'. Fig. 6 (ii) shows the relative movement of modified suction, s', with respect to matric suction, s, from the state (a) to state (d). s' gradually moves to the boundary of the hysteresis region opposite to the movement of matric suction, ds. The paths of the relative movements of s' inside the hysteresis region for its different initial positions is shown in Fig. 7 (iii), which describes that the effective suction is always evolved to cause a delay by reaching state (d) eventually.

4 RESULTS AND DISCUSSIONS

The validity of the proposed concept is hereafter discussed through three series of simulations. A unified set of material parameters listed in Table 2 is applied throughout this paper.

First, a possible range of saturation degrees modeled by the proposed concept of effective suction is compared with that predicted by the ordinary model based on matric suction. Two kinds of pressure paths summarized in Table 3 are applied to get the saturation degrees of air, NAPL, and water, \(S_{aw}^{a}, S_{aw}^{b}, S_{aw}^{c}\) in a triangular coordinate system in Fig. 7. At any point, three lines that are drawn parallel to the respective sides of the triangle intersect them at the corresponding fluid saturation on those axes. The possible range of saturation degrees considered in the proposed concept is obtained by assigning effective suction to Eq. (8), which indicates that the hysteretic variation of saturation degree is incorporated by the non-uniqueness of the saturation degree in the hysteresis region. Meanwhile, a dashed-dotted line represents a unique variation in saturation degrees predicted by the ordinary model based on the matric suction only, which indicates a unique variation in the saturation degrees regardless of past suction histories.

Second, the proposed model is verified with the experimental results of the air–water two–phase soil–water characteristic curve (SWCC) obtained from Huang et al. (2005). The wetting and drying scanning paths in a non–monotonous suction regime as summarized in Table 3 capturing the hysteresis loop in white silica sand...
Fig 7. Triangular figure for the saturation degrees of water, NAPL, and air representing the range of saturation degree in (i) NAPL–water two–phase system, and (ii) varying NAPL pressure three–phase system.

Fig 8. Validation of current model with Huang et al. (2005) depicting SWCC with the presence of a hysteretic loop in the scanning wetting and drying paths is shown in the Fig. 8.

Finally, the realistic behavior of soil is modeled by varying the level of NAPL, which is settled above the groundwater after an oil spill. Vertical soil profiles are modeled by initially increasing the NAPL level from step (a) to step (c), followed by decreasing it until step (e). The corresponding wetting and drying paths are obtained during the increase and decrease of the NAPL level, respectively. Fig. 9 represents the model without considering the existence or movement of the effective suction, whereas Fig. 10 represents the saturation curves considering the phenomenon of hysteresis. It is illustrated that, due to hysteresis, NAPL finds it difficult to wet the soil media during the wetting process and leave the soil media during the drying process.

5 CONCLUSIONS

The limitations of the existing models were briefly discussed, and a novel attempt of parametric study to model both a two–phase and three–phase system in a simple yet effective way is achieved. The primary issue of capturing the phenomenon of hysteresis in three–phase system in a simplistic fashion is carried out along with the transition between arbitrary two– and three–phase systems. Nevertheless, the model itself is not restricted only to the conditions involving hysteresis. In addition to this, an innovative and modest approach to represent the saturation state in a single point along with a corresponding hysteretic yield surface in a triangular figure is accomplished. One of the most essential advantages of the proposed model includes the applicability of it in the other two– and three–phase system models.

However, the mechanism of hysteresis is to be investigated to appropriately implement the magnitude of this effective suction range depending on the soil and fluid types. Apart from the fore–mentioned limitations, if there are non–negligible volume changes during the wetting and drying scanning paths, soil deformation and phase–change effects must be taken into consideration as a part of future research to develop the model further.
Fig 9. Vertical distributions of fluid saturations in a three–phase system predicted by the ordinary model based on matric suction.

Fig 10. Vertical distributions of fluid saturations in a three–phase system predicted by the proposed concept based on the effective suction considering the effect of hysteresis.

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