On the Relationship between Water Flux and Hydraulic Gradient for Clay Media

Hui-Hai Liu and Jens Birkholzer

Earth Sciences Division
Lawrence Berkeley National Laboratory
Berkeley, CA 94706

Submitted to
Journal of Hydrology

* Corresponding author; hhliu@lbl.gov; Phone: (510)486-6452; Fax: (510)486-5686

Abstract

Experimental results indicate that traditional form of Darcy's law is not adequate for describing water flow processes in clay media because the observed relationship between water flux and hydraulic gradient can be highly non-linear. To capture this non-Darcian flow behavior, we proposed a new relationship between water flux and hydraulic gradient by generalizing the currently existing relationships. The new relationship is shown to be consistent with experimental observations for both saturated and unsaturated conditions. In this paper, we also developed an empirical relationship between permeability and threshold hydraulic gradient, an important measure of non-Darcian behavior. The latter relationship is practically useful because it can reduce the number of parameters whose values need to be determined from experiment data in order to model non-Darcian behavior. However, how to incorporate impacts of temperature and electrolyte concentrations into the proposed relationships needs further research.

1. Introduction

Water flow in clay media is an important process for a number of practical applications. For example, clay/shale formations have been considered as potential host rock for geological disposal of high-level radioactive waste because of their low permeability, low diffusion coefficient, high retention capacity for radionuclide, and capability to self-seal fractures (Tsang et al., 2012). Clay and shale formations also serve as the cap rock of geological formations where supercritical CO_2 is stored for the purpose of CO_2 sequestration. Water flow through the cap rock may be an important mechanism that needs to be considered for managing pressure buildup owing to injection of CO_2 into the storage formation below the cap rock (Zhou et al., 2008). Nowadays, exploration and recovery of non-conventional energy resources, including shale oil and shale gas, become increasing important. The recovery of these resources from shale formations requires improved understanding of and modeling approaches for water (and other fluid) flow within these formations under different conditions.

Water flow in porous media is traditionally described by Darcy’s law. In 1856, Henry Darcy investigated the flow of water in vertical homogeneous sand filters in connection with the fountains of the city of Dijion in France. From his experiment results, Darcy discovered that water flux is directly proportional to the hydraulic gradient. However, it has been well documented that Darcy’s law is not adequate for modeling water flow in clay media. For example, Hansbo (1960; 2001) reported that water flux is proportional to a power function of the hydraulic gradient when the gradient is less than a critical value, whereupon the relationship between water flux and gradient becomes linear for large gradient values. He explained this behavior by positing that a certain hydraulic gradient is required to overcome the maximum binding energy of mobile pore water. From their experiment results, Miller and Low (1963) also found the existence of a hydraulic gradient below which water is essentially immobile. After analyzing several data sets for water flow in clay media, Swartzendruber (1963) proposed a modified Darcy’s law in which non-linear behavior of water flux vs. gradient is described by an exponential function. Zou (1996) developed a nonlinear flux-gradient relationship depending on the activation energy of pore liquid. He assumed that the activation energy of pore water in clay is not only variable with the distance from the solid particle surface, but also with the flow velocity of pore water. His model, including several empirical parameters, was able to fit a number of data sets that show nonlinear flux-gradient relationships at low hydraulic gradients and linear relationships at high gradients.

The studies mentioned above are all for saturated flow conditions. Laboratory test results seem to show that non-Darcian flow behavior becomes more significant under unsaturated conditions. Cui et al. (2008) reported non-Darcian behavior for a range of observed hydraulic gradients under unsaturated conditions. Liu et al. (2012) developed a constitutive model for unsaturated flow when water can be considered as a power-law (non-Newtonian) fluid. The model is consistent with the data set of Cui et al. (2008).

While considerable progress has been made for investigating non-Darcian flow in clay media, a general relationship between water flux and hydraulic gradient (that covers full range of non-Darcian flow behavior under both saturated and unsaturated conditions) is still lacking. We will develop such a model in this paper and also demonstrate an empirical relationship between permeability and threshold hydraulic gradient (that will be defined later and is an important measure of non-Darcian behavior). The latter relationship is practically useful because it can reduce the number of parameters whose values need to be determined from experiment data in order to model non-Darcian behavior.

2. Relationships between water flux and hydraulic gradient

The key requirement for accurately modeling water flow in clay media is an appropriate relationship between water flux and hydraulic gradient. This section reviews the currently available relationships and develops an improved one that can capture a relatively large range of water flow behavior.

The well-known flux-gradient relationship is Darcy's law given by

$$q = Ki \tag{1}$$

where q (m/s) is water flux, K (m/s) is hydraulic conductivity and i (-) is hydraulic gradient. The above equation only includes magnitudes of variables for one-dimensional flow and therefore q, K, and i are all positive. The similar treatment, for the convenience, is used for all the other relationships to be discussed in this section.

Darcy's law was initially developed for water flow in saturated porous media. Buckingham (1907) extended Darcy's law to unsaturated conditions, although it is an issue of debate regarding whether he was aware of Darcy's law when developing his relationship. An excellent historic review of Edgar Buckingham and his scientific contributions to unsaturated flow in soils was recently published by Nimmo and Landa (2005).

In his extension, Buckingham (1907) used an unsaturated hydraulic conductivity, a function of water saturation, to replace hydraulic conductivity in Darcy's law. Although not explicitly stated in Buckingham (1907), this development is based on the local equilibrium assumption (LEA) that capillary pressure is uniformly distributed within the pore space at the local scale corresponding to representative elementary volume (REV). The LEA essentially implies that water distribution is completely controlled by capillary force and is independent of its flux. That is why capillary pressure and relative permeability can be expressed as functions of saturations only.

However, in many cases, the LEA does not hold especially when instability (or fingering) occurs. For example, if a numerical grid block (and/or a large-scale REV) contains a number of fingers (which is generally the case because a grid-block size cannot be too small for practical applications), LEA is violated at the grid block scale. Liu (2011) recently proposed a new theory for unsaturated flow to deal with this challenging issue based on a optimality principle that unstable liquid flow pattern are self-organized in such a way that total flow resistance is minimal. In this case, the relative permeability is a function of not only saturations, but also water flux. While detailed discussion of the optimality principle is beyond the scope of this study, the principle is closely related to an important feature of a chaotic (complex) system: adaptation. The latter refers to that emergent patterns (fingering for unsaturated) resulting from self-organization are most efficient for certain processes [XXX]. Nevertheless, it is appropriate to apply the LEA to unsaturated flow in clay media because it is strongly controlled by capillary force and not subject to gravitational instability.

Hansbo (1960, 2001) proposed a relationship between water flux and hydraulic gradient to consider the non-Darcian flow behavior in clay media:

$$q = k^* i^n \qquad \text{for } i \le i_1 \qquad (2\text{-}1)$$

$$q = k^* n i_1^{n-1}(i - I) \qquad \text{for } i \ge i_1 \qquad (2\text{-}2)$$

$$i_1 = \frac{In}{(n-1)} \qquad (2\text{-}3)$$

The formulation of Hansbo (1960, 2001) includes three parameters k* (m/s), n (-) and I (-). Note that k* herein is not the hydraulic conductivity and Eq. (2-2) corresponds to a linear function between water flux and hydraulic gradient i. Parameter I is called threshold gradient in this study and refers to the intersection between the i axis and the linear part of the relationship (Figure 1). Hansbo (1960, 2001) demonstrated that Eq. (2) can fit related experimental observations and developed, based on (2), a theoretical approach to dealing with clay consolidation processes. However, as indicated by Swartzendruber (1961), Eq. (2) consists of two separated mathematical expressions and three related parameters cannot be evaluated unless data are available all the way from i =0 out to and including an appreciable part of the linear portion of the flux-gradient curve.

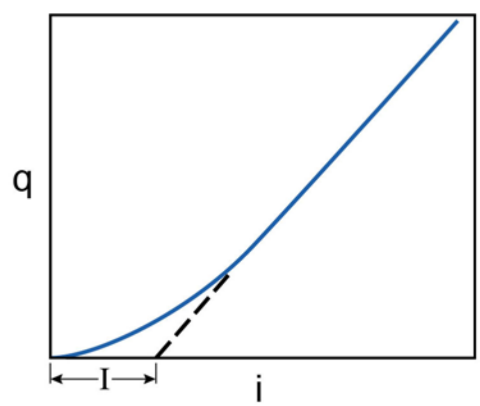

Figure 1. Definition of threshold hydraulic gradient

To overcome the difficulties encountered in applying (2), Swartzendruber (1961) introduced a new version of the modified Darcy's law based on a relation for dq/di:

$$\frac{dq}{di} = K(1 - e^{-i/I}) \qquad (3)$$

For a large value of hydraulic gradient i, dq/di approaches a constant K that is hydraulic conductivity. Integrating, and using q=0 at i=0, leads to

$$q = K[i - I(1 - e^{-i/I})] \qquad (4)$$

Eq. (4) involves two parameters K and I. Compared with commonly used Darcy's law, it contains one additional parameter (I) only. The equation of Swartzendruber (1961) had been evaluated with a number of data sets collected under saturated flow conditions and satisfactory agreements were generally obtained (Swartzendruber1961; Blecker 1970). However, Figure 2 shows a comparison between results calculated from (4) and a data set collected by Cui et al. (2008) under unsaturated flow conditions, indicating the deviation between the theoretical and observed results. Thus, Eq. (4) cannot capture all the range of non-Darcian flow behavior in clay media under different conditions.

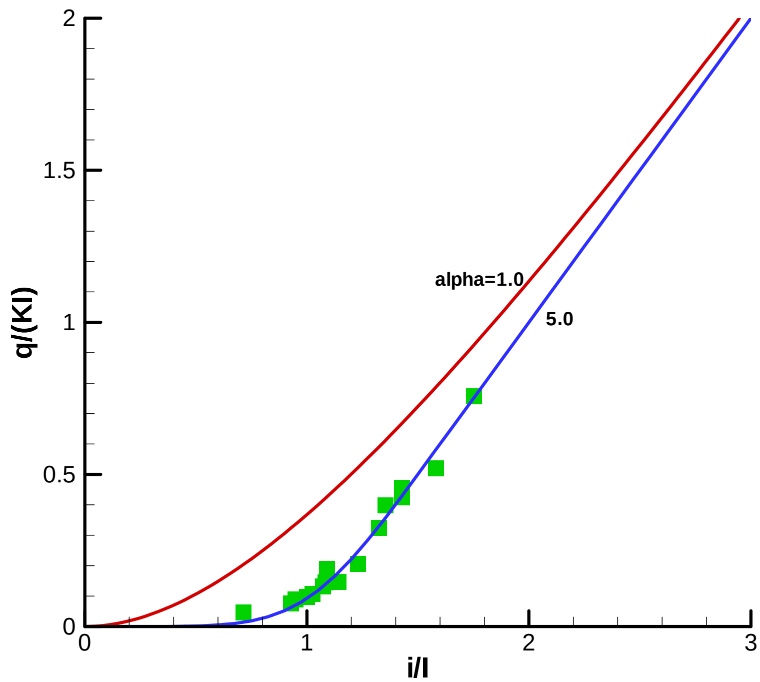

Figure 2. Comparisons between Eq. (8) with two different α values and data of Cui et al. (2008). Note that Eq. (8) is reduced to (4) for α =1.

Another commonly used flux-gradient relationship for clay is given as (e.g., Bear, 1979):

$$q = 0 \qquad \text{for } i \le I \qquad (5\text{-}1)$$

$$q = K(i - I) \qquad \text{for } i \ge I \qquad (5\text{-}2)$$

Similar to (4), the above equation involves two parameters (K and I) only and is mathematically simpler than other relationships. It is obvious, however, from data available in the literature, that (5) cannot adequately capture the non-Darcian flow behavior (or non-linear flux-gradient relationship) at low i values (e.g., Swartzendruber1961; Blecker 1970). Therefore, (5) should be applied only when i is large. It is also of interest to note that (5-2) is a limiting case of (4-2) for $i/I \to \infty$.

Assuming that the activation energy of pore water in clay is not only variable with the distance from the solid particle surface, but also with the flow velocity of pore water, Zou (1996) proposed the following flux-gradient relationship:

$$q = \frac{Ki}{1 + b\exp(-ci)} \tag{6}$$

where b and c are empirical parameters. One problem with this equation is that it will become exactly the Darcy's law, defined in (1), for $i \to \infty$ without considering the threshold gradient that has been often observed from test results, as shown in Figure 2.

From the above discussions, it is clear that a general relationship between water flux and hydraulic gradient (that covers full range of non-Darcian flow behavior under both saturated and unsaturated conditions) is still lacking. As an effort to overcome this problem, we propose to generalize the Swartzendruber's (1961) relationship by using

$$\frac{dq}{di} = K(1 - e^{-\left(\frac{i}{I^*}\right)^{\alpha}}) \tag{7}$$

where α is a positive constant, and I* is a parameter related to α and I. For α =1, (7) is reduced to (3). For $\alpha \to \infty$, $\frac{dq}{di} \to 0$ when $\frac{i}{I^*} < 1$, and $\frac{dq}{di} \to K$ when $\frac{i}{I^*} > 1$. In this case, (7) essentially represents flux-gradient behavior given in (5). Thus, with one more parameter (α), (7) can capture a relatively large range of non-Darcian flow behavior.

Integrating (7) with the condition of q=0 at i=0 yields

$$q = K[i - \frac{I}{\gamma(\frac{1}{\alpha})}\gamma(\frac{1}{\alpha}, \left(\frac{i}{I^*}\right)^{\alpha})] \tag{8}$$

where

$$I = \frac{I*}{\alpha}\gamma\left(\frac{1}{\alpha}\right) \tag{9-1}$$

and γ refers to Gamma functions

$$\gamma(a,x) = \int_0^x t^{a-1}e^{-t}dt \tag{9-2}$$

$$\gamma(a) = \int_0^\infty t^{a-1}e^{-t}dt \tag{9-3}$$

Figure 2 shows a good agreement between Eq. (8) with α = 5 and the data from Cui et al. (2008). Note that Cui et al. (2008) reported flux-gradient data for three capillary pressure values. Each capillary pressure has its own I value. Different I values are used in Figure 2 for different capillary pressure when calculating i/I and dimensionless flux defined as q/(KI).

3. Correlation between permeability and threshold gradient

Previous studies suggest that non-Darcian flow is a result of strong solid-water interaction in clay materials. Since pore size in clay media is on the order of nano-meters, studies on water properties and flow processes in nano tubes are highly relevant to water flow in clay (Wang et al. 2011). The mechanism of non-Darcian flow behavior was attributed to water-clay interaction by Low (1961). This is further confirmed by a number of studies on nanoscale fluid transport based on MD simulations (e.g., Chen et al., 2008; Ma et al., 2010; Farrow et al., 2011). They generally show that water properties and flow processes at that scale could be significantly different from those in course materials. For example, water density near the solid surface is generally much higher than its bulk value. It has been widely accepted that owing to viscose force, water velocity is zero on the solid surface. This non-slippage boundary condition, however, does not apply to nanoscale flow any more because of the sub-continuum feature of water molecules at a nanoscale. Also, resistance to water flow at nanoscale is dominantly from friction between water and solid surface, compared to internal friction among water layers, resulting from small pore sizes (Chen et al., 2008). Several MD simulation results show that flow rate for water flow through a nano-tube is a nonlinear function of shearing stress (that is equivalent to hydraulic gradient for steady-state flow) (e.g., Chen et al., 2008; Ma et al., 2010; Farrow et al., 2011), which is consistent with non-Darcian behavior observed from laboratory measurements.

It is easy to understand that for a clay material degrees of solid-water interaction and non-Darcian flow behavior can be characterized by permeability (or pore size) and threshold gradient (I), respectively. Note that I = 0 corresponds to Darcian law for flux-gradient relationships given in (2), (4), (5) and (8). Thus, it is logical that there exists a correlation between I and permeability. We will demonstrate such a relationship using a number of typical clay data sets of flux vs. gradient (Figure 3). For the transparency purpose, the data sets and procedures to determine parameter I and permeability are briefly given in the rest of this section, while we refer the readers to the cited references for the details of the data sets. This correlation is important because the number of parameters (whose values are required for practical applications) can be reduced.

Lutz and Kemper (1958) are among the first to report impact of clay-water interaction on clay hydraulic properties. Types of clay soils used in their experiments include Utah bentonite, hallosite, Bladen clay, and Wyoming bentonite. The tests were conducted under saturated conditions using solutions with different electrolyte concentrations. A low concentration generally corresponds to a low water flow rate, as a result of swelling, for the given soil and hydraulic gradient. During their tests, water flow rate was monitored as a function of the gradient. The threshold values for all these tests were calculated by Swartzendruber (1961) and used in Figure 3. Swartzendruber (1961) also demonstrated that the data are consistent with Eq. (4). Cross-sectional area of soil samples is needed to determine permeability values from the flow rate vs. gradient data at large gradients corresponding to Darcy behavior. However, Lutz and Kemper (1958) did not report value of the area, but the thickness of soil samples. Thus, we determine the area value from the ratio of sample diameter to its thickness value measured from the test-setup figure in Lutz and Kemper (1958). Since this approximation may involve significant uncertainties, the data of Lutz and Kemper (1958) are not used for estimating relationship between parameter I and permeability in Figure 3, but just for providing corroborative information. Nevertheless, the data seems to be consistent with the general trend in the figure. Also note that impact of soil swelling for this and other data sets to be discussed, to some extent, has been included in permeability values because permeability is a strong function of swelling (or shrinkage).

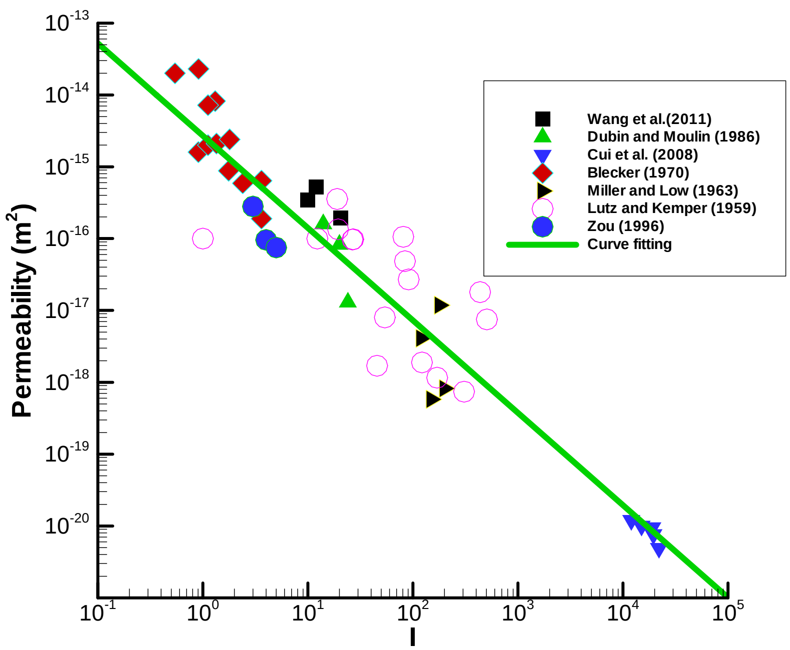

Figure 3. Correlation between permeability and threshold hydraulic gradient.

Miller and Low (1963) presented results of laboratory experiments for Wyoming bentonite. The soil samples have a length of 2.5 cm and diameter of 1 cm. Rates for water flow through soil samples were measured as a function of hydraulic gradient. We estimated permeability values using slope of linear part of the flow-rate vs. gradient curve. When the data points corresponding to the linear part are limited, we use the straight line through the two data points with the highest gradient values to approximate the linear part. Note that Miller and Low (1963) defined their threshold gradient as the gradient below which water does not flow, while in this study we refers the threshold gradient I, as previously indicated, to the intersection between the gradient axis and the linear part of the flux-gradient relationship (Figure 1). Figure 4 shows a match of Eq. (8) with data from one of the experiments conducted by Miller and Low (1963). The fitted α value is 0.9, indicating that the relationship of Swartzendruber's (1961), corresponding to α = 1, should give a similar match.

Blecker (1970) reported test results of flux-gradient relationships for clay soils from Brolliar and Springerville soil series of northern Arizona, USA. His experimental setup is similar to those used by Miller and Low (1963) and Lutz and Kemper (1958). The soil samples were prepared for different densities for a given soil type. His data show a good agreement with Eq. (4). He also reported values for all the fitting parameters including permeability and I. However, his reported values for hydraulic head difference across a soil sample are based on heights of mercury column. To be consistent with other data sets in Figure 3, we converted his reported values by representing head difference with height of water column.

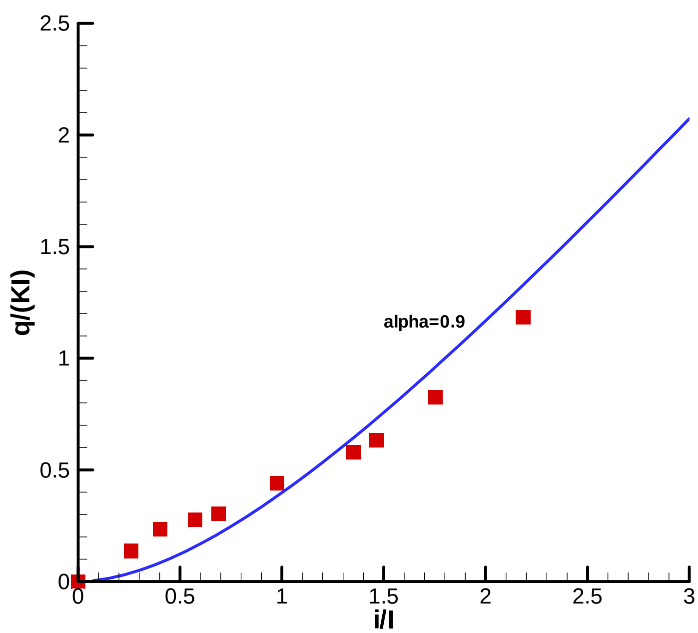

Figure 4. Match of Eq. (8) with test data for a Na-clay paste (Miller and Low, 1963).

Consolidation of clays is important for some civil-engineering applications. Investigation of the consolidation requires data of clay permeability under different mechanical and hydraulic conditions (Hansbo, 2001). For this purpose, Dubin and Moulin (1986) reported measured flux-gradient curves for Saint Herblain clay (their Figure 7) and the corresponding hydraulic conductivity values that are converted to permeability values in Figure 3. To evaluate his relationship for water flux and hydraulic gradient, Zou (1996) also presented a data set for several soil types. Figure 3 only shows his data points for clay soil because no significant non-Darcian behavior is observed from other soils in his paper.

Wang et al. (2011) presented test data from core samples collected from low-permeability Daqing oilfield, China. During their tests, core samples are first saturated with salt water (composition is not available), and then pressure gradient across them are imposed. After steady-state flow condition was achieved, the water flux value was recorded. In this way, they were able generate flow rate data as a function of hydraulic gradient. The non-Darcian flow behavior is represented by a ratio of $(\frac{q}{i})/K$ (their Figure 2) in terms of variables defined in this paper. The permeability values reported in Wang et al. (2011), with appropriate unit conversion, are used in Figure 3. Based on (4), the ratio $(\frac{q}{i})/K$ can be expressed as

$$\frac{q/i}{K} = 1 - \frac{1 - e^{-\frac{i}{I}}}{i/I} \qquad (10)$$

The above equation is used to fit the data sets of Wang et al. (2011). A comparison between calculated curve and the data is given in Figure 5. The fitted I values are 20.4, 12.0 and 10.0 for rock samples with permeability values of 0.195 mD, 0.347 mD and 0.524 mD, respectively. The comparison between data and (10) is reasonable, suggesting that (4) is valid for describing flux-gradient relationships for these rock samples.

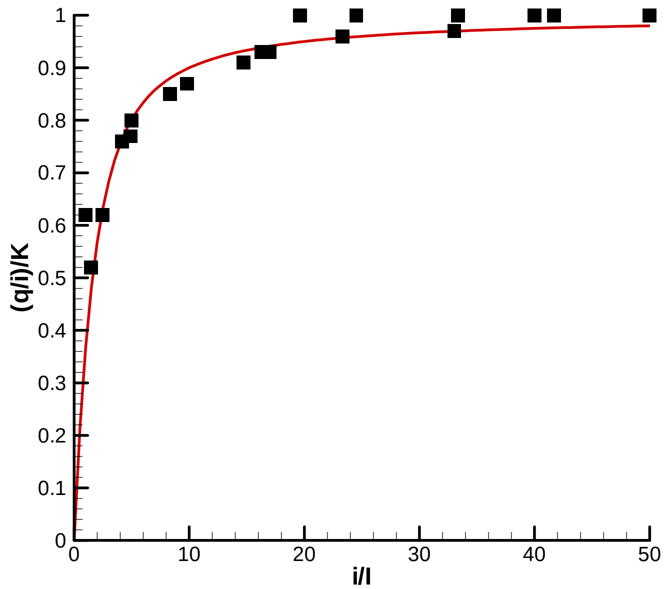

Figure 5. Match of Eq. (10) with test data of Wang et al. (2011).

All the data sets discussed above are for saturated conditions. Cui et al. (2008) reported on measurements of unsaturated hydraulic conductivity for a compacted sand-bentonite mixture. To the best of our knowledge, this work provided the first reliable data set of water flux as a function of hydraulic gradient under unsaturated conditions. The tests of Cui et al. (2008) were conducted under two experimental boundary conditions: constant volume and free swelling. In this study, we focus on the data for constant-volume conditions only, based on the reasoning that under constant-volume conditions and for a given capillary pressure, hydraulic processes and pore structures are approximately the same at different locations within the soil sample (Cui et al., 2008). Cui et al. (2008) used the instantaneous profile method to determine the unsaturated hydraulic conductivity for infiltration tests of a vertical sand-bentonite column. The sand-bentonite mixture was directly compacted in a metallic cylinder (50 mm in inner diameter, 250 mm high). The bottom of the test cell was connected to a water source, and the upper end to an air source under atmospheric pressure. Under transient water-flow conditions, vertical distributions of capillary pressure were directly measured as a function of time at several locations along the column. The relationship between water content and capillary pressure was independently measured under constant volume conditions. This relationship enables them to estimate vertical distributions of water content from the capillary-pressure measurements. Based on these vertical distributions at different times, and on the mass balance at each location within the soil column, they estimated the water flux at that location as a function of capillary pressure and hydraulic gradient (their Figure 12). The details of this instantaneous method can be found in Daniel (1982) and Cui et al. (2008). For this study, linear part of the flux-gradient relationship for a given capillary pressure is approximated by a straight line through the two points with the largest hydraulic gradients. Consequently, the unsaturated permeability (relative permeability for water multiplied by absolute permeability) and the threshold gradient I can be obtained for each capillary pressure. Figure 3 shows the match between (8) and the data set.

As shown in Figure 3, the data sets can be reasonably fitted by a relationship between I and permeability k (m^2)

$$I = Ak^{B} \qquad (11)$$

with A= 4.0 E-12 and B =-0.78. This is very encouraging considering that the data sets were collected for different kinds of clay media and low-permeability materials and by different researchers. This may imply the existence of a universal relationship between I and permeability, although some degree of fluctuation exists in Figure 3.

Equation (11) can be used to evaluate the relative importance of non-Darcian behavior for a given media if its permeability is known. For example, non-Darcian behavior can be safely ignored for permeability values larger than 10^{-13} m^2. Also, when data are not available for estimating site-specific values for A and B, (11) can be directly used for estimating parameter I based on more easily obtained permeability values. Another usefulness of (11) is to provide a criterion for permeability to be accurately measured. For example, if the hydraulic gradient head used in tests is less than the value calculated from (11), the measured permeability will depend on the gradient and does not correspond to the correct value.

It is especially of interest that the data for unsaturated flow follows the same relationship as long as permeability is replaced by unsaturated permeability (relative permeability multiplied by permeability) (Figure 3). This is because unsaturated permeability characterizes sizes of pores occupied by water under unsaturated conditions.

In practical applications, it is very difficult, if not impossible, to determine parameter I for different capillary pressures. Figure 3 seems to indicate that the same A and B values can be used for different capillary pressures, although this needs to be further confirmed when more data are available for unsaturated conditions. If it is the case, A and B values can be either estimated from data at one capillary pressure or using values from Figure 3 when site-specific data are not available. Then I values for different capillary pressures (or saturations) can be estimated from (11) using unsaturated permeability. The relationships between unsaturated permeability and capillary pressures are well established for unsaturated media (van Genuchten, 1980; Brooks and Corey, 1964).

4. Discussion

We developed a new relationship for one-dimensional water flow and hydraulic gradient given in Eq. (8). It can be considered a generalization of two previous formulations (Eqs. (4) and (5)) and is more consistent with laboratory experiments. In many studies, we need to investigate multidimensional water-flow processes. For that purpose, we extend Eq. (8) to three-dimensional, homogeneous and isotropic clay media:

$$\mathbf{q} = -K[i - \frac{I}{\gamma(\frac{1}{\alpha})}\gamma(\frac{1}{\alpha}, \left(\frac{i}{I*}\right)^{\alpha})]\,\mathbf{n_i} \qquad (12)$$

where $\mathbf{q}$ (m/s) and $\mathbf{n_i}$ (-) are water flux vector and unit vector for hydraulic gradient, respectively. Relation between I and I^* is given by (9-1). When threshold gradient I approaches to zero, (12) is reduced to the commonly used form of Darcy’s law. It should be emphasized that (12) is an empirical relationship and developed based on one dimensional test results. It is logical to directly extend (8) to (12) for homogeneous and isotropic cases because both equations are consistent with the one-dimensional-test condition that flux and negative hydraulic gradient are along the same direction. That condition is violated for three-dimensional anisotropic porous media. In this case, it is not clear whether (12) is adequate.

Eq. (12) is developed for both saturated and unsaturated flow processes. As demonstrated in Figure 2, (12) can also be used to satisfactorily describe unsaturated flow, although the related flux-gradient data are very limited. For unsaturated flow, both hydraulic conductivity and threshold gradient I are dependent on capillary pressure or water saturation. In this regard, the developed relationship between permeability and I (Eq. (11)) are especially useful. As previously indicated, it is practically difficult, if not impossible, to experimentally obtain flux-gradient data under constant capillary pressures (or saturations) to determine the threshold gradient I for a large range of capillary-pressure values. However, through relationship given in (11), we can relatively easily to obtain I from unsaturated permeability whose dependence on saturation and capillary pressure has been well established.

Also note it is not clear how parameter α is related to other clay parameters, while a single α value cannot adequately capture all the non-Darcian flow behavior of interest, as demonstrated in Figure 2. One speculation is that parameter α may be related to pore-size distribution, because the observed flux-gradient relation is a combination of water flow processes across pores with different sizes that correspond to different agrees of water-solid interaction. Nevertheless, this needs to be further confirmed in future studies.

Finally, parameters in (12) depend on both electrolyte concentration of solution that flows through clay media and temperature. For example, Swartzendruber (1961) analyzed the data of Lutz and Kemper (1958) that were collected for solutions with different electrolyte concentrations and found that the threshold gradient decreases with the concentration. As previously indicated, the impact of the concentration is partially considered herein through the correlation between permeability and I (Eq. (11)). For example, with increasing solution concentration, permeability will increase because of shrinkage and thus parameter I will decrease based on (11), which is consistent with the analysis results of Swartzendruber (1961). Studies on temperature impact on non-Darcian behavior are rare in the literature. To the best of our knowledge, the only study that is related to the temperature impact is Miller and Low (1963). They experimentally found that the critical hydraulic gradient (below which water is immobile) decreases with increasing temperature. They argue that this is because increased temperature may weaken the bounding between water molecules and clay surface. More experimental and theoretical studies are needed to fully establish temperature dependence of related parameters in (12).

5. Summary

Water flow in clay media is an important process for a number of practical applications. Experimental results indicate that traditional form of Darcy's law is not adequate for describing water flow processes in clay media because the observed relationship between water flux and hydraulic gradient can be highly non-linear. To capture this non-Darcian flow behavior, we proposed a new relationship between water flux and hydraulic gradient by generalizing the currently existing relationships. The new relationship is shown to be consistent with experimental observations for both saturated and unsaturated conditions. In this paper, we also developed an empirical relationship between permeability and threshold hydraulic gradient, an important measure of non-Darcian behavior. The latter relationship is practically useful because it can reduce the number of parameters whose values need to be determined from experiment data in order to model non-Darcian behavior. However, how to incorporate impacts of temperature and electrolyte concentrations into the proposed relationships needs further research.

Acknowledgment

The original version of this paper was reviewed by Dan Hawkes at Lawrence Berkeley National Laboratory, Prasad Nair from US Department of Energy, and Yifeng Wang at Sandia National Laboratories. Their constructive comments are appreciated. We also appreciate comments from the Associate Editor and anonymous reviewers. This work was funded by and conducted for the Used Fuel Disposition Campaign under DOE Contract No. DE-AC02-05CH11231

References

Blecker, R.F., 1970. Saturated flow of water through clay loam subsoil material of the Brolliat and Springerville soil series. Master Thesis, The University of Arizona.

Brooks, R. H., and Corey, A. T., 1964. Hydraulic properties for porous media. Hydro. Rep. No. 3, Colorado State University, Fort Collins.

Buckingham, E. 1907. Studies on the movement of soil moisture. Bulletin 38. USDA Bureau of Soils, Washington, DC.

Burdine, N.T. (1953) Relative permeability calculations from pore size distribution data, Am. Ins. Min. Metail. Pet. Eng, 198, 71-77.

Chen, X., Cao, G.X., Han, A.J., Punyamurtula, V.K., Liu, L., Culligan, P.J., Kim, T., and Qiao.Y., 2008. Nanoscale fluid transport: size and rate effects. Nano Letters, 8(9), 2988-2992.

Cui, Y.J., Tang, A. M., Loiseau, C., and Delage P., 2008. Determining the unsaturated hydraulic conductivity of a compacted sand-bentonite mixture under constant-volume and free-swell conditions. Physics and Chemistry of the Earth 33, S462-S471.

Dubin, B., and Moulin, G., 1986. Influences of critical gradient on the consolidation of clay. In Consolidation of soils, testing and evaluation (eds Young and Townsend), ASTM STP 892, pp. 354-377. West Conshohocken, PA.

Farrow, M.R., Chremos, A., Camp, P.J., Harris, S.G., and Watts, R.F., 2011. Molecular simulations of kinetic-friction modification in nanoscale fluid layers. Tribol Lett. 42, 325-337.

Hansbo, S. 1960. Consolidation of clay, with special reference to influence of vertical sand drains. Swed. Geotech. Inst. Proc. 18, Stockholm.

Hansbo, S., 2001. Consolidation equation valid for both Darcian and non-Darcian flow. Geotechnique 51(1), 51-54.

Liu, H.H., Li, L.C., and Birkholzer, J., 2012. Unsaturated properties for non-Darcian water flow in clay. Journal of Hydrology, 430-431, 173-178.

Liu, H. H., 2011. A conductivity relationship for steady-state unsaturated flow processes under optimal flow conditions. Vadose Zone J. 10(2), 736-740, doi:10.2136/vzj2010.0118.

Low, P.F., 1961. Physical chemistry of clay-water interaction, Advance in Agronomy, 13, 269-327.

Lutz, J.F., and Kemper, W.D., 1959. Intrinsic permeability of clay as effected by clay-water interaction. Soil Sci. 88: 83-90.

Ma, M., Shen, L., Sheridan, J., Liu, Z., Chen C., and Zheng, Q., 2010. Friction law for water flowing in carbon nanotubes. 2010 International Conference on Nanoscience and Nanotechnology. Feb. 20-22, Sydney, Australia.

Miller, R.J., and Low P.F., 1963. Threshold gradient for water flow in clay systems. Soil. Sci. Soc. Am. Proc. 27(6), 605-609.

Nimmo, J. R., and Landa, E.R., 2005. The soil physics contributions of Edgar Buckingham. Soil. Sci. Am. J. 69:328-342

Tsang, C.F., Barnichon, J.D., Birkholzer, J., Li X.L, Liu, H.H., Sillen, X., 2012. Coupled thermo-hydro-mechanical processes in the near field of a high-level radioactive waste repository in clay formations. Int. J. Rock Mech. Min. Sci. 49: 31-44. DOI 10.1016/j.ijrmms.2011.09.015.

Swartzendruber, D. 1961. Modification of Darcy's law for the flow of water in soils. Soil Science 93: 22-29. Soil. Sci. Soc. Am. J. 69, 328-342.

Van Genuchten, M., 1980. A closed-form equation for predicting the hydraulic conductivity of unsaturated soil. Soil. Sci. Soc. Am. J. 44(5), 892-898.

Wang, X.X., Yang, Z. M., Sun, Y.P., and Liu, X.X., 2011. Experimental and theoretical investigation of nonlinear flow in low permeability reservoir. Procedia Environmental Sciences, 11, 1392-1399.

Wang, Y.F., Gao, H.Z., and Xu, H.F., 2011. Nanogeochemistry: nanostructures and their reactivity in natural systems. Frontiers in Geochemistry: contributions of geochemistry to the study of the earth (eds Russell S. Harmon and Andrew Parker), Backwell Publishing Ltd.

Zhou, Q.L., Birkholzer, J.T., Tsang C.F., J. Rutqvist, 2008. A method for quick assessment of CO2 storage capacity in closed and semi-closed saline formations. International Journal of Greenhouse Gas Control, 2(4), 626-639.

Zou, Y., 1996. A non-linear permeability relation depending on the activation energy of pore liquid. Geotechnique 46(4), 769-774.