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Method for Modeling the Gradual Physical Degradation of a Porous Material

Cementitious and other engineered porous materials encountered in waste disposals may degrade over time due to one or more mechanisms. Physical degradation may take the form of cracking (fracturing) and/or altered (e.g. increased) porosity, depending on the material and underlying degradation mechanism. In most cases, the hydraulic properties of degrading materials are expected to evolve due to physical changes occurring over roughly the pore to decimeter scale, which is conducive to calculating equivalent or *effective* material properties.

The exact morphology of a degrading material in its end-state may or may not be known. In the latter case, the fully-degraded condition can be assumed to be similar to a more-permeable material in the surrounding environment, such as backfill soil. Then the fully-degraded waste form or barrier material is hydraulically neutral with respect to its surroundings, constituting neither a barrier to nor conduit for moisture flow and solute transport.

Unless the degradation mechanism is abrupt, a gradual transition between the intact initial and fully-degraded final states is desired. Linear interpolation through time is one method for smoothly blending hydraulic properties between those of an intact matrix and those of a soil or other surrogate for the end-state.

Letting f and F represent the fractions of the final (e.g. soil) and initial (intact matrix) states, respectively, the component fractions satisfy the equation:

$$f + F = 1 \tag{1}$$

The soil fraction f, which can also be interpreted as a degradation fraction, varies between 0 and 1 according to the piecewise linear function:

$$f(t) = \max\left[0, \min\left(1, \frac{t-t_1}{t_2-t_1}\right)\right] \tag{2}$$

where t_1 and t_2 are the degradation start and end times, respectively. The composite porosity [cm^3 void /cm^3 total] is:

$$n = fn_s + Fn_m \tag{3}$$

where n_s and n_m are the porosities of the soil and matrix, respectively. Using the same subscripting convention and weighting scheme, the effective saturated hydraulic conductivity [cm/s] of the composited material is:

$$K = fK_s + FK_m \tag{4}$$

For an anisotropic material, K may represent either horizontal or vertical conductivity. Equation (4) is an arithmetic average consistent with parallel flow through the binary constituents (Freeze and Cherry 1979 Equation (2.32)). Alternative, non-linear, weighting/averaging schemes may be justified depending on the structure of composite material. For example, harmonic averaging would be appropriate for perpendicular flow through the constituents (Freeze and Cherry 1979 Equation (2.31)) and geometric averaging produces the effective conductivity of an isotropic, two-dimensional, medium with log-normally distributed K (Gelhar 1993, Equation [4.1.58]). Equation (4) can be generalized as (Ababou and Wood 1990)

$$K = [fK_s{}^p + FK_m{}^p]^{1/p} \tag{5}$$

where $-1 \le p \le 1$ and special cases include:

arithmetic $p = 1$

geometric $p \to 0$

harmonic $p = -1$

Non-integer values of p are permissible; see Phifer et al. (2006, Section 5.2.2.2) for several examples. Arithmetic averaging produces the largest K for a given f and is adopted for the remainder of this discussion as a conservative-tending assumption and to be consistent with recent Savannah River Performance Assessment examples.

Continuing with the remaining material properties, the saturation [cm^3 liquid /cm^3 void] function of the composited material is:

$$S(\psi) = \frac{fn_sS_s(\psi)+Fn_mS_m(\psi)}{n} \tag{6}$$

where ψ is suction (negative pressure) head [cm]. The relative permeability [-] function becomes:

$$k_r(\psi) = \frac{fk_{rs}[S(\psi)]K_s+Fk_{rm}[S(\psi)]K_m}{k_r(\psi)K} \tag{7}$$

The unsaturated conductivity [cm/s] of the composited material is

$$K_u = k_r(\psi)K \tag{8}$$

The water retention curve for the composited material represented by Equation (6) is presented with suction head as the independent variable, the usual convention. However, the relative permeability curve is also presented in Equation (7) as a direct function of suction head, rather than the usual choice of saturation, to more clearly illustrate how unsaturated conductivity defined by $k_r(\psi)K$ varies with the primary flow variable, pressure head ($-\psi$). That is, rather than viewing unsaturated conductivity variation for the composited material through the separate functions $k_r(S)$ and $S(\psi)$, the confounding intermediate variable S is eliminated between the water retention and conventional relative permeability curves to achieve the single characteristic curve $k_r(\psi)$.

The blending process is most conveniently conducted using analytic expressions for the water retention and relative permeability curves of the intact and soil materials, as opposed to tabular data that may not share the same suction head values (ψ_i). The van Genuchten (1980) / Mualem (1976) functions are a popular choice for analytic characteristic curves. The van Genuchten (1980) water retention function is:

$$S_e = \frac{S-S_r}{1-S_r} = \frac{\theta-\theta_r}{\theta_s-\theta_r} = \left[\frac{1}{1+(\alpha\psi)^n}\right]^m \tag{9}$$

where θ is water content [cm^3 liquid /cm^3 total], the subscripts e, r, and s refer to effective, residual, and saturated conditions, respectively. The model parameters are S_r (or θ_s and θ_r), α, n, and m. The relative permeability function is given by:

$$k_r = {S_e}^{1/2}\left[1-\left(1-{S_e}^{1/m}\right)^m\right]^2 \tag{10}$$

For the Mualem (1976) conductivity model:

$$m = 1 - 1/n \tag{11}$$

leaving S_r, α, and n as the independent model parameters.

Figure 1 reproduced from Jordan and Flach (2013) illustrates example transitions from intact cementitious materials to fully-degraded conditions resembling those of soils. The hydraulic properties of the initial and end-state materials are presented in Table 1. At intermediate blending fractions ($0 < f < 1$) the hydraulic conductivity functions are observed to have a characteristic double-hump or two-tiered nature.

Table 1. Hydraulic parameters for cementitious materials and soils.

Parameter	Intact concrete	Intact grout	Backfill soil	Lower Vadose Zone (LVZ) soil	Sand	Gravel
θ_s	0.082	0.58	0.35	0.39	0.38103	0.29843
θ_r	0	0	0.10062	0.14014	0.1349	0.01564
S_r	0	0	0.2875	0.3593	0.354040	0.0524076
α (cm^{-1})	2.0856E-06	1.9994E-05	0.01464	0.01257	2.95E-02	1.43E-01
n, van Genuchten (1980)	1.9433	1.51012	1.22727	1.39358	1.40995	1.45746
K (cm/s)	9.3E-11	6.4E-09	4.1E-05	9.1E-05	2.8E-04	1.5E-01
n, porosity	0.11	0.58	0.35	0.39	0.38	0.3

Transitioning completely to a soil is conceptually most clearly consistent with a degradation mechanism where porosity and pore size increase but internal stresses are not present that would produce a fractured medium. An example mechanism is decalcification of a concrete or grout. Linear blending of properties is consistent with the concept of parallel flow through distinct intact and fully-degraded regions that vary in proportion, and results in the intermediate conductivity variations that are two-tiered (Figure 1). In this manner, the partially-degraded cementitious materials resemble a soil with a bimodal pore size distribution, that is, having both micro- and macro-porosity (Durner 1994, Simunek et al. 2003, Priesack and Durner 2006). As discussed earlier, alternative (non-linear) weighting schemes are possible, but lacking a specific physical driver, are rejected here in favor of simple linear averaging.

For degradation resulting in an increasingly fractured medium, several alternative representations of degraded conditions could be adopted as shown in Figure 2 reproduced from Altman et al. (1996). The blending process described above falls under the "Equivalent Matrix and Fracture Continuum" representation depicted in Figure 2, or *Equivalent Continuum Model* (ECM) in short. The ECM is most appropriate for steady-state flow conditions and smaller scale fractures, such that local equilibrium is achieved between fractures and matrix. The former conditions are generally expected for cementitious materials degrading by chemical attack and lying several feet below the ground surface.

Within the ECM framework, blending approaches other than a 100% transition to soil could be adopted following Peters and Klavetter (1988) among others (Simunek et al. 2003). Pruess (1998) notes that

> "Recent theoretical and experimental work suggests that relative permeability and capillary pressure behavior of fractures is similar to that of highly permeable media with intergranular porosity (Pruess and Tsang, 1990; Firoozabadi and Hauge, 1990; Persoff and Pruess, 1995). Accordingly, we used the customary van Genuchten correlations, with parameters chosen as for coarse sands".

Thus high-permeability granular materials such as coarse sands or gravels are reasonable surrogates for natural fractures with rough surfaces and/or infilled with granular material.

Figure 3 modified from Jordan and Flach (2013) shows the results of compositing an intact concrete first with the “Gravel” and then alternatively with the “Sand” materials from Phifer et al. (2006) summarized in Table 1, while preserving the saturated conductivity of Lower Vadose Zone (LVZ) soil. The LVZ soil curve is included in the figure as a point of reference. “Sand” is shorthand for sandy native sediments at the Savannah River Site, which contain up to 25% fines under the chosen classification. Thus the “Sand” material from Phifer et al. (2006) has a lower saturated conductivity than a clean coarse-grained sand, and could be questioned as a surrogate for high-permeability fractures. Nonetheless both materials have a higher saturated conductivity than Backfill, and the volume fraction (b) of higher-permeability material needed to achieve the same saturated hydraulic conductivity as Backfill for the fully-degraded concrete is less than 100%. The specific fractions of Gravel or Sand used in Figure 3 are 0.06% and 32.5%, respectively. Considering the tiny fraction of total volume occupied by fractures, Gravel is viewed as the more appropriate surrogate material for fractures. Blending with Gravel produces a more pronounced two-tiered curve, which is consistent with the ease with which fractures are dewatered through tension in the adjoining matrix (Wang and Narasimhan 1985). Blending to 100% LVZ soil is observed to produce higher unsaturated conductivities within the suction head range $-\infty < \psi < 1500$ cm typical of Savannah River Site applications than blending with the two higher permeability materials. In this sense, a choice of LVZ (or Backfill) soil would be a conservative-tending assumption for the end-state surrogate.

Another ECM approach is to match the saturated conductivity of a concrete-gravel blend to that of a fractured medium with specified attributes, principally fracture aperture b and spacing B. For this exercise, gravel from Phifer et al. (2006) is again considered to be a reasonable surrogate for fractures. The saturated conductivity of a saturated fracture is (e.g. Wang and Narasimhan 1985):

$$K_f = \frac{\rho g b^2}{12\eta} \tag{12}$$

where ρ is liquid density [kg/m^3], g is gravitational acceleration [m/s^2], b is fracture aperture [m], and η is liquid viscosity [kg/m-s]. The effective conductivity of the fractured porous matrix is:

$$K = \frac{bK_f + BK_m}{b+B} \approx \frac{b}{b+B} K_f = f K_f \tag{13}$$

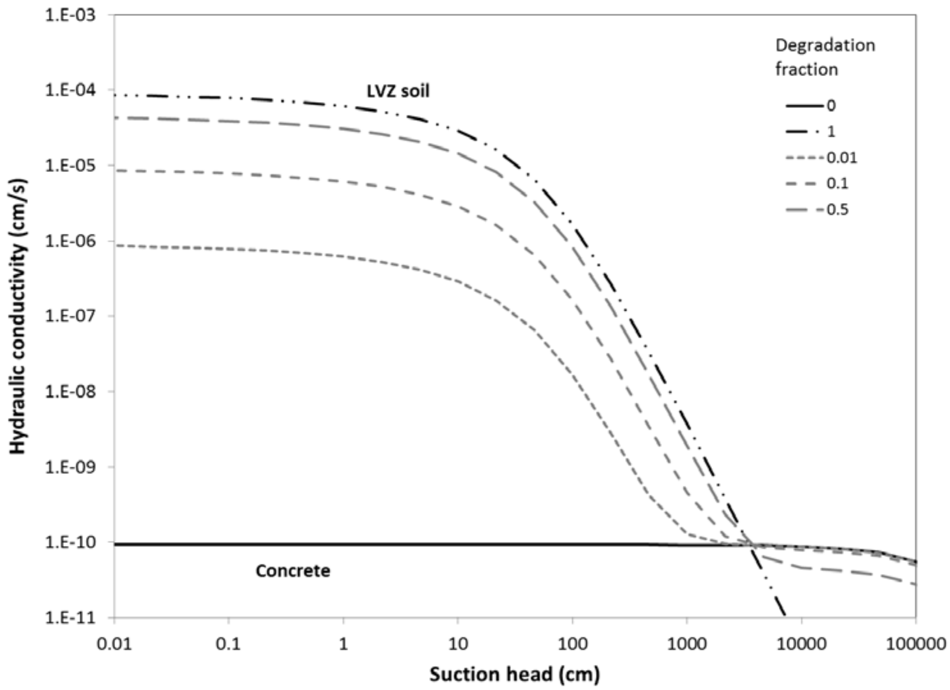
where $f = b/(b+B)$ in Equation (4) and the contribution of the low-permeability matrix is then assumed to be negligible. As an example, the effective saturated conductivity of a low-permeability porous medium with 5 mil (0.127 mm) fractures spaced at 1 cm is 1.65E-02 cm/s. The fraction of gravel required to produce this same saturated conductivity is 10.7% using Equation (4). Unsaturated conductivity for this concrete-gravel blend, which is representative of a severely damaged concrete, is shown in Figure 4 modified from Jordan and Flach (2013). The LVZ soil curve also shown as a point of reference to preceding figures.

Or and Tuller (2000) present a direct method for defining fracture hydraulic properties based on an idealized fracture geometry. These derived fracture properties can be used instead of properties from a surrogate granular material in an ECM model. Following Flach et al. (2009, Section 3.7), Figure 5 from Jordan and Flach (2013) presents example ECM curves for two conditions: a) 5 mil (0.127 mm) fractures spaced at 1 cm (as considered in Figure 4), and b) 50 mil (1.27 mm) fractures spaced at 10 cm. Within the suction head range 50 < ψ < 1500+ cm, conductivities based on Or and Tuller (2000) are lower than both the concrete-gravel blend and LVZ soil. Thus, selecting a granular material to represent a degraded state in the form of fractures is a conservative-tending assumption compared to Or and Tuller (2000).

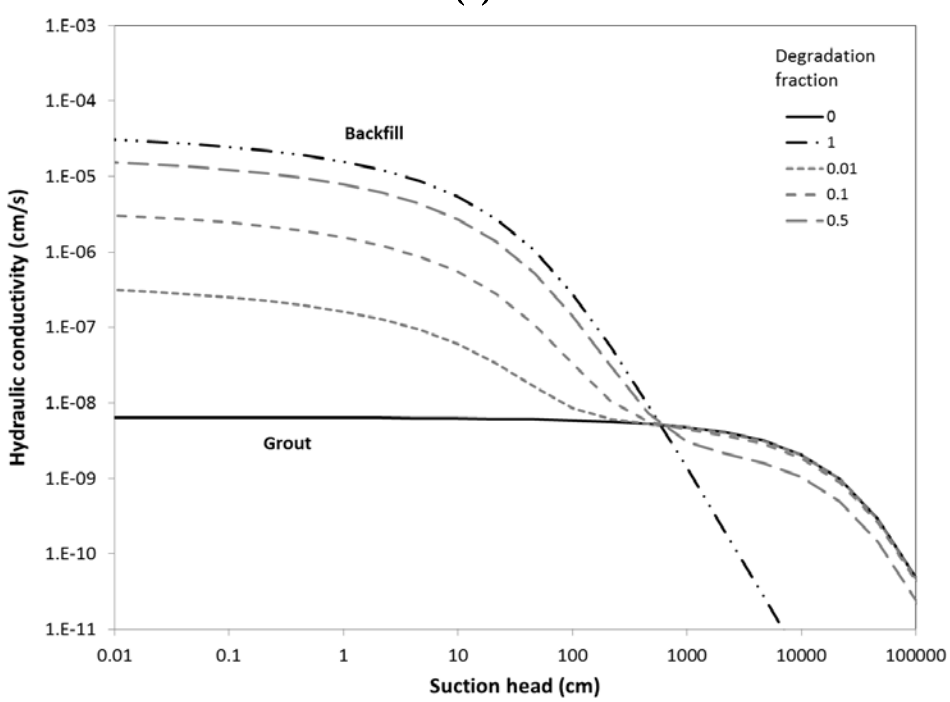
Flach et al. (2015) used an *outflow extraction method* to estimate the unsaturated hydraulic conductivity of a fractured grout and compared their experimental results to LVZ and Backfill materials (Table 1). Similar to Figure 5, the hydraulic conductivity of the fractured specimens was estimated to be lower than that of the soil materials for 100 < ψ < 1000 cm (see Figure 5 in Flach et al. 2015). The authors concluded that:

> "With respect to PA applications, comparisons of the unsaturated conductivity for the micro-fractured grout samples suggests that soils may serve as conservative surrogates for damaged cementitious materials, assuming that higher conductivity at higher tension heads is conservative for facility performance."

Figures 1, 3, 4 and 5 illustrate example applications of the general concept defined by Equations (1) through (8) for smoothly evolving material properties, and are not intended to be limiting. For example, Section 6.3.3.1 of Phifer et al. (2006) uses non-linear weighting per Equation (5) with $p = 0.33$ for a Components-In-Grout material.



(a)



(b)

Figure 1. Example transitions of intact cementitious materials from intact to fully-degraded conditions represented by soils: (a) concrete and (b) grout.

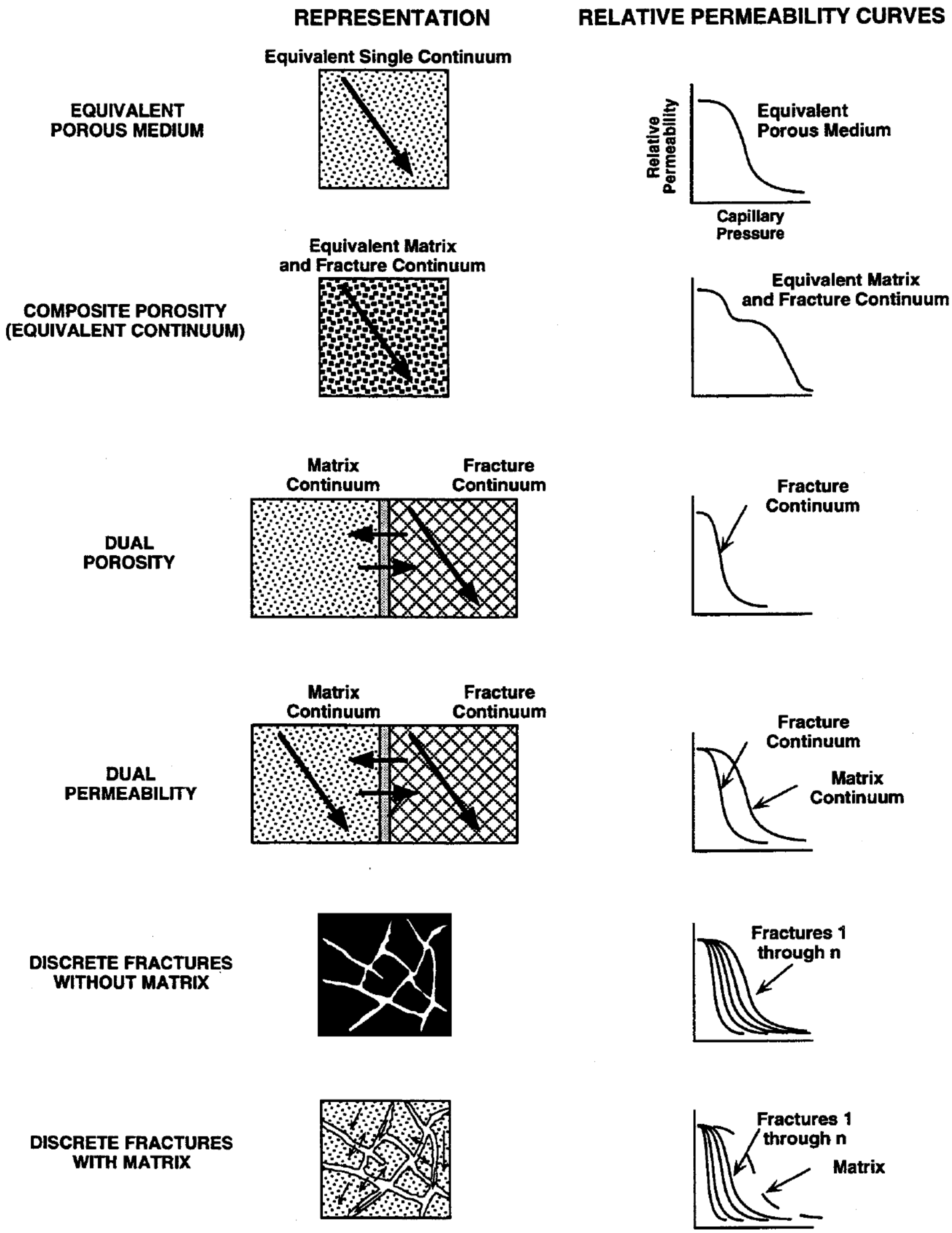


Figure 2. Alternative representations of a fractured medium; reproduced from Altman et al. (1996, Figure 2-2).

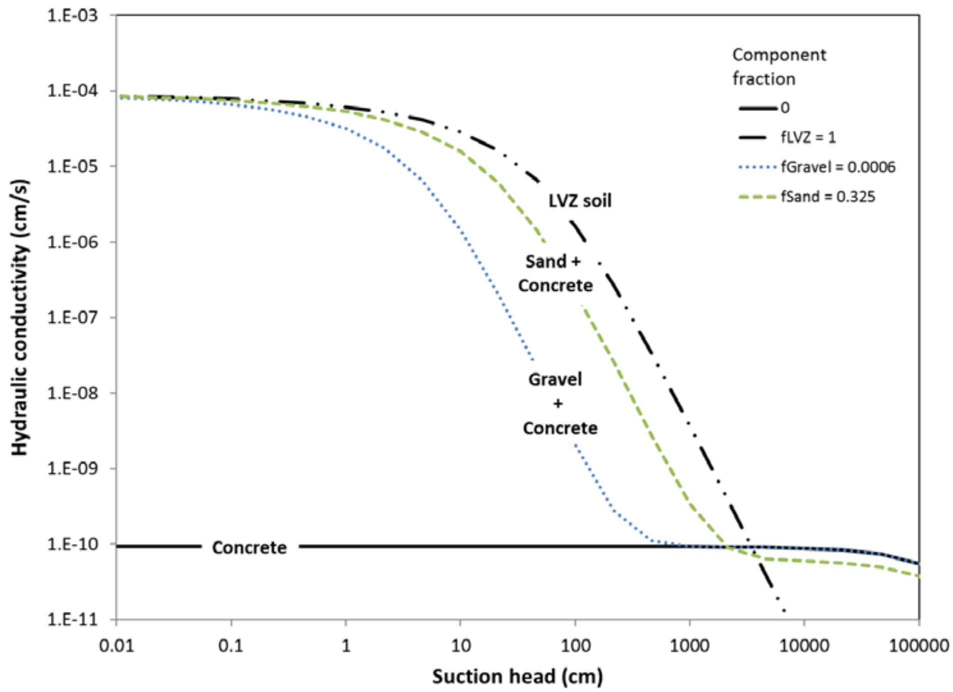


Figure 3. Alternative surrogate soils for blending to fully-degraded conditions.

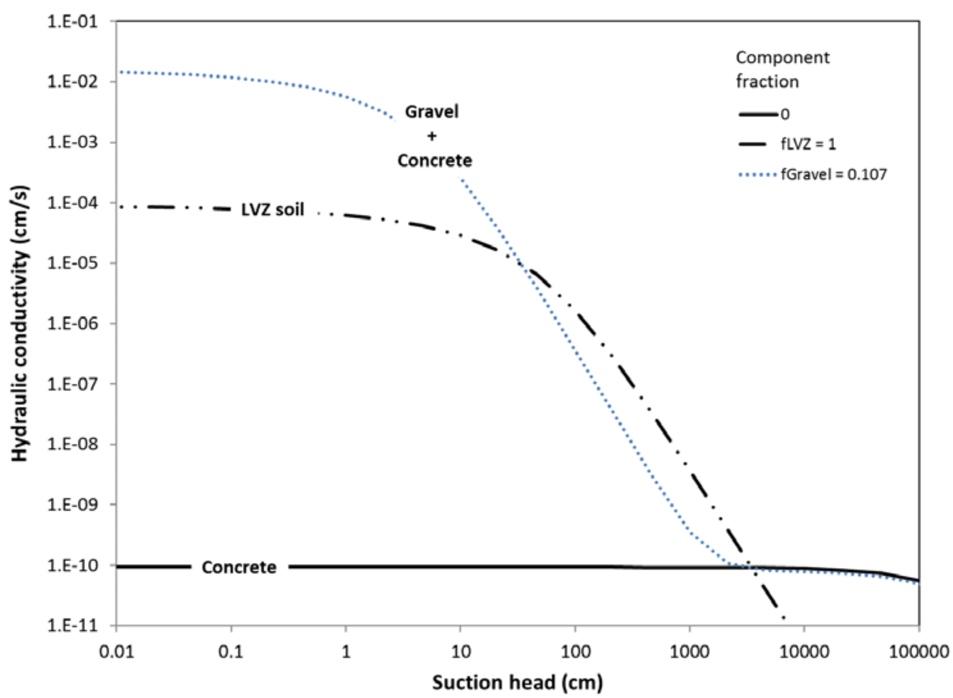


Figure 4. Partial blending (10.7%) to gravel representing degradation to 5 mil fractures spaced at 1 cm.

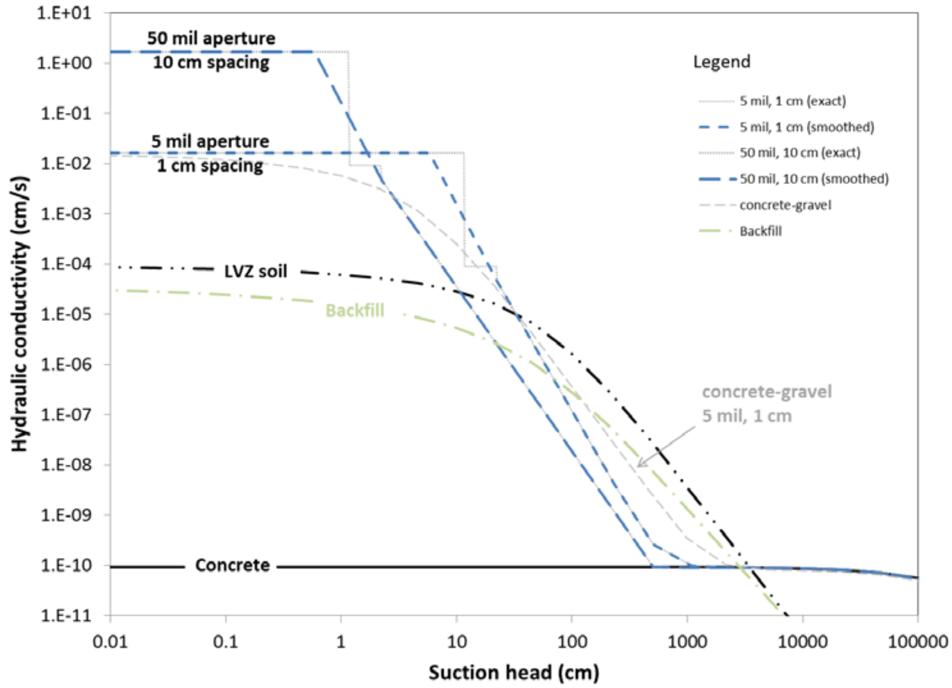


Figure 5. Degradation to selected fractured conditions using fracture properties based on Or and Tuller (2000).

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