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Modeling Geologic Waste Repository Systems Below Residual Saturation

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INTRODUCTION

Unsaturated alluvium is one of several generic deep geologic disposal environments considered in the United States Department of Energy's Spent Fuel and Waste Science Campaign. However, there are physical and numerical challenges in modeling flow and transport associated with unsaturated systems. Exacerbating this, decay heat released from the radioactive decay of spent nuclear fuel packages may significantly elevate temperatures in the repository system, resulting in complex thermal-hydrological interactions. Thus, simulations of spent fuel and high-level waste repository systems must adequately model coupled thermo-hydrologic interactions in multiphase systems.

In an isothermal system, a partially saturated porous medium will drain to a small but non-zero degree of water saturation known as the residual saturation. Many models, such as the van Genuchten model[1], capture this behavior by allowing the capillary pressure to approach an infinite value at some small but non-zero residual saturation. When relative permeability is derived from capillary pressure curves, as in the Mualem[2] model, this also specifies the relative permeability to be zero at residual saturation. Thus, the water saturation can never fall below the residual limit by liquid-phase flow due to both the capillary pressure and relative permeability models.

However, water can and is removed below the residual limit by evaporation and gas-phase flow. This can be encountered in high-level nuclear waste repositories as the decay heat increases the temperature, driving evaporation and convection. As capillary pressure cannot be infinite, either physically or numerically, when water is driven off past the residual saturation, several extensions have been recently implemented in PFLOTRAN as part of the Geological Disposal Safety Assessment (GDSA) development project. However, these extensions can introduce their own physical or numerical idiosyncrasies. This work discusses some criteria a modeler should consider when utilizing these options.

Numerically, the capillary pressure curve must be both continuous and smooth for Newton's method to efficiently solve the implicit system of equations describing mass and heat transfer in multiphase systems. Consequently, smooth extensions to capillary pressure can have significant improvement in performance on a series of benchmark problems. One such benchmark problem is presented here whereby evaporation, boiling from the thermal pulse of a waste package, and infiltration from rainfall in an unsaturated alluvium system all interact to dynamically dry and re-wet the system. Using the new smooth capillary pressure extensions, this problem can finish where it had previously stalled due to unacceptably small timesteps necessary for convergence.

Geologic Waste Repository Reference Case

A broad range of possible scenarios are being considered with the GDSA program. Within this program, the unsaturated zone (UZ) reference case is particularly challenging as it has significant rates of evaporation or even boiling in the porous media. The UZ model consists of a mined repository in unsaturated alluvium located 255 m below the land surface, and the repository is modeled to contain hot commercial spent nuclear fuel[3]. The water table depth is estimated to be at 500 m depth. Aside from infiltration of rainwater, at 245 m above the water table, the initial water content will initially be near residual saturation.

At the same time, due to radioactive decay heat, the temperature in the spent nuclear fuel waste packages will be elevated. Initial calculations have estimated peak temperatures ranging from 150 ºC to 350 ºC. While the temperature in the surrounding porous media will be less the peak, the elevated temperatures will drive elevated rates of evaporation or even boiling, at ambient or possibly elevated pressures.

While water is at an elevated temperature near the waste packages, most of the repository system remains near the ambient temperature of 27 ºC. Consequently, water vapor generated by decay heat will not be removed in a closed repository system. After gas phase transport, the water vapor is anticipated to condense on cooler materials, driving natural convection flows[4].

This situation can be problematic when applying standard approaches to model two-phase flow in porous media. For example, the two-phase flow model in PFLOTRAN was initially adopted from using the model described by Chen *et al*[5]. The numerical difficulty due to the singularity in the capillary pressure model is not new and has previously been addressed using piecewise extensions by Sun *et al*[6]. This work will examine extensions to relative permeability and capillary pressure below the residual on a physical basis.

Two-Phase Darcian Flow Model

A common but ultimately empirical approach is to follow the approach of Muskat model and extend Darcy's law to two-phase systems by introducing the concept of relative permeabilities[7]. The phenomenon of residual saturation was considered to be the saturation where the relative permeability of the wetting phase approaches or is equal to zero. And while it was evident there was residual water, it was presumed to be removeable if evaporative processes were adequately modeled.

Independent of the relative permeability model, Richards[8] modeled the phenomenon of capillary rise in porous media, modelling the flow of a wetting fluid into porous media. More specifically, Richards modelled the only the flow of fluid due to capillary pressure due to cohesive forces that predominate in a wetted porous media. Importantly, this explicitly excludes the forces of adhesion, including phenomenon such as film flow that predominate at very low degrees of saturation[9]. When adsorbed films are neglected, the capillary pressure follows the Young-Laplace equation and is presumed to approach infinite values in the limit of infinitely small pores.

By merging the concepts of relative permeability and capillary pressure[10] the generalized Darcy equations and can be formulated[11].

$$q_w = -k_i \frac{k_{rw}}{\mu_w} \nabla p_w \quad (1)$$

$$q_g = -k_i \frac{k_{rg}}{\mu_g} \nabla p_g \quad (2)$$

$$p_g - p_w = p_c \quad (3)$$

Here, q_w and q_g are the volume flux of liquid water and gaseous air, respectively. k_i is the intrinsic permeability while k_{rw} and k_{rg} are the relative permeabilities of the corresponding phases. Finally, μ and p represent the dynamic viscosity and pressure of their respective phases. While general, the validity of this approach is entirely dependent upon the validity of the underlying models for both relative permeability and capillary pressure.

This problem is numerically challenging as both relative permeability and capillary pressure are functions of the phase saturation. While saturation can be defined in many ways, here saturation will be defined on a fractional volumetric basis such that the sum of the water and air phase saturations equals one (*i.e.*, $S_w + S_{nw} = 1$).

Saturation-based Mass Balance

If the expression for the volume flux is substituted into a mass balance, a nonlinear partial-differential equation is created.

$$\phi \frac{\partial(\rho_w S_w)}{\partial t} + \nabla \cdot (\rho_w q_w) = Q_w \quad (4)$$

Here, ρ_w represents the mass density, ϕ the porosity, and Q_w the source/sink term for the liquid phase of water, including evaporation and condensation. For a fluid that is approximated to be incompressible, the density of the fluid is close to constant, and saturation is the primary solution variable.

Above the residual, the Darcian model applies as usual. But if residual saturation is to be understood as the region where liquid phase flow ceases due to a lack of hydraulic connectivity, the mass balance is trivial below the residual. That is, only the source/sink term contributes, and transport of the water is limited to vapor phase flux. Solving the mass balance is not challenging in either of the individual regimes, but it is in the transition when the simulation spans both regimes.

While relative permeability models generally approach zero at the residual saturation, a common assumption is that capillary pressure is also infinite at residual. Not only that, but to find the pressure gradient, the system requires the derivative with respect to saturation. The Darcian model for volume flux then seeks to model the product of $k_{rw} \frac{dP_c}{dS_w} \frac{dS_w}{dx}$, which is ill-defined near the residual liquid saturation.

Previously PFLOTRAN truncated capillary pressure to a fixed maximum, here described as a flat extension. By bounding the derivative of capillary pressure to finite values, the flow is similarly limited to finite values. However, this introduces a new discontinuity in the model as the slope is discontinuous at the corner.

This is particularly problematic when using Newton's method to solve the system of nonlinear equations, as Newton's method requires dividing by the derivative. Thus, if the derivative is or approaches zero, the next iteration may be driven to a wildly incorrect saturation.

In the approach taken by Sun *et al.*[6], a piecewise but smooth extension to capillary pressure resolves the instability using Newton's method. This was the initial approach taken here as well for PFLOTRAN for capillary pressure. However, this work deviates in the treatment of relative permeability as, by similarly extending the relative permeability models below residual saturation, liquid-phase flow will continue below residual saturation.

The next two sections will detail the justification for both decoupling the value for residual saturation for relative permeability and capillary pressure, as well to support eliminating the assumption of infinite capillary pressure at the capillary pressure residual.

Relative Permeability Models

As seen in equation 1, permeability is essentially the coefficient relating the flow rate of a fluid in a non-equilibrium system with its driving potential. While models based on bundles of tubes in Hagen-Poiseuille flow can be drawn for single-phase flow, for multi-phase flow, the

relative permeability represents the averaging behavior over many discontinuous regions[12].

Measuring relative permeability directly can be experimentally demanding and, rather than making direct measurements at numerous degrees of saturation, only the saturated liquid or intrinsic permeability is often measured – or estimated – directly. The permeability of each phase is then calculated relative to this using an additional model, dependent upon the capillary potential model. Two such models implemented in PFLOTRAN are the Burdine[13,14] and Mualem[2] models.

In the development of the first model, Burdine *et al*. first performed detailed characterizations of petroleum reservoir rocks[13] and later developed a conceptual model for estimating the relative permeability by summing over the volume, area, and tortuosity of the fluid filled pores[14]. To do so, Burdine made a simple linear interpolation between residual and full saturation to represent the "relative" tortuosity of the wetting phase, that is $X_{rw} = \frac{S_w - S_r}{1 - S_r}$. This relative tortuosity is mathematically equivalent to the term effective saturation S_e.

However, the relative tortuosity is distinct from the intrinsic tortuosity, that is the tortuosity of a fully saturated medium. Additionally, while narrow dead-end pores appear inside the Burdine summation, if they lack connectivity, they do not contribute to the resulting permeability in the original Burdine model.

However, detailed pore size and tortuosity distributions are rarely available. Rather, it is assumed that all pores have equal tortuosity as a matter of convenience, and the diameter of filled pores at a given saturation are modeled using the Young-Laplace equation. Assuming cylindrical pores, this can be expressed as $r = \frac{2\gamma}{p_c}$, where γ is the surface tension. In doing so, the so-called Burdine equations result if the summation is replaced with integration[9].

$$k_{rw}^{Burdine} = \left(\frac{S_w - S_r}{1 - S_r}\right)^2 \frac{\int_{S_r}^{S_w} \frac{dS_w'}{p_c^2}}{\int_{S_r}^{1} \frac{dS_w'}{p_c^2}} \tag{5}$$

Later, Mualem[2] took a different approach in deriving a model for relative permeability that more clearly demonstrates it is allowable for small pores to be neglected. That is, when Mualem substituted the Young-Laplace equation to convert the variable of integration from pore diameter to saturation, the limits of integration in terms of pore diameter were explicitly non-zero:

$$k_{rw}^{Mualem} = S_e^{\eta} \left[\frac{\int_{r_{min}}^{r} rf(r)dr}{\int_{r_{min}}^{r_{max}} rf(r)dr}\right]^2 = S_e^{\eta} \left[\frac{\int_{S_r}^{S_w} \frac{dS_w'}{p_c}}{\int_{S_r}^{1} \frac{dS_w'}{p_c}}\right]^2 \tag{6}$$

Thus, in the original formulation from Burdine and Mualem there is a finite capillary pressure at the residual, p_{cr}, that corresponds to the minimum pore diameter r_{min} that contributed to permeability. That is, $p_{cr} = \frac{2\gamma}{r_{min}}$. The value of residual saturation S_r can then be viewed in terms of the saturation that predicts this capillary pressure. That is $p_{cr} = p_c(S_r)$. Thus, for any non-zero minimum pore diameter, the capillary pressure must be finite.

Capillary Pressure Models

As such, we require a capillary pressure model can satisfy the equation $P_{cr} = P_c(S_r)$. At the same time, modeling the capillary pressure in porous media is complex proposition due to the heterogeneity in both pore sizes, and additionally surface chemistries and thereby wetting angles. Consequently, it is most often defined by empirically fitting simple functions to experimentally measured values.

In this work, two frequently utilized but similar empirical approaches for capillary pressure will be considered. The first is the Brooks-Corey[15], which applied the concept of the power law distribution of pore sizes to capillary potential.

$$S_e = \left(\frac{p_b}{p_c}\right)^{\lambda} \tag{7}$$

Where p_b is air entry pressure and λ is the pore size distribution constant. The effective saturation S_e is mathematically equivalent to the "relative tortuosity" of Burdine. This assumption allows the integrals inside the Burdine equations to evaluated analytical if and only if capillary pressure is allowed to approach infinity, neglecting the presence of dead-end pores or thin films.

Later, Van Genuchten[1] expanded on this model to allow for a second power law distribution exponent for both large and small pores.

$$S_e = \left(\frac{1}{1 + (\alpha P_c)^n}\right)^m \tag{8}$$

Where m, n, and α are empirical parameters dependent on the material. Again, the effective saturation is assumed commensurate with the relative tortuosity and this model too ignores the existence of dead-end pores, requiring capillary pressure to approach infinity at the residual.

Of course, dead end pores and thin films exist, and isolated pockets of water are retained in porous media. Residual water is associated with dead-end pores that have a non-zero radius and thereby a finite, no matter how large, capillary pressures. Adsorbed films also exist, though the Young-Laplace equation does not model these. While it is analytically convenient residual saturation to be commensurate in the relative permeability and capillary pressure models, it is not physically realistic to do so.

There are, however, experimental challenges in measuring capillary pressure below residual. Gravitational drainage is generally not enough, and some additional water

may be removed by centrifugal methods. However, flow below this threshold will be dominated by film flow or vapor phase transport. As originally suggested by Muskat and Meres[7], liquid transport below the residual should be modeled using source/sink terms in the mass balance, rather than via *ad hoc* extensions to the Darcy model. While the relative permeability of the gas phase approaches a maximum at residual liquid saturation, transport of vapor may be reduced at saturations below residual by additional vapor pressure lowering.

To date, PFLOTRAN has been programmed with the analytic expressions from Brooks-Corey and van Genuchten, and while residual values are allowed to differ in the two expressions, the resulting relative permeability is not consistent with either equations 5 or 6. Provided that the piecewise extension to the capillary pressure model occurs at a large capillary pressure, the impact on relative permeability is a minor perturbation as it is inversely proportional to square of capillary pressure.

RESULTS

Allowing the residual saturations to be dissimilar and extending the capillary pressure below residual was found to be sufficient to resolve the numerical instability in the UZ model. Using a reference case with 24 pressurized water reactor spent fuel waste packages, simulations were conducted on Sandia National Laboratories Skybridge cluster, which hosts Intel Xeon processors (E5-2670 2.60 GHz, 20 MB L2 caches). Using the conventional Newton solver and the truncated or flat capillary pressure, the simulation failed to complete in 48 hours. Using either the linear or exponential extensions, such simulations completed in 33.8 and 31.8 minutes respectively.

In addition to modifying the physical model, recently, a more robust numerical method, Newton Trust Region Dogleg Cauchy, was implemented into PETSc to be utilized by PFLOTRAN[16]. This algorithm, the UZ simulation with the capillary pressure extension completed despite the discontinuity in the capillary pressure derivative. It nevertheless requires considerably more computational resources (551.0 minutes). Combining NTRDC with smooth capillary extensions, but the original relative permeability expression provided the highest performance, with simulations requiring only 19.4 or 18.3 minutes., reducing computation time by approximately 42% over the traditional Newton solver.

CONCLUSIONS

In this work, the basis for the decoupling of residual saturation for relative permeability from the residual saturation for capillary potential has been presented. While previously described as an *ad hoc* approach, this approach is supported by the original works of Burdine and Mualem. Rather, the need for an infinite capillary pressure appears as a matter of convenience in the Brooks-Corey and van Genuchten models.

In doing so, the cessation of liquid flow at residual saturation can be adequately modeled in terms of relative permeability alone. This permits capillary pressure to be described by any number of extensions to the empirical models below this relative permeability residual. While more robust numerical algorithms can resolve capillary pressure models with discontinuities in slope, it is nevertheless advantageous to use smooth extensions where possible.

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