Stochastic pore-scale growth models of DNAPL migration in porous media

Robert P. Ewing a, Brian Berkowitz b,*

a Department of Agronomy, Iowa State University, Ames, IA 50011, USA
b Department of Environmental Sciences and Energy Research, Weizmann Institute of Science, Rehovot, Israel

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Abstract

Stochastic models that account for a wide range of pore-scale effects are discussed in the context of two-phase, immiscible displacement problems in natural porous media. We focus on migration of dense, nonaqueous phase liquids (DNAPLs) through water-saturated geological materials. DNAPL movement is governed by buoyancy, capillary, and viscous forces, as well as by the details of the porous medium. We examine key issues relevant to development of stochastic growth models. We then present a particular stochastic growth model, based on a generalization of invasion percolation and Eden growth approaches, which realistically simulates two-phase flows in a computationally efficient manner. Fingering patterns are shown to depend critically on the competing buoyancy, capillary, and viscous forces between the DNAPL and the water, and their interactions with the porous medium at the local scale. We conclude with recommendations for future research. © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

Characterizing and quantifying the migration of immiscible liquids in geological formations has been the subject of intense investigation over many decades. Initially, attention was focused on secondary recovery methods in the petroleum industry, where injected water and/or other liquids are used to displace oil. More recent study of two-phase immiscible flows has been motivated by the need to treat spills and leaks of so-called non-aqueous phase liquids (NAPLs), such as trichloroethane (TCA), trichloroethylene (TCE), and benzene, which can severely reduce the quality of subsurface water supplies. Although NAPL solubilities in water are generally low, their toxicity is such that NAPLs that leak into an aquifer and dissolve slowly over long periods of time will contaminate large volumes of water. Clearly, there is a strong need for realistic simulation models with predictive capabilities, both for application to problems of pollution remediation and prevention as well as for oil recovery.

In the case of secondary recovery in petroleum reservoirs, water displacing oil is generally an imbibition process, if we assume that the rock is predominantly water-wet. In contrast, the advance of a dense NAPL (DNAPL) into a previously water-saturated porous medium is a drainage process, in that a nonwetting fluid displaces a wetting fluid. Generally speaking, the initial downward migration of a DNAPL is due mainly to gravity, whereas the lateral spreading usually results from capillary resistance to entering layers of lower pore-size material, such as clay lenses. The interactions among the capillary, buoyancy, and viscous forces and the details of the porous medium can result in a complex flow pattern characterized by ramified features that range in size from the pore scale to tens of meters or more.

The physics of two-phase, immiscible fluid movement in porous media are well understood under simple conditions e.g. [1,2,11,20]. Realistic simulation in well-defined, continuum-scale media has been achieved using macroscopic multi-phase flow equations (e.g. [1,2,29–31]). The simulations generally succeed in reproducing the finger-and-pool configuration observed in some DNAPL spills (e.g. [9]). Despite being 2-D, however, these continuum-based simulation models are generally cpu-intensive. More significantly, DNAPL flow patterns are often highly ramified and sensitive to small-scale
details. As a consequence, these models are severely limited because use of a coarse grid will likely yield meaningless results, but the memory requirements and computation times required for finer grids are prohibitive. These computational difficulties become more marked in full 3-D simulations, which are necessary to properly capture realistic flow patterns.

More recent alternatives to these macroscopic approaches to describing multi-phase flows have been developed on the basis of stochastic growth models (e.g. [14,18]). These models seem particularly promising because of their inherent ability to account for the dynamic, and often unstable, interaction among capillary, buoyancy, and viscous forces, along with the high sensitivity to pore-scale structure. Stochastic, discrete growth models are based upon frameworks such as invasion percolation (IP) and diffusion-limited aggregation (DLA). Such models capture the essential physics of a process, and migration patterns can be simulated without the need to solve large systems of equations. Simple growth models that mimic physical growth processes are known for specific instances, such as viscous fingering [34,40], capillary fingering [34,50], and gravity fingering [21,37]. A broad review of stochastic and continuum two-phase flow models is given by Sahimi [41]. Note that in some physical situations (e.g., slow capillary-driven processes), large-scale behavior is controlled by small-scale mechanisms, whereas in other cases (e.g., high pressure injection of an oily phase), small-scale behavior is controlled by large-scale mechanisms. In general, models that use local rules (as do most stochastic growth models) will work better when local mechanisms dominate, so conditions that are controlled by small-scale mechanisms will play to the growth model’s strength, whereas the latter one will not.

In this contribution, we first discuss the application of various stochastic growth models to modeling two-phase, immiscible displacements in geological porous media. We focus on drainage-type processes relevant to the migration of DNAPLs. Our purpose is to examine current growth models to identify places where they fail to realize their potential as useful predictive tools. We then present a particular growth model, based on a generalization of invasion percolation and Eden growth approaches, which realistically simulates DNAPL migration in a computationally efficient manner. We conclude with a discussion of future research directions.

2. Stochastic growth models for DNAPL movement

Invasion percolation was developed as an analog to quasi-static capillary displacement [50], and drying (or drainage) processes in particular. DNAPL migration is certainly a drainage process at the advancing front, but the contributions of viscous and buoyancy forces demand that a growth model more complex than simple IP be used. In addition to considerations as fundamental as which growth model and conceptual framework to use, other complications are encountered in bringing the model from an abstract concept to a real-world application. In this section, we discuss some of the challenges of developing percolation-based DNAPL models and examine how these challenges may be addressed.

2.1. Stochastic growth models: background

The patterns formed by immiscible displacement of a wetting fluid by a nonwetting fluid in the absence of buoyancy forces fall into three regimes [34], with an intermediate transition zone that links them. These regimes are defined as regions in a 2-D “phase diagram” (Fig. 1(a)), with one axis being log(M) (M ≡ viscosity ratio = μ_d/μ_r, where μ is viscosity, r the resident fluid, and d is the displacing fluid), and the other log(Ca) (Ca ≡ capillary number = μ_d/V, where V is velocity of the displacing fluid, and γ is interfacial tension). At high viscosity ratios and high capillary numbers, stable displacement is found (Fig. 1(a)). At high M and low Ca, capillary-driven fingering occurs, whereas at low M, viscous fingering is seen. Significantly, these three flow regimes each correspond to a specific stochastic growth model: IP is an analog of capillary fingering [50], stable growth is mimicked by anti-DLA [36,40], and DLA is similar to viscous fingering [40,52]. Some researchers (e.g. [14]) have used Eden growth [13] rather than anti-DLA to represent stable growth; we shall address this issue later.

As discussed in the following section, in order to be applicable to DNAPL migration, growth models must account for density differences between liquids in addition to dealing with viscous and capillary forces. The experiments of Lenormand et al. [34] were conducted in flat, horizontal model porous media, so gravity effects were negligible. But, for DNAPLs invading a porous medium that is not 2-D and horizontal, buoyancy forces can affect the stability of the interface (e.g. [17,21,25,51]). For example, a light NAPL (LNAPL) injected at the top of the medium would tend to have its interface with the aqueous phase stabilized by buoyancy [27], whereas a DNAPL injected at the top would have its interface destabilized [17]. As such, one can consider that Lenormand et al.’s [34] phase diagram could be extended to include a third dimension describing buoyancy forces (Fig. 1(b)). Position in this dimension is given by the bond number (Bo = grl/Δρ/γ, where g is the gravitational acceleration, r the characteristic pore radius, l the characteristic distance between pores, and Δρ is the fluid density difference). The usual convention is that Bo > 0 stabilizes the interface, whereas Bo < 0 destabilizes it. This is reflected in the diagram by the stable regime growing larger with positive bond numbers and
shrinking with negative ones. The viscous fingering regime will likewise change size according to the bond number and the critical velocity, as given in Chuoke et al. [11]. Yortsos et al. [54] have developed a stable/unstable flow phase diagram for immiscible displacement which also includes the influence of the bond number on stabilization and destabilization. Combining this work with that of Lenormand et al. [34] may better define the 3-D stable and unstable displacement regions.

The literature on the stochastic growth models is large and well developed, and so here we will not dwell on their descriptions. For more detailed discussions of these different growth models, we refer the interested reader to the original articles referenced in the preceding paragraph in addition to, e.g., Stauffer and Aharony [47] for percolation in general and Barabási and Stanley [4] for Eden and DLA. Briefly, the IP algorithm involves defining a medium as a regular or irregular grid, with each site being randomly assigned an “invasion probability” between 0 and 1. For each invasion step, the interface along the invading boundary is scanned for the site with the highest probability; then this site is invaded (Fig. 2(a)). The DLA algorithm, on the other hand, begins with a “seed”, which can be a point, line, or other shape. “Sticky” particles are then released from random locations at a distance from the interface and diffuse freely until they touch the seed, whereupon they stick to it and become part of it (Fig. 2(b)).

Eden and anti-DLA algorithms lead to patterns that initially look similar, but over time anti-DLA remains stable while Eden develops features. Historically, Eden was developed as an analog for bacterial growth [13], and anti-DLA for stable flow [40]. In terms of implementation, Eden scans the interface and chooses one of the interface sites at random; in contrast to the IP algorithm, all sites are equally likely (Fig. 2(c)). Anti-DLA, on the other hand, starts a random walker at the DNAPL source and allows it to diffuse to the interface, where it sticks and becomes part of the DNAPL (Fig. 2(d)). The result is that Eden is more unstable because parts of the interface that protrude slightly have extra interfacial area and so a slightly higher chance of growth. Conversely, with anti-DLA, interface sites along a protuberance are at a greater distance from the source, so random walkers that start at the source are more likely to encounter closer sites than those at the end of the protuberance, thus dissipating instabilities or perturbations.

Generalizations and combinations of these and other growth processes have been attempted for several years. In physics, Martín et al. [35] noted that the Eden stable growth model and IP are essentially extremes of a continuum, and the transition between them can be accomplished by using an exponent to bias a pseudorandom selector toward zero (for IP) or unity (for Eden). Similarly, Smith and Collins [44] hypothesized that the Eden and DLA models could be generalized to a single model using a method based on variable diffusion lengths, with large steps corresponding to DLA and infinitely short to Eden. The next year, Siddiqui and Sahimi [43] developed a model that interpolated smoothly from Eden to DLA. In petroleum engineering, Leclerc and Neale [33] used a Monte Carlo method that transitioned among three models (IP, DLA, and anti-DLA).
anti-DLA) using probabilities, based on values of $M$ and $Ca$, to select a random walker type and a sticking probability. Their model produced excessively stable behavior at very low $Ca$ and did not properly predict the stable to capillary transition, but otherwise showed good agreement with experiments. The low $Ca$ problems were partially corrected by Kiriakidis et al. [28], but results were still not close enough to experimental behavior. In geophysics, Stark [46] modeled patterns of plateau erosion by interpolating among three models (IP, Eden, DLA). Stark’s [46] algorithm assigned an original strength to each point (as in IP), then used random walkers to weaken perimeter sites by a factor $g$, as well as Eden-like selection to weaken perimeter sites by a factor $h$. The choice of internal or external walkers (stable flow or viscous fingering) and the balance between $g$ and $h$ shifted the process among the different growth models. As an aside, we note that in DLA, anti-DLA, and Eden as originally formulated, the medium is implicitly assumed to have no effect, so medium effects are essentially ignored.

Some researchers (e.g. [14,46]) have used the Eden growth model [13] for simulating stable flow, despite Paterson’s [40] and Lenormand et al.’s [34] identification of stable flow with anti-DLA. The Eden model is likely chosen because generalizing across Eden and IP is much more straightforward than generalizing across anti-DLA and IP. As discussed previously, however, Eden and anti-DLA do not behave identically: anti-DLA resists perturbations in the front just as viscous-stabilized flow does, whereas the Eden model tends to propagate them. Other researchers (e.g. [18,49]) have used DLA to model flow that is primarily capillary-driven. Although the fractal dimensions of the resulting patterns are similar, which may lead the human eye to initially perceive the fluid patterns as similar, the driving physics are different, as are the resulting patterns and often their topologies. The key issue here is that the model should closely follow the relevant physics of the process of interest: it is not sufficient to simply reproduce the essential features of the process being simulated.

![Fig. 2. Schematic illustration of typical migration patterns generated by different stochastic growth models: (a) IP; (b) DLA; (c) Eden; (d) anti-DLA.](image-url)
2.2. Complicating issues in stochastic growth model development and application

In this section, we identify and discuss six factors that contribute to the complexity of modeling DNAPL movement in the real world: buoyancy, viscosity, sensitivity to local detail, geological uncertainty, partial saturation, and dynamics. These factors complicate both continuum scale and stochastic growth models, but our focus here is on the stochastic growth models. A seventh factor, capillarity, poses difficulties for continuum models but is built into invasion percolation-based models at a fundamental level, so we will not discuss it here. Our purpose in this section is to summarize areas of needed future work.

2.2.1. Buoyancy

Most NAPLs have a density different from that of water, so that in addition to capillary forces, buoyancy forces are exerted on NAPLs and affect their movement. At first glance this is not a problem: percolation concepts can be adapted by assigning an invadability value to each site that accounts for buoyancy as well as capillarity (see, e.g. [21,37]), such that the effective invadability $I_{\text{eff}} = I_0 + \Delta \rho gh$, where $I_0$ is the intrinsic (capillary) invadability and $h$ is the vertical distance from the source. But this approach assumes no viscosity (a basic assumption of invasion percolation), which can cause notable deviations from the equation. For example, a DNAPL finger migrating downward at nonzero velocity does not exert the full force given by that simple equation, because some of the pressure difference is lost by viscous drag within the finger. Similarly, where the DNAPL encounters and spreads along the top of a confining layer, viscous forces cause the DNAPL to accumulate into an inverted bowl shape with nonzero depth [19]. In other words, the assumption of no viscosity implies that simple hydrostatic forces dominate everywhere, unaffected by viscosity, which is clearly an extreme. We propose an alternative extreme: by allowing for viscosity effects, we assume that DNAPL height effects do not propagate unaltered through the entire DNAPL-invaded region. Specifically, we can assume that the hydrostatic force at the bottom of a pool feeding a finger is that given from the top of the pool (viscous drag through the pool itself is negligible). For this alternative, buoyancy force calculations should be updated dynamically as the configuration changes. The two extremes will serve to constrain the solution and point toward a more reasonable middle ground.

A further complication is that buoyancy forces have a different sensitivity to length than do capillary forces [27]. To put it simply, if the grid is made too coarse, then the buoyancy forces driving invasion of a site one level lower will overwhelm any capillary considerations. Specifically, the vertical grid size $l$ at which buoyancy forces would equal capillary forces is approximately $l \sim P_c / g \Delta \rho$, where $P_c$ is the capillary pressure at breakthrough [27]. Because $l$ should be much smaller than this, the vertical grid spacing must be relatively small, which may greatly increase memory requirements for a realistic 3-D simulation. An alternative approach would be to use the calculated invadability when comparing sites on the same level but to moderate local buoyancy effects by a much smaller $l$ when invading either upward or downward. To our knowledge this approach has never been used, but it offers a potential way to avoid being forced into impractically fine discretization.

2.2.2. Viscosity

Viscous forces affect the configuration of an advancing front [34], but because invasion percolation assumes zero viscosity, viscous forces are not explicitly accounted for by a pure invasion percolation model. As discussed in Section 2.1, invasion percolation can be blended with one or more other growth models to account for viscous forces. For high viscosity NAPLs like coal tars, viscous fingering effects may be safely ignored. However, low viscosity NAPLs such as carbon tetrachloride or chloroform require a model that includes DLA behavior. A general growth model must be able to handle both high and low viscosity ratios.

However complex it may be to blend growth models, it is only the first step: the role of viscous forces (as quantified by the capillary number, for example) changes with time, position, and interactions between them. Time, because if a DNAPL is spilled at a constant rate, the front will be smaller initially than later, and so the average velocity of the interface will decrease, decreasing the average capillary number. (Of course, if the spill does not proceed at a constant rate, that also will change the capillary number.) Position, because the capillary number at the tip of an advancing finger is clearly greater than that along its static midsection, which is essentially functioning as a conduit. And interactions, because DNAPL movement may oscillate as pools fill and empty, causing finger midsections to break via snap-off, then re-form as pressure builds up behind them [19], with threshold pressures possibly changing due to differences in advancing and retreating contact angle. Modeling of these oscillations may be avoided if the objective is to determine where the DNAPL has been, rather than its configuration at any given instant. In other words, if the model does not allow oscillations, it is less realistic but it is likely to be faster, and the results may be identical from the point of view of remediation. But even without modeling oscillations, viscous forces require the modeler to constantly update a changing invadability in addition to tracking a constantly changing accessibility.
2.2.3. Sensitivity to local detail and upscaling considerations

Invasion percolation was developed to mimic pore-scale properties, and the capillary forces that dominate DNAPL movement in medium- and fine-grained materials operate at the pore scale. Nonetheless, it is of course impractical to model an aquifer at the pore scale. Several authors (e.g. [15,21,32,53]) have used a form of upscaling to move percolation models to larger scales than a few pores, and aside from the concerns about grid size raised by Ioannidis et al. [27], mentioned earlier, upscaling percolation theory appears to work well. We note in passing that upscaling a pore-scale model raises some question as to what exactly constitutes pore-scale modeling: if scale alone is the determining factor, then a finite-element model could presumably be made pore-scale by simply reducing the size of the elements. However, we believe the key point is that what are called pore-scale models treat the medium as being composed of discrete pores rather than being a continuum; when they are also at or near the pore scale, it is convenient to call them pore-scale models.

Features smaller than the grid size, e.g., a thin clay layer, may not show up in a coarsely discretized model but will stop DNAPL migration in actuality [19]. Conversely, a small fracture that crosses an otherwise impermeable layer will provide an ample pathway for migration [48]. Sensitivity to detail implies that, given absolute knowledge of the medium, finer resolution will always lead to more accurate simulations, whether one is using a continuum-based or a stochastic growth model. Because accuracy is always desirable, modelers will likely be tempted to work near the memory limitations of their computers, and so memory limitations will frequently be a limiting factor for both kinds of models. One approach that has not yet been taken by any percolation modelers (to the best of our knowledge) is to use an adaptive memory scheme with finer grids at the interface and coarser grids where detail is not needed. The implementation is difficult, but it would greatly reduce the memory overhead and may thereby speed execution. In the end, however, cpu time is more likely than memory to be the limiting computer resource, because inversion of the large sparse matrices that result from high resolution 3-D simulations requires far more cpu time than do stochastic growth processes.

2.2.4. Geological uncertainty

All models of DNAPL movement, be they continuum or percolation based, are limited by geological uncertainty. At best, the model is only as good as the data it is given, and geological information is known only sketchily. Worse, many kinds of relevant data are not known at all. For example, DNAPL movement is locally sensitive not only to porosity and conductivity, but also to contact angle [8], pore shape, pore roughness, and so on. These parameters are in turn functions of position in the profile (which influences weathering and organic matter content) and lithology, among other factors. Because the data requirements are so high and what is known so sparse, many of these parameters are simply assumed to be homogeneous. This means that the model is run within a virtual porous medium created by a combination of data, inference, assumptions, and stochastic generation. Because both uncertainty and sensitivity to detail are so great, a single simulation has a very low chance of correctly predicting the DNAPL disposition.

As suggested by Borchers et al. [7], a more useful approach is to generate many realizations of the porous medium and run the DNAPL movement simulation on each to obtain probabilistic estimates of the DNAPL location. This Monte Carlo approach is a very powerful way of handling geological uncertainty. But, the price of that power is the need for many realizations, which requires a model that executes rapidly. This can be partially dealt with by a coarse-grained parallelism: one can run simulations in parallel on many different machines. But a phenomenological growth model such as modified invasion percolation has the advantage of being much more abstract than either a continuum model with its multiple linked partial differential equations, or a pore-scale model that handles each of the many possible pore-scale events explicitly. Stochastic growth models are fast to the extent that they remain abstract, but accurate to the extent that they remain true to all the relevant physics; this tension is part of the challenge of modeling.

2.2.5. Partial saturation

DNAPL movement below the water table is a two-phase phenomenon, but above the water table it involves three phases. Because DNAPL leaks and spills generally start above the water table, and LNAPLs “perch” on the water table, it is important to consider how the presence of a third phase affects the NAPL behavior. The interaction of air, water, and NAPL is much more complex than that of water and NAPL only, with both apparent wettability [45] and pressure-saturation–conductivity relationships [16] being sensitive to the order in which fluids occupy each pore. (We note in passing that this three-phase nonlinearity is a major consideration in continuum models as well; see, e.g. [38].) In addition, buoyancy forces acting to move the DNAPL downward are affected by the degree of saturation, with the effective density of the resident fluid being (to a first approximation) the mean of the densities of the air and water as weighted by the local degree of saturation. Meanwhile, local saturation itself is a function of (at least) pore size, local wetting angle, height above the water table, and wetting history, and the rewetting that this involves is not a trivial matter [6,16].
As discussed in Section 2.2.4 this is an area that can quickly become too complex to model with reasonable speed, and percolation-based NAPL models aimed at practical application will need to find ways to abstract these complexities into simpler patterns if they are to succeed.

2.2.6. Dynamics

The aquifer into which the DNAPL moves is not a static system. Even aside from the DNAPL’s influence, the water table may rise and fall, and the water may flow. The medium is likely to have hysteretic water retention properties, so the history as well as the current height of the water table will need to be known. Schwille [42] performed experiments in which DNAPL was injected with a high water table, which was subsequently lowered. He also indicated that water flow had little effect on DNAPL movement, but at the least it will influence ganglion dissolution rates. To our knowledge, no one has yet replicated these experiments either quantitatively or in a simulation model, suggesting that changes in the water table height and flow in an aquifer represent areas of poor current understanding.

Once the DNAPL comes into contact with water, further changes will take place. The DNAPL will slowly dissolve into water, changing the interfacial tension as well as the density and viscosity of water. Likewise, water will slowly dissolve into the DNAPL, changing its properties. Because these changes are partially a function of the time of contact between the two phases, fluid properties, such as interfacial tension, density, and viscosity of both fluids, and contact angle, may change locally over time. In fact, they will likely even differ at different points along a single finger. Finally, the oscillations mentioned previously (Section 2.2.2) may also occur, further complicating the movement.

When the DNAPL source is removed, the pressure at the injection point changes and the DNAPL will retreat downward, generating ganglia by snap-offs as buoyancy-induced pressure is reduced. A complete DNAPL movement model will need to inform us where we will find DNAPL in pools, where in ganglia, and where in intact fingers. Then, after the initial DNAPL migration has occurred and the ganglia are apparently stable, slow dissolution of DNAPL into the water may eventually remobilize them [3]. Experimentally, these dynamic effects are generally avoided by holding the water table constant (e.g. [26]), pre-equilibrating the water and DNAPL (e.g. [23]), and collecting data only up to the point of breakthrough. Real spills are not so convenient, however, and eventually experiments will be needed to assess the magnitude of the effect of these dynamics on the eventual DNAPL disposition so that models can be altered as needed.

3. An in-depth example of a generalized growth model

The following model exposé is not given with the claim that the model fully addresses all issues raised above. We examine this model because it is one of the few stochastic growth models being developed, and because we are quite familiar with it. Both new developments and deficiencies in the model are examined in this discussion.

Berkowitz and Ewing [5] noted that the phase diagram relating viscous, capillary, and stable flow regimes should have a third dimension to account for buoyancy forces (Fig. 1). This implies that the growth models must allow modification or biasing to account for gravity stabilizing and destabilizing. Onody et al. [39] accomplished this through the use of four adjustable parameters that controlled the bond number, the downward bias due to gravity, the local change in capillary forces due to curvature of the interface, and the number of simultaneous invasions (which allowed multiple fingers to form). Their model required a data structure that identified fingers as distinct entities, but finger dynamics such as splitting and sensitivity to local details appeared to be lacking.

We [14] blended the IP and Eden models similarly to Márton et al.’s [35] model. The IP portion of the model involves assigning an invadability I to each cell, using a function of capillary forces with buoyancy-biasing (e.g. [21])

$$I = g(\rho D_p h + h\Delta \rho) - 2\gamma \cos(\theta)/r,$$

where \(g\) is the gravitational acceleration, \(\rho\) the NAPL density, \(D_p\) the depth of NAPL ponding above the water table, \(h\) the vertical position (positive downward with \(h = 0\) at the water table), \(\theta\) the local contact angle, and \(r\) is the local mean pore size. The Eden portion involves making a stochastic selection of what interface cell to invade, rather than always invading the most invadable as in normal IP. As with Márton et al. [35], the model can be biased toward the IP and/or Eden ends of the continuum by generating a pseudo-random number \(R\), raising it to an exponent \(W\), and choosing the cell in the interface list whose invadability-weighted position in the sorted list corresponds to \(W\). To obtain multiple fingers with merging and splitting behavior, as often seen in laboratory and field experiments, the model used an intermediate stochastic step that first stochastically selected among the continually changing number of individual fingers, then chose a cell within that finger. Eq. (1), combined with a bi-scale (finger and cell) adaptation of Márton et al.’s [35] biased selection procedure, is the core of the Ewing and Berkowitz [14] model.

The capillary number’s effect on the flow regime was controlled through stochastic weighting of the exponent \(W\), which served to bias the selection of fingers and
cells at each time step. Time was explicitly accounted for by assuming that the flow rate at the source was constant, and calculating the time required to NAPL-saturate one cell given the capillary number at the injection point. A more detailed model might include partial saturation capabilities, but our model currently assumes a given location is either water- or NAPL-saturated. The resulting model is the only percolation-based NAPL migration model to date that can span a range of capillary numbers and produce dynamic fingers. Given the improvement in model behavior when fingers were explicitly included in the model’s data structure, we speculate that even better simulations would be obtained if the data had a hierarchical, recursive pool and finger data structure instead of the current flat model.

Since the publication of Ewing and Berkowitz’s [14] model, we have implemented a trapping rule [12], based on a path-finding algorithm that determines whether the resident fluid has a possible escape route. Trapping occurs when the displacing fluid surrounds an “island” of the resident fluid. If the resident fluid is incompressible and the invasion process is assumed to proceed sufficiently quickly that film flow is negligible, then the island is considered trapped: it cannot be invaded. The decision of whether to implement the trapping rule in a given instance is therefore partially dependent on the time scale, and the two extremes – with and without trapping – should serve as constraints to intermediate cases. Additionally, if a NAPL invades an aquifer quickly but then stays in place for an extended time, film flow may be important. In such a case, examining the difference between invasion with trapping and without may give some hints as to where slow changes should appear. In 2-D, trapping causes holes to appear in the invaded region (Fig. 3), but does not by itself cause flow to have, for example, more or fewer fingers. However, given the history-dependent progression of IP, a small change in the invasion sequence may result in large differences in the final configuration.

We have also extended the model to the third dimension. This is important for several reasons. First, 2-D and 3-D flow patterns are different, partially because connected regions across the domain boundaries can co-exist for each of the two fluid phases in 3-D, but not in 2-D. Additionally, real-world problems are 3-D, and modeling a single 2-D slice of a 3-D site is not as useful, or potentially as accurate, as modeling the complete problem domain. Interestingly, trapping is less extensive in 3-D than in 2-D [53], but it can still be useful to be able to examine how important it is in any given situation. When modeling a quasi-2-D experiment, such as those of Hofstee et al. [26] or Glass et al. [19], it may actually be advantageous to simulate the process in 3-D, with the third dimension (thickness) being a few layers thick, because observations indicate that saturation is often not uniform from one face to the other [19]. Because these physical quasi-2-D experiments are, strictly speaking, 3-D, modeling them as 2-D is likely to introduce model artifacts that could be avoided in 3-D.

Finally, we have developed a procedure to readjust the capillary number in time and space based on local changes in the area of the interface (see Section 2.2.2). Capillary readjustment is performed because if the DNAPL/water interfacial area increases while the spill rate and the porosity remain constant, the mean velocity of the interface – and therefore of the invading fluid – must decrease. Because a decrease in velocity corresponds to a decrease in the capillary number, the capillary number should be continually updated to account for this change in velocity. Simply multiplying the injection capillary number by the proportional increase in interfacial area will not produce accurate results, however, because the velocity of the invading fluid is not uniform along the entire interface. A viscous-stabilized (high capillary number) invasion may expand fairly uniformly, but a slowly advancing DNAPL finger moves at the tip but changes little along the rest of the finger. In other words, the degree to which increases in interfacial area should be used in updating the capillary number, itself depends on the capillary number.

Our capillary readjustment algorithm therefore proceeds as follows. First, the capillary number from the previous time step \( t - 1 \) is used to calculate the stochastic finger weighting parameter \( W \)

\[
W = \frac{3}{4} + \frac{1}{(4P)},
\]

(2a)

where

\[
P = m + \frac{\log(Ca_{t-1}) + n}{n}.
\]

(2b)

The intermediate value \( P \) is simply a log-transformed capillary number normalized to the range 0–1 from the original range \( Ca = 10^{m-n} \), for \( m \approx 0 \) to \( -1 \) and \( n \approx 5-9 \). This original range corresponds to the range over which transition occurs from viscous-stabilized flow to capillary fingering [34], itself a function of lattice size and, likely, other variables such as dimension of the medium. In our simulations, we used \( m = 0 \) and \( n = 6 \), but we show the exponents as being variables to emphasize that their actual values may vary somewhat depending on the specifics of the system. Eq. (2a) is a strictly ad hoc formulation for relating the normalized capillary number to the behavior produced by Martín et al.’s biasing scheme. When \( P \leq 0 \), \( W \) is set to infinity, resulting in pure IP.

Once a finger \( F \) is stochastically selected using \( W \) (as detailed in [14]), the “effective” interfacial area \( A_F \) of that finger is calculated by summing the first \( I_F/W \) worth of entries in the sorted interface list, where \( I_F \) is the sum of the invadabilities of all entries in the list for finger \( F \).
It is evident that at low capillary numbers, $W$ will be large, and so $A_F$ will only be a small fraction of the number of entries in the list. In other words, at low flow rates, only a small fraction of the interface is actually moving. The effective area for $F$ is then used to calculate the effective area of the entire interface $A_i$.
The discussions so far have implied that the flow patterns are largely functions of the two fluids. However, the structure of the porous medium also exerts considerable influence in developing flow patterns. As an illustration, Fig. 4 shows invasion patterns for media similar to those in the previous figures, but differing in spatial arrangement. The imprint of the porous medium structure is evident in the DNAPL migration patterns, showing that the final pattern is the result of interaction of the porous medium with the capillary, buoyancy, and viscous forces.

A series of detailed experiments reported in [22,23] affords us an opportunity to compare our model results to experimental data. The experiments are a series of controlled DNAPL spills performed in a water-saturated, uniformly packed sand tank. Following DNAPL detection at the lower tank outlet, the DNAPL source was removed and the tanks were excavated in serial horizontal sections. The addition of red dye to the
DNAPL allowed discrimination of the DNAPL in photographs. In the current paper, we compare our results to their experiment IV, injection of TCE into medium sand. Held’s [22] results are statistical, in the sense that duplicate experiments may produce similar statistics with respect to (for example) saturation with depth, but would yield different specific fingering patterns. The photographic results are not strictly quantitative, however, because the DNAPL spill volume calculated from image analysis is approximately 200% of that measured probably due to their methodology; they [22] used a red filter, which masked much of the contrast between the sand and the red dye, resulting in a pixel brightness histogram approximately seven brightness values wide. Partially NAPL-saturated pores would frequently be interpreted as NAPL-saturated during thresholding, over-estimating the amount of NAPL in the image.

In Fig. 5, we present two photographs from Held [22], showing DNAPL configuration at 16.5 and 73.5 cm below the injection port, and results of one of our simulations of that experiment. Simulation inputs corresponded to experimental parameters in terms of fluid properties (DNAPL density, viscosity, contact angle, and interfacial tension), porous medium properties (porosity, pore size distribution, and a [assumed] random pore structure), size of the tank, size and location of the DNAPL injection port, and experimental parameters such as DNAPL head and injection rate. A listing of numerical values of simulation and experimental parameters, taken from the input file, is given in Table 1. There are no fitting parameters as such: numbers are either derived directly from the experiment (e.g., the NAPL density), or are arbitrary (e.g., the random number seed). Values for \( n \) and \( m \) do not appear in the input file; as mentioned earlier, we assume here that \( m = 0 \) and \( n = 6 \). Held [22] used a constant head upper boundary with an initially rising water table. The resulting DNAPL injection rates were nearly constant.

![Fig. 5. Comparison of Held’s [22] photographs of DNAPL at depths of 16.5 (a) and 73.5 (b) cm below the injection port, and simulated horizontal cross-sections at corresponding depths ((c) and (d)). Note that the photographs are at a higher resolution than the simulations, so that there is more detail in the photographs.](image-url)
over time, so for convenience we used a constant rate injection, also with an initially rising water table. Simulations used trapping and capillary readjustment and were performed on a 556 \times 144 grid with unit cell size of 1.8 mm \(^3\). Simulations required approximately 5 min on a 800 MHz Athlon processor.

The qualitative comparison between horizontal sections is quite good, indicating that the model captured the essentials of the DNAPL behavior. Examination of a side view of the simulation results (Fig. 6) shows that, as Held [22] stated, “A long stretched plume reached 35 cm down (below the injection point), where multiple fingers developed”. In agreement with Held’s [22] data, the region just below the injection point showed high saturation and stable behavior. Additionally, below about 35 cm under the injection point the number of fingers started to decline, with only a small number of fingers still advancing near the bottom. Quantitative comparison between experiment and simulation can be made using (inferred) saturation with depth, area/perimeter ratio, or fractal dimension [22], but the pattern itself is more difficult to quantify. A more quantitative comparison between Held’s [22] experimental results and our simulations will be presented in a future publication.

Strictly speaking, our model is not directly applicable to Held’s [22] experiment. Their experimental viscosity ratio, \( M = 57 \), is slightly less than unity, whereas our model is intended for \( M > 10 \). Moreover, there is no DLA component to our model. However, their viscosity ratio places the experiment in the transition region, but closer to the stable flow regime than to the viscous fingering regime, so little DLA-like behavior is to be expected. It is encouraging that the model captures the observed DNAPL movement, without adjusting an array of fitting parameters; all needed input parameters are specified in [22]. Examining the range of behaviors and robustness of the model, afforded by changing the random number seed, or the changes induced by (for example) changing the input capillary number, is beyond the scope of the current study. In this context, we stress that we are not adver-

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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<tr>
<td>Mean pore radius, m</td>
<td>0.00011</td>
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<td>Positive = gravity-unstable</td>
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<tr>
<td>Initial depth of water table, m</td>
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</tr>
<tr>
<td>Initial ponding depth of NAPL at surface, m</td>
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<td>Grid size, Y</td>
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<td>Grid size, Z</td>
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<td>Stop injection here. –1 denotes circular injection.</td>
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<td>Total volume of NAPL injected, m(^3)</td>
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<td>Random number seed</td>
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4. Future directions and perspectives

A consistent difficulty in evaluating both continuum-based and stochastic growth DNAPL flow models is that there is no straightforward way to validate a model. Proper validation requires a data set that is sufficiently detailed in both space and time that comparisons can be made at all stages of the simulation, not just at the end. In fact, comparison with many experimental results spanning a wide range of experimental parameter space is needed before we can have confidence that the model performs both correctly and generally. Currently available data fall into three general classes: micromodel studies (e.g. [24]), macroscopic experiments using an artificially structured porous medium (e.g. [26]), and macroscopic experiments using an unstructured porous medium (e.g. [23]). For the purposes of this article, micromodel studies are less relevant, so we will discuss only the macroscopic experiments. Laboratory studies are few in number (see, e.g. [10]) and fewer yet when one requires quantitative descriptions of the porous medium, the NAPL and the conditions under which it was injected, and the migration pattern. Structured medium experiments are useful for verifying that a model performs in a generally correct manner, but unless the medium is fairly intricate and very well characterized, a single dataset is of limited value because it tests so few aspects of the model. Unstructured media, meanwhile, will produce somewhat random patterns due to sensitivity to unknown local details. It is difficult to find meaningful ways to compare experimental results to simulations, effectively comparing a single physical Monte Carlo realization to many virtual ones. Comparisons between experimental and model results, across many depths, of the number of fingers, NAPL-saturated area, and NAPL area/perimeter ratios give some indication of how well a model performs, but because of the random nature of the medium it is not useful to be too strict in the comparisons.

The most useful experiments will be those that use intricately constructed, well-characterized media and quantitatively captured saturation data over time. To our knowledge, the only data that meet these criteria are from the recent experiments of Glass et al. [19]. Their medium was designed to test several hypotheses concerning capillary barriers, and data were collected at frequent intervals and analyzed to yield NAPL saturation as a function of position and time. It is our hope that more experiments of this caliber will be performed and published, as they offer a more stringent and quantitative avenue for model validation than has heretofore been available.

To summarize, percolation-based models of DNAPL movement have much to recommend them over models based on partial differential equations, in terms of conceptual foundation, speed, and potential for accuracy.
However, we identify five principal ways in which percolation-based NAPL movement models need to advance. First, the overly simplistic invadability equation, by which invadability increases linearly with depth, needs to be modified dynamically to account for the NAPL's common pool/finger structure and the viscosity of the NAPL. Second, ways must be found to blend the anti-DLA and IP models so the correct process model is used for stable flow. Third, models should be expanded to function properly in the unsaturated zone and where the water table is fluctuating, even incorporating three-phase flow dynamics as needed. Fourth, the number, quality, and availability of data sets for model validation must be increased. And finally, modelers should work with computer scientists to overcome some of the speed and memory issues that are already evident, and which could eventually delay or prevent practical application of the models.

Acknowledgements

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References

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