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Counter-current imbibition and non-linear diffusion in fractured porous media: Analysis of early- and late-time regimes and application to inter-porosity flux

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ABSTRACT

The displacement of non-aqueous phase liquid (NAPL) in permeable porous rock by water saturating the surrounding fractures is studied. In many situations of practical interest, capillarity is the dominant driving force. Using the *global pressure* concept, it is shown that the water saturation is driven by a generic non-linear diffusion equation governing the matrix block counter-current spontaneous imbibition. In addition, in most cases, the saturation-dependent diffusion coefficient vanishes at the saturation end points, that renders the driving equation highly singular.

In this paper, two exact asymptotic solutions valid for short and long times are presented, under the assumption that the conductivity vanishes as a power-law of both phase saturations at the extreme values of the fluid saturation.

Focusing on the late-time domain, the asymptotic solution is derived using an Ansatz that is written under the form of a power-law time decay of the NAPL saturation. In the spatial domain, this solution is an eigenvector of the non-linear diffusion operator driving the saturation, for a problem with Dirichlet boundary conditions. If the diffusion coefficient varies as a power law of the NAPL saturation, the spatial variations of the solution are given analytically for a one-dimensional porous medium corresponding to parallel fracture planes. The analytical solution is in very good agreement with results of numerical simulations involving various realistic sets of input transport parameters.

Generalization to the case of two- or three-dimensional matrix blocks of arbitrary shape is proposed using a similar Ansatz, solution of a non-linear eigenvalue problem. A fast converging algorithm based on a fixed-point sequence starting from a suitable first guess was developed. Comparisons with full-time simulations for several typical block geometries show an excellent agreement.

These results permit to set-up an analytical formulation generalizing linear single-phase representation of matrix-to-fracture exchange term. It accounts for the non-linearity of the local flow equations using the power-law dependence of the conductivity for low NAPL saturation. The corresponding exponent can be predicted from the input conductivity parameters. Similar findings are also presented and validated numerically for two- or three-dimensional matrix blocks. That original approach paves the way to research leading to a more faithful description of matrix-to-fracture exchanges when considering a realistic fractured medium composed of a population of matrix blocks of various size and shapes.


1. Introduction

10 In many situations, fluid transport processes in complex
11 porous systems may be described by the solution of a non-
12 linear diffusion equation with a diffusion coefficient that
13 depends on the local concentration or saturation of the
14 solute or phase of interest. In the geosciences context, that
15 can be the case when considering NAPL¹ displacement by
16

17 water in aquifers or hydrocarbon recovery in rock matrix
18 [9, 22, 29, 39, 41, 21, 26]. In the context of soil mechanics,
19 one derives semi-analytical or numerical solutions of the
20 Richards equation describing unsaturated flows with such
21 non-linear forms [8, 34, 35, 5]. Other systems may receive
22 similar descriptions ranging from filtration to astrophysics,
23 including compressible gas flows in porous media, granular
24 media flows [18, 17], among others.

25 The non-linearity of the driving diffusion equation im-
26 plies that most of standard methods based on superposition
27 properties such as Green's functions and Fourier decompo-
28 sition fail down. In addition, the popular assumption that
29 the diffusion coefficient vanishes at the limiting saturations,
30 sharing a power-law dependence with the saturation adds
31 a mathematical difficulty [2]. In particular, singular be-
32 haviours are to be expected close to the forcing boundaries
33 where the diffusion coefficient may vanish.


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¹Hydrocarbons in liquid or gaseous state, pollutants, organic compounds, air...

34 In order to be more specific, in the geosciences context
35 such a generic problem arises when considering a counter-
36 current capillary imbibition process on a finite size matrix
37 block approximated by a layer of length L embedded in
38 a fracture network of high conductivity [22, 29, 41, 26].
39 The most standard approach to account for the presence of
40 fractures is to adopt a dual-porosity description that involves
41 two averaged equations for the macro-scale fracture and
42 matrix effective media, in which some exchange term needs
43 to be modeled [3, 16, 37, 25, 14]. In the single-phase case,
44 the simplest model is the pseudo-steady-state dual-porosity
45 model, that can be improved to account for additional inter-
46 nal matrix relaxation times using a time convolution with
47 some kernel that is the solution of a linear diffusion problem
48 on a representative matrix block [25, 30]. Such convolution
49 models are not practical in numerical modeling but retain the
50 multiple time-scales which are naturally observed.

51 Dual-porosity models involving a linear closure (local
52 inter-porosity flux proportional to pressure/saturation/con-
53 centration difference) imply an exponential relaxation of the
54 average quantity of interest, i.e. a single relaxation time. The
55 associated characteristic time corresponds to the diffusion
56 time over the matrix size $\tau_0 = L^2/D_0$, defined using some
57 representative diffusion coefficient D_0 and a representative
58 matrix block size L . At short times, a “boundary layer” be-
59 haviour is observed and can be estimated using the standard
60 Boltzmann transformation, leading to a matrix-to-fracture
61 flux varying as $1/\sqrt{t}$ [17, 26]. This regime may be observed
62 while the diffusion distance remains lower than the typical
63 size of the matrix blocks.

64 Fewer results are reported regarding the long time regime
65 description occurring when water has invaded most of the
66 pore volume of the rock. Using an overall dual-porosity
67 description with a linear closure leads to first-order differ-
68 ential equations for the description of the matrix-fracture
69 exchange, the solution of which implies an exponential
70 relaxation of the NAPL saturation at long times [39]. That
71 is reminiscent of the dual-porosity solutions obtained while
72 interpreting pumping tests in fractured formations. The asso-
73 ciated relaxation time still involves a characteristic diffusion
74 time τ_0 conveniently weighted by a dimensionless shape
75 factor that characterizes the overall matrix block geometrical
76 shape [25].

77 In order to account more accurately for non-linear ef-
78 fects, an alternative approach is to change the analytical form
79 of the exchange term by looking for a form involving a non-
80 linear function of the average matrix block saturation. Semi-
81 analytical developments in that direction were proposed by
82 [41, 26]: solving a weak form of the full problem at hand, the
83 authors introduce an approximation of the late-time solution
84 using a suitably chosen spatial Ansatz. The model predicts a
85 power-law time decay of the NAPL average saturation in the
86 blocks. The corresponding exponent may be related directly
87 to the exponent involved in the local diffusion coefficient
88 dependence with saturation. Both papers show a good agree-
89 ment between experimental data and simulation results. For
90 completeness, [26] proposed an explicit representation of the

exchanges using a non-linear term valid over the entire time
scale.

In the present paper, we develop an alternative approach
for the late-time asymptotics, looking for solutions having a
power-law time decay at every location of the matrix block.
Following this assumption, a complete determination of the
solution can be obtained. This solution, which compares
very well with numerical solutions, can be useful to propose
a non-linear formula relating the matrix to fracture exchange
flux to the remaining average NAPL saturation in the matrix.
Generalizations of our approach to more complex fractured
media are discussed.

The paper is organized as follows: first, the generic
problem to be solved is presented in Section 2.2, and the
notations and driving equations are introduced in Section
2.3. In particular, it is shown that the emergence of a non-
linear diffusion equation driving the time-evolution of wa-
ter saturation is not limited to specific fracture network
geometries. A straightforward derivation is provided using
the concept of *global pressure* that allows to show that the
mean fluid velocity vanishes under various geometrical and
physical conditions.

Restricting ourselves to one-dimensional counter-current
displacements, results of semi-analytical short-time anal-
ysis using Boltzmann transformation are reviewed shortly
in Section 3. Then the long-time asymptotic solution is
presented in Section 4, the spatial dependence of which
can be fully determined analytically, still in the case of a
one-dimensional matrix geometry. In both sections, these
solutions are compared with numerical simulations carried
out at both short and long times.

In Section 5, the time variation of the overall flux at
the matrix boundary is studied. That allows to set-up a
formulation of the exchange term accounting for the non
linearity.

Coming back to 2D or 3D cases, the approach is general-
ized for *arbitrary matrix block shapes* in Section 6. The cor-
responding Ansatz that describes the space dependency of
the asymptotic behaviour is solution of a non-linear steady-
state eigenvalue problem. That solution can be evaluated
numerically using a fast converging original fixed-point al-
gorithm presented in Section 6.3. That solution plays the role
of the analytical solution developed in the one-dimensional
case. Comparisons to full simulations showing an excellent
agreement are presented in Section 6.4 for various matrix
block geometries, including the one-dimensional previous
solution.

Section 7 concludes and an appendix detailing the nu-
merical scheme used to solve the counter-current equation
closes the article.

2. Analytical and numerical models

In this section, we present the mathematical model and
the numerical solution that will be used as a reference.

145
146 In a network of diffuse fractures where flow is taking
147 place, the exchange interaction with the matrix blocks
148 delineated by the fracture network, as sketched in Fig. 1,
149 involves four forces: gravity, viscous forces, capillarity and
150 molecular diffusion. In the following we will assume on the
151 one hand that the molecular diffusion forces, which are by far
152 the weakest, are negligible. On the other hand we consider
153 such a high permeability contrast between the fractures and
154 the matrix that the overall pressure gradient acting on one
155 matrix block through the fracture is negligible. This leaves
156 capillarity and gravity as dominant forces.

157 The capillary length l_{cap} is defined as the length beyond
158 which gravity becomes important [15]. In porous media,
159 it can be estimated by comparing the Laplace pressure
160 $(2\gamma \cos \theta)/R$, where γ is the interfacial tension between water
161 and NAPL, θ is the contact angle and R is the characteristic
162 capillary tube radius of the equivalent porous medium
163 represented as a bundle of capillary tubes, to the gravity-
164 driven hydrostatic pressure $\Delta\rho gh$, where $\Delta\rho = \rho_w - \rho_o$ is the
165 density difference between water and NAPL, h denotes the
166 matrix block height and g the gravity acceleration. Equating
167 these two pressures, setting $R = \sqrt{8k/\phi}$ where k and ϕ
168 denote the matrix block average permeability and porosity,
169 defines the capillary length

$$l_{\text{cap}} = \frac{2\gamma \cos \theta}{\Delta\rho g R} = \frac{\gamma \cos \theta \sqrt{\phi}}{\Delta\rho g \sqrt{2k}}. \quad (1)$$

170 This capillary length, which corresponds to a block height
171 such that gravity and capillary forces are balanced, i.e. a unit
172 Bond number

$$\text{Bo} = \frac{\Delta\rho gh R}{2\gamma \cos \theta} = \frac{\Delta\rho gh \sqrt{2k}}{\gamma \cos \theta \sqrt{\phi}} = \frac{h}{l_{\text{cap}}}, \quad (2)$$

173 defines two regimes depending on whether $l_{\text{cap}} \leq h$ or
174 $\text{Bo} \geq 1$. Gravity is negligible for block height $h \ll l_{\text{cap}}$
175 ($\text{Bo} \ll 1$). When this condition is met, it is as sought the
176 fluid is in a zero-gravity environment and capillary effects
177 dominate. The opposite case, when $h \gg l_{\text{cap}}$ is referred to
178 as the gravity regime ($\text{Bo} \gg 1$).

179 In what follows we consider only the capillary regime,
180 that is rock-fluid configurations such that the block height is
181 much lower than the capillary length, $h \ll l_{\text{cap}}$. ?? gives a
182 few examples. For instance, assuming NAPL is a light oil,
183 $\phi = 0.1$ and $\theta = \pi/3$, one gets a capillary length of about
184 $l_{\text{cap}} \sim 4-9$ m for $k = 10$ mD, depending on temperature and
185 pressure conditions. Basically, we are considering blocks of
186 small height of the order of a few tens of centimeters to one
187 meter.

188 **2.2. Counter-current imbibition in the** 189 **three-dimensional incompressible two-phase** 190 **flow case without gravity**

191 We start with general considerations about three-dimen-
192 sional spontaneous counter-current capillary imbibition,

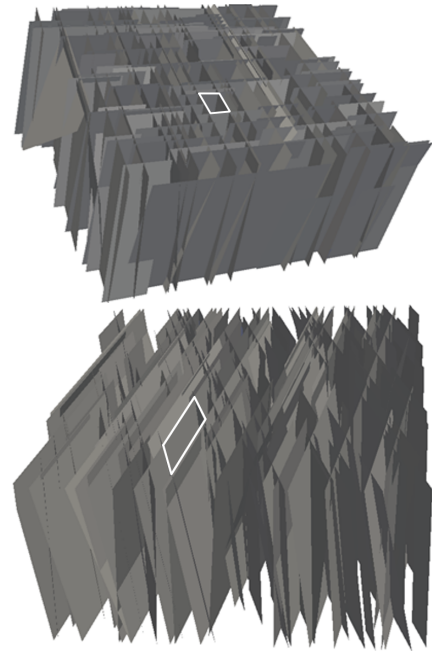


Figure 1: Examples of discrete fracture networks with two near-vertical fracture families (of mean spacing about 3 and 5 m). These fractured geometries delineate matrix blocks (not shown) of various shapes with two typical quadrangular contours shown in white that are the focus of this work (adapted from [21]).

193 then detail in Section 2.3 the corresponding one-dimensional
194 problem under the same assumptions. We consider two-
195 phase flow of incompressible fluids without gravity. Fol-
196 lowing the generalized multiphase Darcy's laws, the driving
197 equations read [4]:

$$\phi \frac{\partial S_w}{\partial t} - \nabla \cdot (k \lambda_w \nabla P_w) = 0, \quad (3)$$

$$\phi \frac{\partial S_o}{\partial t} - \nabla \cdot (k \lambda_o \nabla P_o) = 0, \quad (4)$$

$$S_w + S_o = 1, \quad (5)$$

$$P_o - P_w = P_c(S_w), \quad (6)$$

198 where ϕ and k denote the porosity and the permeabil-
199 ity of the porous medium under consideration (here and
200 in the following we assume that the permeability tensor
201 is isotropic), $S_w(\mathbf{x}, t) \in [S_{wi}, 1 - S_{orw}]$ is the aqueous
202 phase saturation, $S_o(\mathbf{x}, t) \in [S_{orw}, 1 - S_{wi}]$ is the non-
203 aqueous phase saturation, S_{wi} is the connate (immobile)
204 aqueous phase saturation, S_{orw} is the residual (immobile)
205 non-aqueous phase saturation, $\lambda_\varphi(S_w) = k_{r\varphi}(S_w)/\mu_\varphi$ is
206 the phase φ mobility where $\varphi = w, o$ denotes the aqueous
207 and the non-aqueous phase respectively, $k_{r\varphi}$ is the relative
208 permeability to phase φ , μ_φ is phase φ viscosity, P_φ the
209 intrinsic average pressure in phase φ , and $P_c(S) = P_o - P_w$ is
210 the capillary pressure between the non-aqueous and aqueous
211 phases.

212 It is useful to introduce the concept of global pressure P_I
213 that depends implicitly on position \mathbf{x} and time t introduced

214 by [11, 12, 13, 20]:

$$P_t = P_w + \int_{S_i}^S \frac{\lambda_o}{\lambda_t} \frac{dP_c}{dS'} dS', \quad (7)$$

215 where $\lambda_t = \lambda_w + \lambda_o$ is the total mobility and $S \in [0, 1]$ the
216 normalized mobile aqueous phase saturation defined as

$$S(\mathbf{x}, t) = \frac{S_w(\mathbf{x}, t) - S_{wi}}{1 - S_{orw} - S_{wi}} \quad (8)$$

217 where the mobile saturation range $1 - S_{orw} - S_{wi}$ is non-zero.
218 Global pressure is defined up to an arbitrary constant which
219 is accounted for by choosing S_i that is an arbitrary saturation
220 value in $[0, 1]$. With some algebraic manipulations, the
221 original set of equations (3)-(6) can thus be transformed as

$$\nabla \cdot \mathbf{u}_t = 0, \quad (9)$$

$$\mathbf{u}_t = -k \lambda_t \nabla P_t, \quad (10)$$

$$\phi \frac{\partial S_w}{\partial t} + \nabla \cdot \left(\frac{\lambda_w}{\lambda_t} \mathbf{u}_t + k \frac{\lambda_w \lambda_o}{\lambda_t} \nabla P_c \right) = 0, \quad (11)$$

222 where $\mathbf{u}_t = \mathbf{u}_w + \mathbf{u}_o$ denotes the total Darcy velocity, with
223 $\mathbf{u}_\phi = -k \lambda_\phi \nabla P_\phi$ and $\phi = w, o$. Equation (10) can be derived
224 from equations (3)-(6) and using equation (7) defining global
225 pressure. It can be noticed that $\lambda_t \nabla P_t = \lambda_w \nabla P_w + \lambda_o \nabla P_o$.
226 Global pressure may be interpreted as a pressure which
227 would give, for a fictitious fluid with mobility λ_t equal to the
228 sum of the mobilities of aqueous and non-aqueous phases,
229 a flux equal to the sum of the flux of the aqueous and non-
230 aqueous phases.

231 Combining Eqs (9) and (10), one is led to solve a quasi-
232 Laplace equation that reads $\nabla \cdot (k \lambda_t \nabla P_t) = 0$ to be solved on
233 the matrix domain with given boundary conditions. Imposing
234 the rather general boundary conditions at the boundaries
235 of the block, i.e. uniform pressure and saturation, we obtain
236 that the global pressure (7) P_t is uniform at the block bound-
237 aries. Thus it can be shown that the unique solution of the
238 quasi-Laplace equation fulfilling these boundary conditions
239 is a constant: P_t does not depend on position. Therefore,²
240 the total velocity vanishes: $\mathbf{u}_t = \mathbf{0}$. This result requires
241 the absence of gravity effects, incompressible fluids, and
242 the matrix permeability k may be heterogeneous. Setting
243 $\mathbf{u}_t = \mathbf{0}$ in Eq. (11) shows that the saturation is driven by
244 the following non-linear diffusion equation:

$$\frac{\partial S}{\partial t} = \nabla \cdot [D(S) \nabla S], \quad (12)$$

$$D(S) = -\frac{k}{\phi(1 - S_{orw} - S_{wi})} \frac{\lambda_w(S) \lambda_o(S)}{\lambda_t(S)} \frac{dP_c(S)}{dS}. \quad (13)$$

245 That result shows that counter-current imbibition is not
246 restricted to one-dimensional cases. Up to our knowledge,
247 that result as well as its direct derivation introducing the
248 global pressure for 2D or 3D situations under quite general
249 conditions appear to be original and shows this non-linear
250 diffusion equation is likely to describe the water saturation
251 evolution in many situations of interest.

²This result can be derived using a variational formulation of Laplace equation.

2.3. Driving equations in the one-dimensional case

252 Spontaneous counter-current capillary imbibition of a
253 one-dimensional matricial porous medium block of size L
254 initially saturated with a non-aqueous phase that is drowned
255 in water can be described, neglecting gravity and assuming
256 the two phases are incompressible, as the non-linear diffu-
257 sion equation (12) for the normalized mobile aqueous phase
258 saturation (8) with initial condition $S = 0$ for all $x \in [0, L]$
259 and $t = 0$ and boundary conditions $S = 1$ for $x = 0$ and
260 $x = L$ for all $t > 0$.

261 In order to analyze the early- and late-time behaviours,
262 we express $D(S)$ as a function of the mobile aqueous phase
263 saturation S (mobile non-aqueous phase saturation being
264 $1 - S$) setting the relative permeabilities to the aqueous and
265 non-aqueous phases and the capillary pressure as Brooks-
266 Corey saturation power laws [7]:

$$k_{rw}(S) = \kappa_w S^p, \quad (14)$$

$$k_{ro}(S) = \kappa_o (1 - S)^q, \quad (15)$$

$$P_c(S) = P_e / S^{1/m}, \quad (16)$$

268 where p, q and m are positive exponents, κ_ϕ is the maximum
269 relative permeability to phase ϕ , and P_e is the entry pressure
270 of the porous medium under consideration. While specifying
271 relative permeabilities and capillary pressure saturation
272 dependence is certainly a restriction, it should be noted that
273 Brooks and Corey relationships are widely used for many
274 rock types. In addition, it does not matter what the choice is
275 as long as a power-law behavior is obtained at long times
276 for $S \rightarrow 1$ (this last statement is less true regarding the
277 early-time regime, which is not the major contribution of this
278 paper, as it is discussed later). We highlight this by making
279 a more versatile choice, later at the end of this section. The
280 diffusion coefficient $D(S)$ given in Eq. (12) then writes

$$D(S) = D_0 \frac{S^r (1-S)^q}{M S^{p+1} (1-S)^q} = D_1 \frac{S^r (1-S)^q}{S^{p+1} (1-S)^q}, \quad (17)$$

281 with

$$D_0 = \frac{k \kappa_w P_e}{m \phi \mu_w (1 - S_{orw} - S_{wi})} = M D_1, \quad (18)$$

$$M = \frac{\mu_o \kappa_w}{\mu_w \kappa_o} = \frac{\lambda_w(S=1)}{\lambda_o(S=0)}, \quad (19)$$

$$r = p - \frac{m+1}{m}, \quad (20)$$

282 where M denotes the end-point mobility ratio between the
283 aqueous and the non-aqueous phases, defined as the maxi-
284 mum aqueous phase mobility k_{rw}/μ_w , taken at $S = 1$, over
285 the maximum non-aqueous phase mobility k_{ro}/μ_o , taken at
286 $S = 0$ [6].

287 It is worth noting that Eq. (12) is of singular type for
288 $r > 0$ or $q > 0$ because $D(S)$ cancels with either the aqueous
289 phase mobility for $S = 0$ or the non-aqueous phase mobility
290 for $S = 1$. It means that mathematical singularities can be
291 expected near the boundary where $D \rightarrow 0$ for $S \rightarrow 0$, as well
292 as in the long-time limit inside the rock for which $D \rightarrow 0$ for
293 $S \rightarrow 1$. A singular regime can be expected at very short
294 times too, due to the sharp jump of the water saturation from
295 1 to 0 near the boundary.

Clearly, the balance of forces at play that lies in a capillary pressure difference between the fracture (where $P_c = 0$) and the matrix (where $P_c \neq 0$) media commands that a flow is established and terminates when the equilibrium condition $P_c(S) = 0$ is reached, i.e. $S(x, t) = 1$ or $S_w(x, t) = 1 - S_{orw}$. Noteworthy, a non-zero solution of Eq. (12) should satisfy the boundary condition $(\partial S / \partial x)_{x=0} = -\infty$ and therefore present a vertical asymptote at the front face of the porous medium. Indeed, this is a necessary condition to get a non-zero fracture-to-matrix flux $(D(S) \partial S / \partial x)_{x=0}$ while D cancels at the boundary.

Thereafter, two limiting cases are particularly useful to consider: on the one hand, the case where $S \rightarrow 0$, which corresponds to the early-time counter-current spontaneous imbibition of the non-aqueous-phase-saturated block by an aqueous phase, and, on the other hand, the case where $S \rightarrow 1$, which corresponds to the late-time imbibition of the same type. For early- and late-time regimes, $D(S)$ behaves asymptotically as

$$D(S) \sim \begin{cases} D_0 S^{\alpha_0} & \text{with } \alpha_0 = r \text{ for } S \rightarrow 0, \\ D_1 (1 - S)^{\alpha_1} & \text{with } \alpha_1 = q \text{ for } S \rightarrow 1. \end{cases} \quad (21)$$

Thus each of these two limiting cases reduces to a non-linear diffusion equation of the singular type with a power-law $D(S)$. Specifically, one gets for $S \rightarrow 0$

$$\frac{\partial S}{\partial t} = \frac{\partial}{\partial x} \left(D_0 S^{\alpha_0} \frac{\partial S}{\partial x} \right), \quad (22)$$

and for $S \rightarrow 1$

$$\frac{\partial(1 - S)}{\partial t} = \frac{\partial}{\partial x} \left[D_1 (1 - S)^{\alpha_1} \frac{\partial(1 - S)}{\partial x} \right], \quad (23)$$

where D_0 , D_1 , α_0 and α_1 are given in Eqs (18), (19), (20) and (21). Typical $D(S)$ curves are reported in Fig. 2.

Coming back to the general case, considering a matrix block Ω of boundary $\partial\Omega$ with initial boundary value problem $S(\mathbf{x}, t) = 0$ in Ω for $t = 0$ and $S(\mathbf{x}, t) = 1$ on $\partial\Omega$ for all $t \geq 0$, Eqs (22) and (23) read respectively

$$\frac{\partial S}{\partial t} = \nabla \cdot (D_0 S^{\alpha_0} \nabla S), \quad (24)$$

$$\frac{\partial(1 - S)}{\partial t} = \nabla \cdot [D_1 (1 - S)^{\alpha_1} \nabla(1 - S)]. \quad (25)$$

Given the initial boundary value problem under investigation, the late-time regime has an expected limit $S \rightarrow 1$ when $t \rightarrow \infty$, the early-time regime has a more subtle behavior since the initial condition corresponds to a jump at $x = 0$ because of the boundary condition $S = 1$ and the initial condition $S = 0$. As a consequence, any series solution will feature a full spectrum of saturation functions. We come back to this point in Section 3.

To finish with, it is worth noting that relationships (21)-(23) still hold when considering more versatile relative permeabilities such as [28, 27]

$$k_{rw}(S) = \kappa_w \frac{S^p}{S^p + \beta(1 - S)^u}, \quad (26)$$

$$k_{ro}(S) = \kappa_o \frac{(1 - S)^q}{(1 - S)^q + \gamma S^v}. \quad (27)$$

In this case one gets

$$D(S) = D_0 \frac{S^{p - \frac{m+1}{m}} (1 - S)^q}{M S^p [(1 - S)^q + \gamma S^v] + (1 - S)^q [S^p + \beta(1 - S)^u]}, \quad (28)$$

hence $D(S) \sim D_0 S^{p - \frac{m+1}{m}} / \beta$ for $S \rightarrow 0$ and $D(S) \sim D_0 (1 - S)^q / (M \gamma)$ for $S \rightarrow 1$, which is precisely Eq. (21) up to the dimensionless constants β and γ . Although this type of correlation has no physical basis, unlike power-law relative permeabilities (14)-(15) which can be derived by analogy with a bundle of capillary tubes [36, 10, 45, 46, 43, 44, 40, 38], it can nevertheless be useful when considering natural porous media for which power laws are not good enough to match measurements.

Our starting point is thus relatively general and is valid as long as an asymptotic power-law behaviour is obtained at the extreme saturations, independently of any detail at intermediate saturation values and regardless of any empirical correlation used.

3. Early-time solution

The goal of present work is to set-up a description of the fluid transfer valid for the whole time range. The literature is very abundant on the short time scale regime that can be assimilated to flow in a semi-infinite medium. We recall some outstanding works of [18, 34] that are relatively little cited nowadays, whereas they give very accurate fudge-factor-free solutions.

In the early-time regime, following [8, 18, 34] and references therein, the idea is to consider that the overall size of the block may be ignored, leading to considering a semi-infinite medium. That suggests to seek a solution using the Boltzmann variable $\xi = x / \sqrt{4D_0 t}$, with boundary conditions $S(0) = 1$, $S(\infty) = 0$ and $D(S(\infty)) (dS/d\xi)(\infty) = 0$. That yields thus the following equation

$$\frac{d}{d\xi} \left(\frac{D(S)}{D_0} \frac{dS}{d\xi} \right) + 2\xi \frac{dS}{d\xi} = 0. \quad (29)$$

A remarkable fact observed by many authors [18, 9, 34, 22, 41, 17, 26] is that the solution of this equation has a finite toe located at a distance ξ_0 from the boundary that depends on the input parameters of the equation.

In order to go farther, the saturation dependence of $D(S)$ suggests to express ξ as a function of S . A further transformation allows to treat the case of a $D(S)$ function given by Eq. (17). Following [8, 34], Eq. (29) can be recast under the equivalent form

$$D(S) = -2D_0 \frac{d\xi}{dS} \int_0^S \xi dS. \quad (30)$$

This form assumes the existence of a finite front ξ_0 for which the saturation and the flux, proportional to $D(S) dS/d\xi$, cancel out.

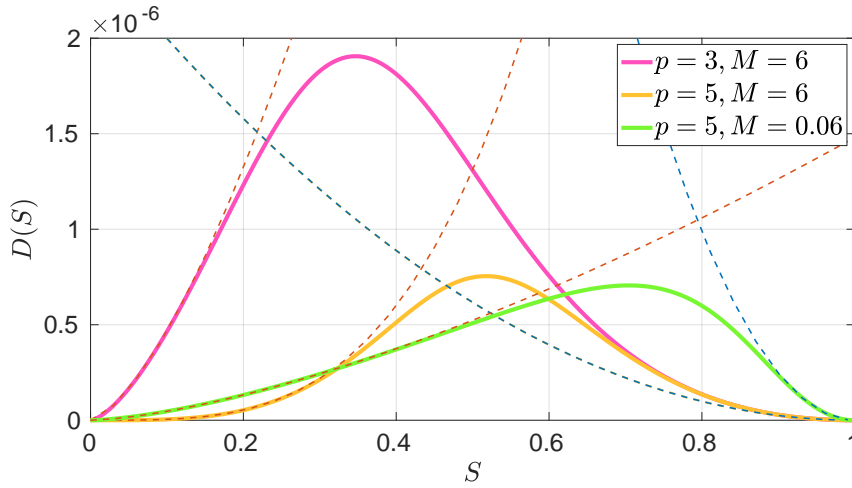


Figure 2: Several $D(S)$ given by Eq. (17) for $p = 3, 5$ and $M = 6, 0.06$ with $q = 2, m = 2, k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25$, $S_{wi} = S_{orw} = 0$. Asymptotic power laws for the extreme saturations given by Eq. (21) are reported as dashed lines.

378 An iterative sequence of approximations [34] can be built
 379 by approximating the right-hand-side integral of Eq. (30)
 380 which yields, at the lowest order approximation

$$\frac{1}{2D_0} \int_0^S \frac{D(S')}{S'} dS' \approx \xi_0(\xi_0 - \xi), \quad (31)$$

381 and, at the higher order approximation

$$\frac{1}{2D_0} \int_0^S \frac{D(S')}{S'} dS' \approx \xi_0(\xi_0 - \xi) - \frac{A}{2}(\xi_0 - \xi)^2, \quad (32)$$

382 where A is a constant equal to $1/(\alpha_0 + 1)$ for a power-law
 383 $D(S)$, which is approximately the case for a $D(S)$ function
 384 given by Eq. (17) if α_0 is large enough. The determination of
 385 ξ_0 follows directly from the boundary condition $S(\xi = 0) =$
 386 1 . These approximations provide an excellent accuracy not
 387 only in the neighborhood of ξ_0 for low saturation values.

388 In the power-law case, the solution proposed by [34] is
 389 consistent with the findings of [18] whose solution, later
 390 extended by [35], reads

$$S(\xi) = \begin{cases} [2\alpha_0\xi_0(\xi_0 - \xi)]^{1/\alpha_0} f\left(\frac{\xi}{\xi_0}\right) & \text{if } \xi \in [0, \xi_0], \\ 0 & \text{if } \xi > \xi_0. \end{cases} \quad (33)$$

391 Here, the function $f(\xi/\xi_0)$ is a regular function that may be
 392 expressed as a power series of ξ/ξ_0 involving the exponent
 393 α_0 as a parameter [18].

394 The complete early-time saturation profile thus may be
 395 estimated analytically with an excellent accuracy, as re-
 396 ported in Fig. 3 which compares, for a few configurations,
 397 [34] solution (32) obtained by setting $A = 1/(\alpha_0 + 1)$
 398 with the numerical solution obtained with the numerical
 399 scheme described in Appendix A. Saturation presents a
 400 vertical asymptote at the front face of the porous medium
 401 for $x = \xi = 0$ because a non-zero fracture-to-matrix flux
 402 $D(S) \partial S / \partial x = (D(S) / \sqrt{4D_0 t}) dS / d\xi$ while $D(S) = 0$
 403 for $S = 1$ at the boundary is only possible if $\partial S / \partial x =$
 404 $dS / d\xi = -\infty$. Flux vanishes as well at the toe position

$\xi = \xi_0$ where $dS / d\xi = -2\xi_0 [2\alpha_0\xi_0(\xi_0 - \xi)]^{(1-\alpha_0)/\alpha_0} \sim$ 405
 $1 / (\xi_0 - \xi)^{(\alpha_0 - 1)/\alpha_0}$ [18], therefore the larger α_0 the sharper the 406
 front as shown in Fig. 3. To finish with, dimensional analysis 407
 suggests to normalize the timescale by the diffusion time 408
 $\tau_0 = L^2 / (4D_0)$. Following our work objective, expressions 409
 for the matrix-to-fracture exchange term are investigated in 410
 Section 5. 411

4. Late-time asymptotic solution 412

Now, we focus on the late-time regime solution, for 413
 which the medium can no longer be assumed to be semi- 414
 infinite, and for which the authors have not found, to their 415
 best knowledge, any published exact solution. 416

An asymptotic solution for large t is sought using the 417
 Ansatz $1 - S(x, t) = y(x) / t^\beta$. Inserting this solution into 418
 Eq. (23) shows that this may be possible if the following 419
 condition is satisfied: 420

$$\beta = \frac{1}{\alpha_1}. \quad (34)$$

The asymptotic solution of Eq. (23) thus writes 421

$$1 - S(x, t) = \frac{y(x)}{t^{1/\alpha_1}}, \quad (35)$$

where $y(x)$ is a solution of the ordinary differential equation 422
 $\frac{d}{dx} (y^{\alpha_1} \frac{dy}{dx}) = -\frac{1}{\alpha_1 D_1} y$, which we shall rewrite 423

$$\frac{d^2}{dx^2} y^{\alpha_1+1} = -\frac{\alpha_1+1}{\alpha_1 D_1} y. \quad (36)$$

Further transformations $g = y^{\alpha_1+1}$ and $h = \lambda g$ with λ a 424
 constant yield 425

$$\frac{d^2 h}{dx^2} = -\lambda \frac{\alpha_1}{\alpha_1+1} \frac{\alpha_1+1}{\alpha_1 D_1} h^{\frac{1}{\alpha_1+1}}. \quad (37)$$

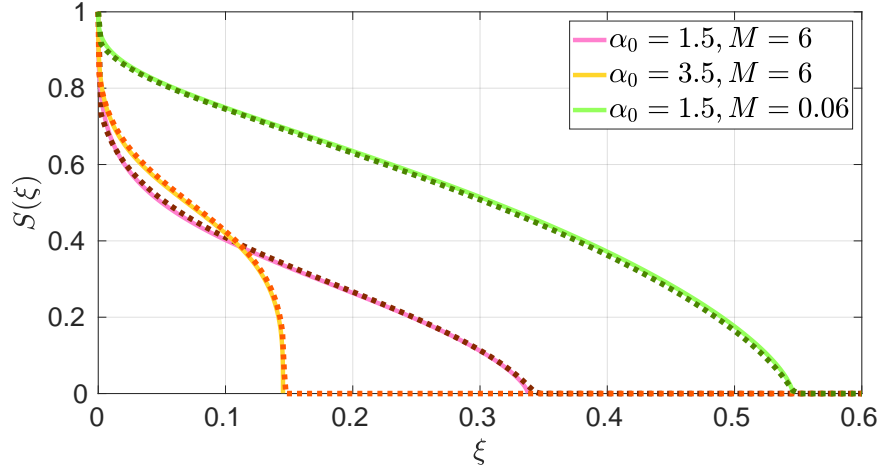


Figure 3: Comparison of early-time analytical solution given in Eq. (32) (solid lines) with the simulated one (dotted lines) for $\alpha_0 = 1.5, 3.5$ ($p = 3, 5$) and $M = 6, 0.06$ with $q = 2, m = 2, k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25, S_{wi} = S_{orw} = 0$.

426 Choosing $\lambda = \left(\frac{\alpha_1 D_1}{\alpha_1 + 1}\right)^{\frac{\alpha_1 + 1}{\alpha_1}}$, Eq. (37) may be written under
 427 the following form:

$$\frac{d^2 h}{dx^2} = -h^{\frac{1}{\alpha_1 + 1}}, \quad (38)$$

$$h = \lambda g = \lambda y^{\alpha_1 + 1} = \left(\frac{\alpha_1 D_1}{\alpha_1 + 1}\right)^{\frac{\alpha_1 + 1}{\alpha_1}} y^{\alpha_1 + 1}. \quad (39)$$

428 For $x \in [0, L]$, boundary conditions $1 - S = 0$ for all
 429 $t > 0$ at $x = 0$ and $x = L$ yield $y(0) = y(L) = 0$, that is
 430 $h(0) = h(L) = 0$. The solution being symmetric around
 431 $x = L/2$, one must have $dy/dx > 0$ for $x \in [0, L/2]$
 432 and $dy/dx < 0$ for $x \in [L/2, L]$. Therefore the condition
 433 $(dy/dx)(L/2) = 0$ must hold.

434 Before going further it is worth noting that Eq. (38)
 435 simplifies into $d^2 h/dx^2 = -1$ when exponent α_1 is very
 436 large, whose solution is the parabola $h(x) = \frac{1}{2}x(L - x)$ such
 437 that $h(L/2) = L^2/8$.

438 Coming back to the general case, it can be remarked that
 439 multiplying each member of Eq. (38) by $\frac{dh}{dx}$, the equation can
 440 be integrated once to yield

$$\frac{1}{2} \left(\frac{dh}{dx}\right)^2 = \frac{\alpha_1 + 1}{\alpha_1 + 2} \left(h^{\frac{\alpha_1 + 2}{\alpha_1 + 1}} \left(\frac{L}{2}\right) - h^{\frac{\alpha_1 + 2}{\alpha_1 + 1}} \right), \quad (40)$$

441 where the right hand side $\frac{\alpha_1 + 1}{\alpha_1 + 2} h^{\frac{\alpha_1 + 2}{\alpha_1 + 1}} \left(\frac{L}{2}\right)$ is an integration
 442 constant that is determined by the condition $\frac{dh}{dx} \left(\frac{L}{2}\right) = 0$. For
 443 $x \in [0, L/2]$, this last form integrates into

$$\begin{aligned} x &= \sqrt{\frac{\alpha_1 + 2}{2(\alpha_1 + 1)}} \int_0^h \frac{dh}{\sqrt{h^{\frac{\alpha_1 + 2}{\alpha_1 + 1}} \left(\frac{L}{2}\right) - h^{\frac{\alpha_1 + 2}{\alpha_1 + 1}}} } \\ &= \sqrt{\frac{\alpha_1}{2(\alpha_1 + 2)}} \int_0^{[h/h(L/2)]^{\frac{\alpha_1 + 2}{\alpha_1 + 1}}} \frac{t^{\frac{\alpha_1 + 1}{\alpha_1 + 2} - 1}}{\sqrt{1 - t}} dt, \end{aligned} \quad (41)$$

setting $t = [h/h(L/2)]^{\frac{\alpha_1 + 2}{\alpha_1 + 1}}$. This relationship set with $x = \frac{L}{2}$
 yields $h(L/2)$ in implicit form:

$$\frac{L}{2} = \sqrt{\frac{(\alpha_1 + 1)h^{\frac{\alpha_1}{\alpha_1 + 1}} \left(\frac{L}{2}\right)}{2(\alpha_1 + 2)}} \int_0^1 \frac{t^{\frac{\alpha_1 + 1}{\alpha_1 + 2} - 1}}{\sqrt{1 - t}} dt. \quad (42)$$

Using Euler incomplete beta and gamma³ functions, the
 solution rewrites as

$$x = \sqrt{\frac{(\alpha_1 + 1)h^{\frac{\alpha_1}{\alpha_1 + 1}} \left(\frac{L}{2}\right)}{2(\alpha_1 + 2)}} B\left(\left[\frac{h}{h(L/2)}\right]^{\frac{\alpha_1 + 2}{\alpha_1 + 1}}; \frac{\alpha_1 + 1}{\alpha_1 + 2}, \frac{1}{2}\right), \quad (43)$$

with $h(L/2)$ given by

$$h\left(\frac{L}{2}\right) = \left(\frac{L}{2\sqrt{2\pi}} \frac{\alpha_1}{\sqrt{(\alpha_1 + 1)(\alpha_1 + 2)}} \frac{\Gamma\left(\frac{\alpha_1}{2(\alpha_1 + 2)}\right)}{\Gamma\left(\frac{\alpha_1 + 1}{\alpha_1 + 2}\right)}\right)^{\frac{2(\alpha_1 + 1)}{\alpha_1}}. \quad (44)$$

Noteworthy, this solution converges for $\alpha_1 \rightarrow \infty$ to the
 parabola that was previously obtained by direct calculation
 in the specific case of a very large α_1 exponent, which writes
 implicitly $x = L/2 - \sqrt{(L/2)^2 - 2h}$ with $h(L/2) = L^2/8$.

Going back to the main unknown y , related to h by Eq.
 (38), the following implicit expression that fulfills all the
 boundary conditions may be obtained:

$$x = \sqrt{\frac{\alpha_1 D_1 y^{\alpha_1} \left(\frac{L}{2}\right)}{2(\alpha_1 + 2)}} B\left(\left[\frac{y}{y(L/2)}\right]^{\alpha_1 + 2}; \frac{\alpha_1 + 1}{\alpha_1 + 2}, \frac{1}{2}\right), \quad (45)$$

with $y(L/2)$ given by

$$y\left(\frac{L}{2}\right) = \left(\frac{L}{2} \sqrt{\frac{\alpha_1}{2\pi(\alpha_1 + 2)D_1}} \frac{\Gamma\left(\frac{\alpha_1}{2(\alpha_1 + 2)}\right)}{\Gamma\left(\frac{\alpha_1 + 1}{\alpha_1 + 2}\right)}\right)^{\frac{2}{\alpha_1}}. \quad (46)$$

³Incomplete beta, beta and gamma function are respectively defined
 as [1] $B(x; a, b) = \int_0^x t^{a-1}(1-t)^{b-1} dt$, $B(x, y) = \int_0^1 t^{x-1}(1-t)^{y-1} dt =$
 $\frac{\Gamma(x)\Gamma(y)}{\Gamma(x+y)}$ and $\Gamma(z) = \int_0^\infty t^{z-1} e^{-t} dt$.

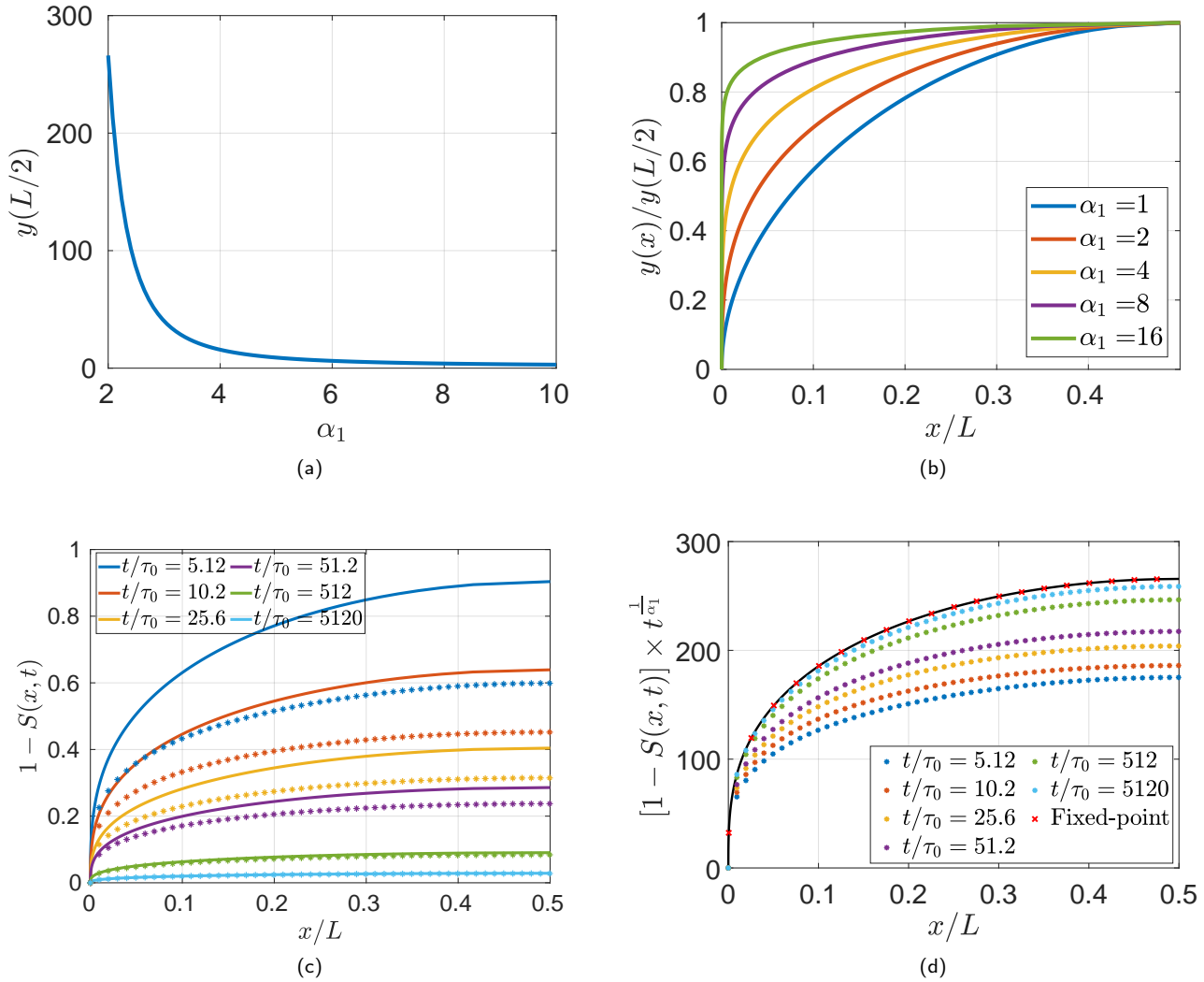


Figure 4: (a)-(b) Late-time analytical solution $y(x)$ given in Eqs (45)-(46) for several α_1 values. (c)-(d) Comparison of the late-time analytical solution $1 - S(x, t) = y(x)/t^{1/\alpha_1}$ and rescaled $y(x) = [1 - S(x, t)]t^{1/\alpha_1}$ (solid lines) given in Eqs (35), (45) and (46) with the simulated one (symbols) for $\alpha_1 = 2$, $p = 3$, $M = 6$, $m = 2$, $k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25$, $S_{wi} = S_{orw} = 0$. Timescale is normalized by the diffusion time $\tau_0 = L^2/(4D_0)$. Red crosses for the fixed-point solution indicated in (d) correspond to a direct numerical solution of the asymptotic Ansatz detailed in Section 6.

458 The solution for $y(\frac{L}{2})$ and $y(x)/y(\frac{L}{2})$ are reported in Figs
 459 4(a) and (b). Unlike solutions (32) and (33) obtained for the
 460 early-time regime by assuming the porous block to be semi-
 461 infinite, the late-time asymptotic solution (45) derived on a
 462 finite-size domain involves a characteristic length which is
 463 the block length L . Contrary to the early-time solution, in
 464 the late-time regime the water saturation is close to unity
 465 at every location in the matrix, therefore the power-law
 466 assumption given in Eq. (23) is automatically satisfied. A
 467 good agreement can be expected for the long time asymptotic
 468 behaviour independently of the details of $D(S)$ over the
 469 whole range of saturations, as reported in Figs 4(c)-
 470 (d) where the timescale is normalized by the diffusion time
 471 $\tau_0 = L^2/(4D_0)$.

472 In the displayed example, the time convergence of the
 473 numerical solution to the asymptotic one given in Eqs (35),

(45) and (46) is pretty slow. That can be accurately quantified
 by comparing the time evolution of the numerical solution
 $1 - S$ or $(1 - S)t^{1/\alpha_1}$ with the asymptotic one for fixed
 x/L values. Specifically, the numerical solution time conver-
 gence to the asymptotic one is slower than the asymptotic
 solution $1/\sqrt{t}$ trend for $\alpha_1 = 2$ and intermediate t/τ_0
 values. The reader may wish to consult Fig. 5 given later in
 Section 5, which reports the time evolution of the matrix-
 to-fracture flux, to spot the transition between the early-
 and the late-time regimes that occurs around $t/\tau_0 = 40$ -
 50 in the example displayed in Figs 4(c)-(d). Ultimately,
 the numerical solution time evolution matches the asymptotic
 solution one as Figs 4(c)-(d) indicate about 7 and 2%
 relative difference between numerical and asymptotic
 solutions, $|S_\infty - S_{num}|/(1 - S_\infty)$, for $t/\tau_0 = 512$ and 5120,
 respectively. The larger the exponent α_1 , the slower this

490 convergence. This is consistent with the typical behaviour of
 491 a porous medium that is preferentially wettable to the non-
 492 aqueous phase, for which the exponent $q = \alpha_1$ of the relative
 493 permeability to the non-aqueous phase, assuming a power-
 494 law saturation dependency (14)-(16), is generally large, for
 495 it imbibes more slowly than a water-wet porous medium.

496 To conclude this section, unlike the early-time regime,
 497 in the late-time regime, the low NAPL assumption $S(t) \approx$
 498 1 becomes more and more valid as time increases. The
 499 asymptotic solution is likely to be independent on the details
 500 of $D(S)$ once $D(S) \approx D_1 (1 - S)^{\alpha_1}$ as $S \rightarrow 1$.

501 5. Towards a non-linear closure for 502 matrix-to-fracture flux

503 When large discrete fracture networks embedded in a
 504 permeable porous matrix are considered, it is customary to
 505 adopt a dual-porosity framework in which the coupling with
 506 the matrix may be accounted for via a source term $\Phi_{mf}(t)$
 507 [25, 14, 32, 30, 33]. This source term $\Phi_{mf}(t)$ corresponds
 508 to a volume average of the normal flux between matrix and
 509 fractures. In the case of linear local flow equations, the
 510 generic closure of $\Phi_{mf}(t)$ appears as being a time convo-
 511 lution of the local variable $S(x, t)$ with a time-dependent
 512 kernel that may be evaluated solving a relaxation problem
 513 on a representative matrix block [25, 30, 33]. In particular,
 514 focusing on the long-time limit, when the relaxation of
 515 the block reaches its exponential decay, it can be shown
 516 that $\Phi_{mf}(t) \approx -\frac{S(x,t) - S_f(x,t)}{\tau_m}$ with x the block centroid and
 517 S_f the surrounding fracture saturation. The relaxation time
 518 τ_m may be related to the block typical dimensions via the
 519 relation $\tau_m \approx \sigma L^2 / D_0$ where σ is the so-called shape-
 520 factor of the matrix block. In the stratified case, $\sigma = 12$.
 521 In the more general case, it is a geometric quantity that is
 522 known for several block shapes, and that is related to the
 523 smallest eigenvalue of the Laplacian operator with Dirichlet
 524 boundary conditions on the block [25].

525 Coming back to the non-linear case, at first sight, the
 526 convolution approach is useless, so other closure must be
 527 proposed, if any. In both short- and long-time regimes,
 528 the time variation of the matrix to fracture flux $\Phi_{mf}(t) =$
 529 $(D(S) \partial S / \partial x)_{x=0}$ can be estimated using the previously
 530 presented solutions. In the short-time case, using the Boltz-
 531 mann variable, $\Phi_{mf}(t)$ may be estimated directly as $\Phi_{mf}(t) =$
 532 $(D(S) / \sqrt{4D_0 t}) (dS/d\xi)_{\xi=0} \sim 1/\sqrt{t}$ up to a prefactor
 533 A_0 . In the long-time case, it is useful to introduce the
 534 spatial average of the matrix saturation defined by $\langle S \rangle(t) =$
 535 $\frac{1}{L/2} \int_0^{L/2} S(x, t) dx$. Averaging the diffusion equation (12)
 536 and swapping the time partial derivative and the spatial aver-
 537 age, one gets $\frac{L}{2} \frac{d\langle S \rangle}{dt} = \int_0^{L/2} \frac{\partial}{\partial x} (D(S) \frac{\partial S}{\partial x}) dx = - (D(S) \frac{\partial S}{\partial x})_{x=0}$
 538 hence $\Phi_{mf}(t) = \frac{L}{2} \frac{d\langle S \rangle}{dt}$. In all the considered cases, sim-
 539 ilar results were obtained using the time derivative of the
 540 average saturation or the global flux at the boundary, so
 541 the former definition was retained. Inserting the late-time
 542 Ansatz in this expression, it may be shown that the matrix to
 543 fracture flux varies asymptotically as $\Phi_{mf}(t) \sim 1/t^{(\alpha_1+1)/\alpha_1}$.

The proportionality factor A_∞ can be evaluated using the
 analytical solution $y(x)$ starting from equations (40) and
 (44). The same algebraic decay was already discovered by
 [41, 26]. In both papers, the authors selected Ansatz dealing
 with the spatial variable valid close to the matrix/fracture
 boundary. Although that approach leads to analogous long
 time variations of the NAPL saturation, it does not provide
 a complete solution in the space domain, and it appears less
 general, especially in the multi-dimensional case.

Summarizing the results, we get:

$$\Phi_{mf}(t) \approx \begin{cases} A_0/\sqrt{t} & \text{for } t \rightarrow 0, \\ A_\infty/t^{\frac{\alpha_1+1}{\alpha_1}} & \text{for } t \rightarrow \infty. \end{cases} \quad (47)$$

Both constants A_0 and A_∞ encapsulate the spatial details of
 the corresponding asymptotic solutions and may be obtained
 manipulating the expressions (29) for A_0 and (39), (40), (43)
 and (44) to obtain A_∞ , that gives the following expressions:

$$A_0 = \sqrt{D_0} \int_0^1 \xi(S) dS, \quad (48)$$

$$A_\infty = \frac{1}{\alpha_1} \left(\frac{\alpha_1+1}{\alpha_1 D_1} \right)^{\frac{1}{\alpha_1}} \sqrt{\frac{2(\alpha_1+1)}{\alpha_1+2}} \times$$

$$\times \left(\frac{L}{2\sqrt{2\pi}} \frac{\alpha_1}{\sqrt{(\alpha_1+1)(\alpha_1+2)}} \frac{\Gamma(\frac{\alpha_1}{2(\alpha_1+2)})}{\Gamma(\frac{\alpha_1+1}{\alpha_1+2})} \right)^{\frac{\alpha_1+2}{\alpha_1}}. \quad (49)$$

If one plots $\Phi_{mf}(t)$ using a log-log scale, two straight lines
 can be observed. The transition time from one regime to the
 other corresponds to a time t such that $\xi_0 = L/2$.

This is illustrated in Fig. 5(a) which reports the time
 evolution of the matrix-to-fracture flux for a few $\alpha_{0,1}$ values.
 Simulated early- and late-time slopes are in excellent agree-
 ment with the early- and late-time predictions $\frac{1}{2}$ and $\frac{\alpha_1+1}{\alpha_1}$,
 respectively. The transition between the early- and late-time
 regimes, which is observed for $t/\tau_0 = 5-10$ in Fig. 5(a) and
 is very clear with a very tight and sharp cross-over, could
 be rescaled to $t/\tau_0 = 1$ by considering not the characteristic
 time $\tau_0 = L^2/(4D_0)$ suggested by dimensional analysis but
 $\tau_0 = L^2/(16\xi_0^2 D_0)$ derived from $\xi = x/\sqrt{4D_0 t}$ setting
 $\xi = \xi_0$ and $x = L/2$. It is worth noting the low influence
 of the early-time exponent α_0 .

Fig. 5(b) reports the time evolution of the simulated
 fracture-to-matrix flux Φ_{mf} multiplied by its asymptotic time
 dependencies (47) and divided by its asymptotic prefactors
 A_0 and A_∞ (48) and (49) for the same α_0 and α_1 exponents as
 in Fig. 5(a). Except for the cross-over between the short and
 long time regimes, the asymptotic solutions (47) are found
 with excellent accuracy and dominate most of the exchange
 dynamics over several orders of magnitude.

It can be observed that using the long-time asymptotics
 $\langle S \rangle \sim t^{1/\alpha_1}$, one can write the matrix to fracture flux under
 the following form: $\Phi_{mf}(t) \sim -\langle S \rangle^{\alpha_1+1}$. That non-linear
 closure between the local matrix to fracture flux and the
 average matrix saturation generalizes the usual linear closure

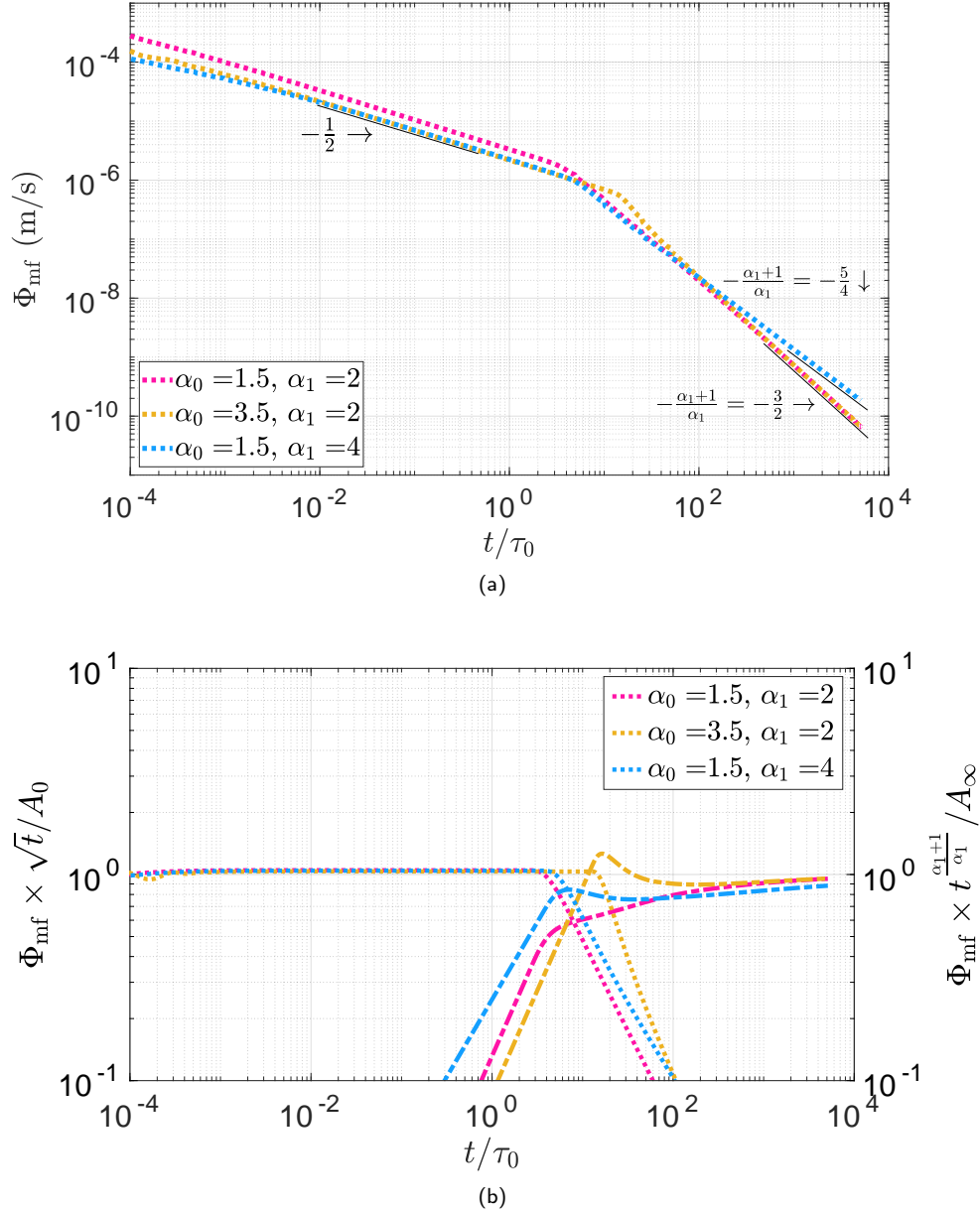


Figure 5: (a) Time evolution of the matrix-to-fracture flux for a few α_0 and α_1 exponents $\alpha_0 = 1.5, 3.5$ ($p = 3, 5, m = 2$) and $\alpha_1 = 2, 4$ with $M = 6, k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25$ and $S_{wi} = S_{orw} = 0$. Simulated early- and late-time slopes $-\{0.48, 0.46, 0.44\}$ and $-\{1.45, 1.48, 1.21\}$, respectively, are in excellent agreement with the early- and late-time predictions (thin solid lines) $-\frac{1}{2}$ and $-\frac{\alpha_1+1}{\alpha_1} = -\frac{3}{2}$ and $-\frac{5}{4}$ for $\alpha_1 = 2$ and 4 , respectively. Timescale is normalized by the diffusion time $\tau_0 = L^2/(4D_0)$. (b) Time evolution of the matrix-to-fracture flux multiplied by its asymptotic time dependencies (47) and divided by its asymptotic prefactors A_0 and A_∞ (48) and (49) for the same α_0 and α_1 exponents.

586 relation that gives rise to most dual-porosity models. The
 587 underlying proportionality factor may be obtained directly
 588 from the fixed-point solution that will be presented in Section
 589 6, and could be related to usual shape factors of the
 590 literature [25].

591 To finish with, it can be remarked that using the relation
 592 $\Phi_{mf}(t) = \frac{L}{2} \frac{d(S)}{dt}$, if $\alpha_1 = 0$ is inserted in that formula, it
 593 provides an exponential relaxation of the matrix saturation.
 594 That is consistent with the corresponding findings of the

constant diffusion case. In the non-linear case, a non-linear
 595 closure is obtained. Estimations of that closure for a whole
 596 range of time-scales accounting for both the short time and
 597 long time scales were proposed by [26].
 598

6. Generalization to 2D or 3D matrix blocks

599 We now generalize the results obtained in one dimension
 600 of space to any dimension of space for any matrix
 601

block geometry. Under the given boundary conditions, the porous medium still undergoes counter-current imbibition as demonstrated in Section 2.2 without the need for the matrix block to exhibit symmetries.

We first indicate in Sections 6.1 and 6.2 which quantitative features are preserved with respect to the one-dimensional early- and late-time asymptotic solutions and corresponding fluxes, and which are not and require additional investigation.

We show in Section 6.3 how the late-time asymptotic solution separate form (35) can be exploited to develop a simple and efficient fixed-point numerical solution method, which advantageously replaces the delicate solving of the initial nonlinear singular diffusion equation.

Finally, we demonstrate in Section 6.4, by considering some simple two-dimensional block geometries, that the full dynamics of capillary imbibition can be accurately and efficiently predicted. The matrix block imbibition still presents two regimes whose saturation and fracture-to-matrix flux can be computed: a diffusive early-time regime and an anomalous late-time regime that cross-over tightly. Eventually, residual points to investigate are discussed.

6.1. Early-time regime

In the general case of matrix blocks having arbitrary shapes, there is no simple approximation working yielding a detailed description of the short time regime. A generalization of Parlange's results might be worth investigating, although nothing simple is apparent at first sight. At very short time, keeping the Boltzmann assumption of a dependency on \mathbf{x}/\sqrt{t} is equivalent to set-up a boundary layer approximation [23, 24, 42]. Physically it corresponds to consider that locally the block boundary can be assumed to be planar, and then to use the one-dimensional solution. Such an approximation can be assumed to be valid if the diffusion length $\sqrt{D_0 t}$ is much smaller than any characteristic lengthscale of the block. The occurrence of such a regime is confirmed once considering the matrix-to-fracture flux that is observed hereafter in Fig. 11 of Section 6.4 to vary as $1/\sqrt{t}$ at short times for simple matrix block geometries. Then a transient regime leading to the long time asymptotic solution can be observed.

6.2. Late-time regime

As detailed in the next section, the late-time asymptotic solution in the separate form (35) remains valid for any dimension of space and any block shape, except that the function $y(x)$, whose analytical expression (45) has been derived in one dimension of space, and which we will henceforth denote $f(\mathbf{x})$, remains to be determined. Thus, except for this detail which we will show hereafter how to manage quickly and accurately numerically, the late-time asymptotic solution $1 - S(\mathbf{x}, t)$ preserves a time dependence in $1/t^{1/\alpha_1}$ and the corresponding asymptotic flux (47) still presents an algebraic anomalous decay that is proportional to $1/t^{(\alpha_1+1)/\alpha_1}$, whose prefactor A_∞ remains to be computed and no longer writes as in (49) as it obviously depends on the space solution $f(\mathbf{x})$.

6.3. Fixed-point algorithm

The late-time asymptotic approach (35) remains valid for any dimension of space and any block shape. The solution of (25) writes

$$1 - S(\mathbf{x}, t) = \frac{f(\mathbf{x})}{t^{1/\alpha_1}} \quad (50)$$

where $f(\mathbf{x})$ satisfies $\nabla \cdot (f^{\alpha_1} \nabla f) = -f/(\alpha_1 D_1)$, which is rewritten as already done in Eqs (36)-(39)

$$\nabla^2 h = -h^{\frac{1}{\alpha_1+1}}, \quad (51)$$

$$h = \left(\frac{\alpha_1 D_1}{\alpha_1 + 1} \right)^{\frac{\alpha_1 + 1}{\alpha_1}} f^{\alpha_1 + 1}. \quad (52)$$

As already noted in Section 4, h satisfies $\nabla^2 h = -1$ when the exponent α_1 is very large, whose solution is a parabola in one dimension. This will serve as an initial guess to implement a fixed-point method to solve (51).

We are thus led to solve a Laplace equation in a domain Ω of boundary $\partial\Omega$ on which $h = 0$. Specifically, a sequence of functions $(h_k)_{k \geq 0}$ such that

$$\begin{aligned} \nabla^2 h_k &= -h_{k-1}^{\frac{1}{\alpha_1+1}} & \text{in } \Omega, \\ h_k &= 0 & \text{on } \partial\Omega, \end{aligned} \quad (53)$$

for all $k \geq 1$ is looked for with the following initial guess

$$\begin{aligned} \nabla^2 h_0 &= -1 & \text{in } \Omega, \\ h_0 &= 0 & \text{on } \partial\Omega. \end{aligned} \quad (54)$$

At each iteration of the algorithm, we solve the non-singular elliptic problem (53) using a Newton-Raphson method embedded in the finite element method framework provided by the FreeFEM++ open-source PDE solver [19]. In 2D, that code generates meshes with triangular elements and Lagrangian P1 basis functions. The iterative algorithm is performed until $\|h_k - h_{k-1}\|_\infty$ and $\|h_k - h_{k-1}\|_2$ are small enough. Residuals less than 10^{-12} have been imposed so that a few iterations (about 20) are enough to converge which makes the algorithm fast and allows the use of fine meshes to obtain very accurate solutions. Also, as the initial guess corresponds to the solution of Eq. (54) with $\alpha_1 \rightarrow \infty$, a faster convergence is expected for higher values of α_1 for a given geometry.

6.4. Results

Running the algorithm in the one-dimensional case gives an excellent agreement with the analytical solution as reported in Fig. 4(d). A similar result has been obtained (but is not shown here as it does not bring much) by considering not the one-dimensional segment $[0, L]$ but a two-dimensional rectangle as shown in Fig. 6 with the same initial and boundary conditions, i.e. one pair of faces facing each other with imposed saturation and the other with zero flux.

Four two-dimensional geometries were considered in order to validate the fixed-point algorithm, considering the

696 saturation imposed on the whole boundary of the matrix
 697 block, from the most to the least symmetrical, as shown in
 698 Fig. 6:

- 699 • Disk of radius R ,
- 700 • Square of side L ,
- 701 • Rectangle of sides L_x and L_y ,
- 702 • Quadrangle of medians L_x and L_y .

703 For the record, the spherical geometry has also been treated
 704 but is not reported because except for some elementary algebra
 705 details it does not differ significantly from the cylindrical
 706 geometry shown hereafter (both are systems with one degree
 707 of freedom).

708 In each case, the diffusion time is defined by $\tau_0 =$
 709 $L_c^2/(4D_0)$ where L_c is a characteristic length of the consid-
 710 ered geometry, that is $L_c = 2R, L$ or L_x or L_y depend-
 711 ing on whether the medium is a disk, square, rectangle or
 712 quadrangle. We will come back to this point later, which
 713 at this stage is more of a dimensional analysis but requires
 714 further analysis as we shall see when analyzing the fluxes.
 715 The considered flow configuration is one of those previously
 716 studied in Sections 4 and 5 such that $\alpha_1 = 2, p = 3,$
 717 $M = 6, m = 2, k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25$ and
 718 $S_{wi} = S_{orw} = 0$.

719 Figs 7(a), 8(a), 9(a)-(b) and 10(a)-(b) report the satu-
 720 ration profiles $1 - S(\mathbf{x}, t)$ projected on the domain paths
 721 indicated in Fig. 6 while Figs 7(b), 8(b), 9(c)-(d) and 10(c)-
 722 (d) give the space dependence of the solution by plotting
 723 $[1 - S(\mathbf{x}, t)] t^{1/\alpha_1}$. In all cases, an excellent agreement be-
 724 tween the numerical and the fixed-point solution is observed
 725 for late times, specifically from $t/\tau_0 = 511$ for the disk,
 726 $t/\tau_0 = 511$ for the square, $t/\tau_0 = 5$ for the rectangle, and
 727 $t/\tau_0 = 838$ for the quadrangle.

728 Now let us look at the flux time evolution over the entire
 729 dynamic range. As already explained in Section 5, flux can
 730 be computed in two ways: either by the time derivative of
 731 the block average saturation, or by the flux at the boundary.
 732 Indeed, integrating Eq. (12) on the considered domain Ω ,
 733 defining the average saturation as $\langle S \rangle(t) = \frac{1}{|\Omega|} \int_{\Omega} S(\mathbf{x}, t) d\mathbf{x}$
 734 and using Stokes theorem, one gets a NAPL flux (of opposite
 735 sign to the water flux) of dimension $L^d T^{-1}$, where d is the
 736 space dimension, that reads

$$\Phi_{mf}(t) = -|\Omega| \frac{d\langle S \rangle(t)}{dt} = - \int_{\partial\Omega} D(S) \mathbf{n} \cdot \nabla S d\sigma \quad (55)$$

737 with \mathbf{n} the $\partial\Omega$ outward normal unitary vector and $|\Omega|$ the
 738 measure of Ω . In two (three) space dimensions $|\Omega|$ is Ω
 739 area (volume), $\partial\Omega$ is its contour (surface) and $d\sigma$ is an
 740 infinitesimal contour (surface) element of $\partial\Omega$.

741 Flux time evolution, that is more convenient to compute
 742 by the average saturation but which we made sure does not
 743 depend on the mode of calculation [using the left or right
 744 hand side of (55)], is reported in Fig. 11 for all the tested ge-
 745 ometries. Whatever the geometry considered, several points
 746 are worth noting:

- 747 • We observe once again, as in one dimension, an early-
 748 time diffusive regime that is characterized by a flux

749 which varies proportionally to $1/\sqrt{t}$, and an anoma-
 750 lous late-time regime that is driven by a flux propor-
 751 tional to $1/t^{(\alpha_1+1)/\alpha_1}$.

- 752 • Flux prefactor A_∞ is no longer known analytically
 753 as in one dimension, following Eq. (49), but can
 754 be quickly obtained numerically from the fixed-point
 755 algorithm subject to having an accurate estimate of a
 756 finite value of $(t/\tau_0)_\infty$ for which the numerical solu-
 757 tion has almost converged to the asymptotic solution
 758 for $t \rightarrow \infty$.
- 759 • The cross-over between the early- and late-time regimes
 760 is very narrow. Therefore the early- and late-time
 761 asymptotic solutions that dominate completely the
 762 fracture-to-matrix dynamic exchange are sufficient to
 763 determine the flux over the whole exchange dynam-
 764 ics. In other words, if the asymptotic solutions are
 765 determined, then the whole dynamic response of the
 766 exchange is also determined to a very good approx-
 767 imation. This remains of course to be demonstrated
 768 in practice on more complex geometries than those
 769 tested.
- 770 • Flux prefactor A_∞ can be computed numerically by
 771 a spatial integration of the fixed-point solution. This
 772 being said, Fig. 11 clearly shows that fluxes are all
 773 translated in time with respect to each other depending
 774 on the geometry considered. Only the square and
 775 the disk are very close. This means that the relevant
 776 characteristic length L_c involved in the diffusion time
 777 $\tau_0 = L_c^2/(4D_0)$ remains to be found, instead of setting
 778 as we did $L_c = 2R, L, L_x$ or L_y depending on whether
 779 the medium is a disk, square, rectangle or quadrangle.
 780 Once this one is found, if it exists, whatever the ge-
 781 ometry considered, all the corresponding fluxes would
 782 be in phase and would superimpose if $\Phi_{mf}(t)/A_\infty$ is
 783 considered for $t \rightarrow \infty$, following (47). This will be the
 784 subject of another work.

785 To finish with, the constant flux that can be observed at
 786 very short times in Fig. 11 for the square and the quadrangle
 787 should be discarded because they are artifacts related to the
 788 domain discretization, the exchange affecting the first row
 789 of cells near the domain boundary only. This was verified by
 790 further refining the mesh (the plateau shifts to even shorter
 791 times).

7. Summary and discussion 792

793 The long-time relaxation of the NAPL saturation inside a
 794 matrix block in contact with water was studied in the case of
 795 counter-current imbibition for one- or two-dimensional ma-
 796 trix blocks of arbitrary shape. Gravity effects were neglected,
 797 so only the pure capillary regime was investigated. The
 798 time variations of the matrix-to-fracture transfer flow rate
 799 was also studied. The transition from a Boltzmann square-
 800 root regime to an ‘‘anomalous’’ algebraic power-law decay
 801 was confirmed qualitatively and quantitatively by numerical
 802 simulations. In the short-time regime, the semi-infinite as-
 803 sumption is quite robust, so the results may remain useful

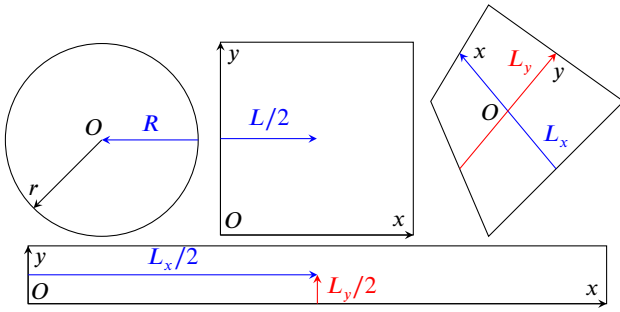


Figure 6: Two-dimensional geometries studied: disk, square, rectangle and quadrangle (all surfaces are to scale except the rectangle, magnified three times). The characteristic lengths as well as the paths on which the saturation field $1 - S(\mathbf{x}, t)$ is projected in Figs 7, 8, 9 and 10 are indicated in blue and red (direction of the path is given by an arrow, coordinate system is shown in black).

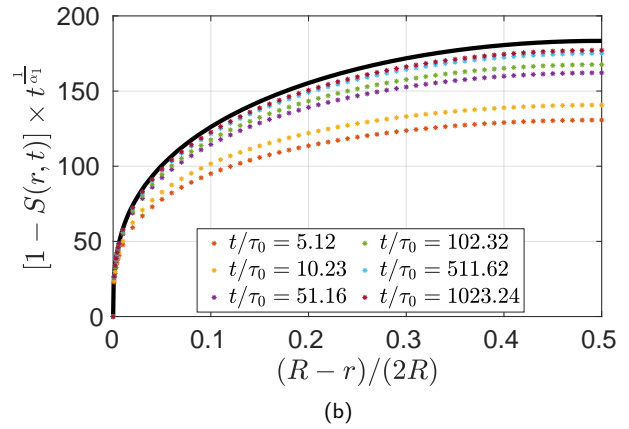
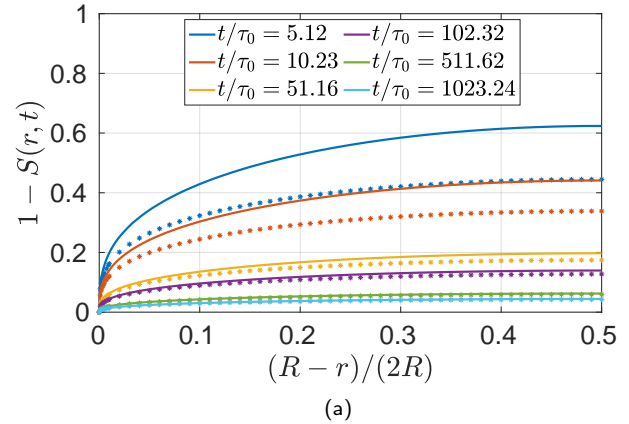


Figure 7: Disk — comparison of the fixed-point solution (solid lines, 1 654 elements) with the numerical solution (symbols, 500 nested rings). Timescale is normalized by the diffusion time $\tau_0 = L_c^2/(4D_0)$ with $L_c = 2R = 1$ m (see Fig. 6).

804 even considering general fractured media response in that
 805 regime. On the other hand, the long-time decay exponent is
 806 directly related to input data combining capillary pressure
 807 and relative permeabilities features. As in the long-time
 808 regime, the NAPL saturation is uniformly small inside the
 809 matrix block, the power-law assumption may be expected
 810 to be quite robust, controlled by the low NAPL saturation
 811 transport properties. The transition between both regimes
 812 is sharp, occurring as soon as the toe of the short-time
 813 Boltzmann solution reaches half the block size. The asso-
 814 ciated matrix-to-fracture flux term $\Phi_{mf}(t)$ closure may be
 815 represented as being proportional to $\langle S \rangle^{\alpha_1+1}$, the exponent
 816 α_1 describing the singularity of the diffusion coefficient with
 817 NAPL saturation.

818 For general matrix blocks in 2 dimensions, a similar
 819 study was carried out under the same counter-current flow
 820 regime. The occurrence of such regime appears to be quite
 821 general thanks to a derivation involving the global pressure
 822 concept. A similar Ansatz driving the long-time regime was
 823 proposed. It predicts a power-law time dependency, and the
 824 spatial dependence can be determined numerically using a
 825 fast fixed-point algorithm. These findings were confirmed
 826 by several comparisons with direct simulations for various
 827 typical matrix block shapes. Similar results are obtained for
 828 the time evolution of the corresponding matrix-to-fracture
 829 flux. A quite sharp transition between a Boltzmann regime
 830 involving a $1/\sqrt{t}$ diffusion time-dependence and the anom-
 831 alous long-time regime driven by the exponent $1/\alpha_1$ can
 832 be observed. That transition corresponds to the interplay
 833 between the diffusion lengthscale and the characteristic size
 834 of the matrix block.

835 In summary, a promising very fast and accurate compu-
 836 tational approach is emerging, which should eventually
 837 make it possible, after some additional efforts, to predict
 838 counter-current imbibition over a distribution of blocks of
 839 various shapes and sizes, which is more representative of
 840 naturally fractured porous media. To this end, the following
 841 points require further investigation:

- A generalization of Parlange's asymptotic early-time 842
 solution in any dimension of space might be worth 843
 investigating, although nothing simple is apparent at 844
 first sight. 845
- Late-time estimates of $(t/\tau_0)_\infty$, such that the fixed- 846
 point asymptotic solution obtained for $t \rightarrow \infty$ is valid 847
 and the flux prefactor A_∞ is accurately computed, 848
 should be consolidated. 849
- To that end, the relevant characteristic length involved 850
 in the diffusion time τ_0 should be determined be- 851
 forehand as previously discussed, to make $(t/\tau_0)_\infty$ 852
 estimates generic (indeed, if the timescale differs from 853
 one geometry to another, determining $(t/\tau_0)_\infty$ on a 854
 case by case basis may be tedious). 855

856 These findings pave the way to research leading to a more 857
 858 faithful description of matrix-to-fracture exchanges when 859
 860 considering a realistic fractured medium composed of a 861
 862 population of matrix block of various size and shapes. In 863
 particular, the proportionality constant relating the late-time
 flux $\Phi_{mf}(t)$ to $-\langle S \rangle^{\alpha_1+1}$ appears to play a similar role than a
 shape factor that may be evaluated using single-phase aver-
 aging techniques [37, 31, 33]. Looking for simple geometric

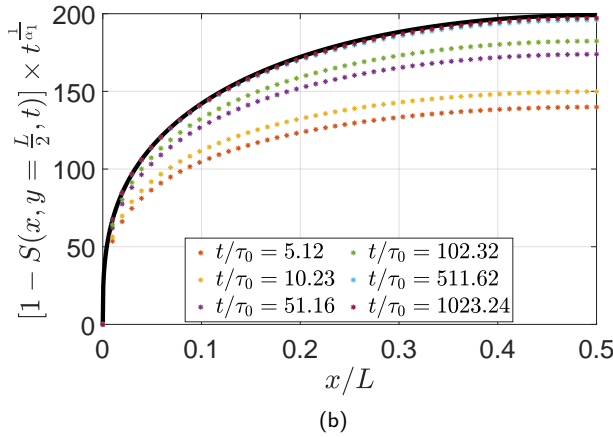
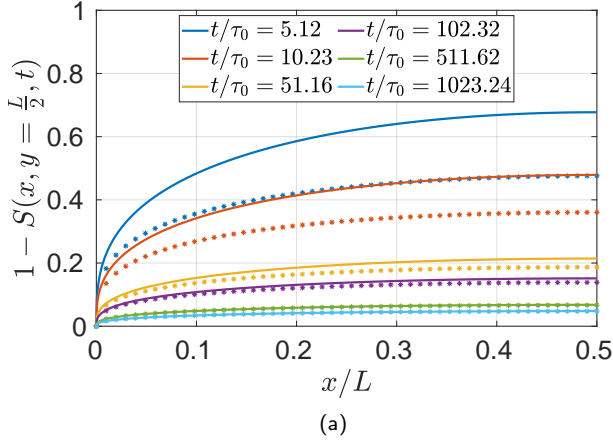


Figure 8: Square — comparison of the fixed-point solution (solid lines, 4050 elements) with the numerical solution (symbols, $101 \times 101 = 10201$ cells). Timescale is normalized by the diffusion time $\tau_0 = L_c^2/(4D_0)$ with $L_c = L = 1$ m (see Fig. 6).

descriptors quantifying the transition time between diffusion regimes will be of interest for such applications. In parallel, developing averaging methods lumping these various matrix blocks within one macro-scale single exchange flux description will be another research avenue.

A. Appendix: conservative numerical scheme

In this Appendix, we detail the numerical scheme used to solve the singular elliptic partial differential equation (12) in the matrix block domain Ω . We consider neither the Boltzmann variable form (29) for short times, nor the asymptotic form (23) for long times, but the generic PDE (12) that drives the solution $S(\mathbf{x}, t)$ for all \mathbf{x} and t . The singular character of this equation comes from the diffusion coefficient D given by Eq. (13) which vanishes and is not differentiable with respect to S , for $S = \{0, 1\}$, except for particular values of the exponents r , p and q . Because D cancels at the boundary $\partial\Omega$ of the porous medium, one must have $(\nabla S)_{\partial\Omega} = -\infty$ in order to get a non-zero fracture-to-matrix flux $(D(S)\nabla S)_{\partial\Omega}$, as can be seen in Fig. 3. To prevent numerical instabilities or large rounding errors, we set-up a conservative scheme

which does not require to compute the product $D(S)\nabla S$, and is based on the following reformulation of Eq. (12) as a conservation law:

$$\frac{\partial S}{\partial t} = \nabla^2 G(S) \quad \text{where} \quad G(S) = \int_0^S D(s) ds, \quad (\text{A1})$$

with S the normalized mobile aqueous phase saturation (8). The physical domain considered is Ω with boundary conditions $S(\mathbf{x}, t) = 1$ on the boundary $\partial\Omega$ for all $t \geq 0$.

To start with, the function G defined in Eq. (A1) does not admit a simple expression that can be easily handled, except in the limiting cases $M \ll 1$ and $M \gg 1$ (G is then a power-law or an incomplete beta function, respectively) where M is the mobility ratio defined in Eq. (19). To derive a conservative numerical scheme, we resort to a discrete integration of the diffusion function D denoted G^h . To do so, we discretize the normalized saturation interval $[0, 1]$ with the sequence $(\tilde{S}_k)_{k \in \{0, \dots, N_S\}} = k\Delta\tilde{S}$ composed of $N_S + 1$ points and where $\Delta\tilde{S} = 1/N_S$. For convenience, let us introduce the operator I_G^h which defines the discrete integral of G over two increasing saturation values S_α and S_β . Using the third order Simpson's method, I_G^h writes

$$I_G^h(S_\alpha, S_\beta) = \frac{S_\beta - S_\alpha}{6} \left[D(S_\alpha) + 4D\left(\frac{S_\alpha + S_\beta}{2}\right) + D(S_\beta) \right]. \quad (\text{A2})$$

For any saturation value S such that $S \in [\tilde{S}_k, \tilde{S}_{k+1}]$, we approximate the function $G(S)$ with $G^h(S) = I_G^h(\tilde{S}_k, S) + \sum_{l=1}^k I_G^h(\tilde{S}_{l-1}, \tilde{S}_l)$.

Regarding the domain Ω discretization, let \mathcal{M}_h be an admissible finite volume mesh of the domain Ω given by a family of control volumes or cells noted K : for any K of \mathcal{M}_h , $|K|$ is its measure, $KL = \partial K \cap \partial L$ is the common interface between K and a neighbouring cell L . The set of neighbors of cell K is denoted $\mathcal{N}(K)$, that is $\mathcal{N}(K) = \{L \in \mathcal{M}_h; \partial K \cap \partial L \neq \emptyset\}$. Time is discretized with the non-decreasing sequence $\{t^n\}$ such that $t^0 = 0$ and $\Delta t = t^{n+1} - t^n$. Let also S_i^n be the approximation of the saturation on the interval $C_i \times [t^n, t^n + \Delta t)$. Finally, it is useful to introduce the vector \mathbf{S}^n such that $(\mathbf{S}^n)_i = S_i^n$ in order to formulate the scheme vectorially.

Partial differential equation (A1) is recast in a discrete manner with the following finite-volume implicit scheme. Accounting for the boundary condition of an imposed saturation, i.e. $S(x = 0, t) = 1, \forall x \in \partial\Omega, \forall t \geq 0$, the numerical scheme can be written by introducing the function \mathbf{Z}^n defined for all $K \in \mathcal{M}_h$ as

$$[\mathbf{Z}^n(\mathbf{S})]_K = S_K - S_K^{n-1} - \sum_{L \in \mathcal{N}(K)} \lambda_{KL} (G_K - G_L) - \lambda_{K, \partial\Omega} (G_K - G(1)), \quad (\text{A3})$$

with $G_K = G(S_K)$, $\lambda_{KL} = \Delta t |KL| / (|K| d_{KL})$ where $|KL|$ is the face measure and d_{KL} the distance between the center of cells K and L . The term $\lambda_{K, \partial\Omega} (G_K - G(1))$ stands for the boundary condition contributions, non null when the cell K

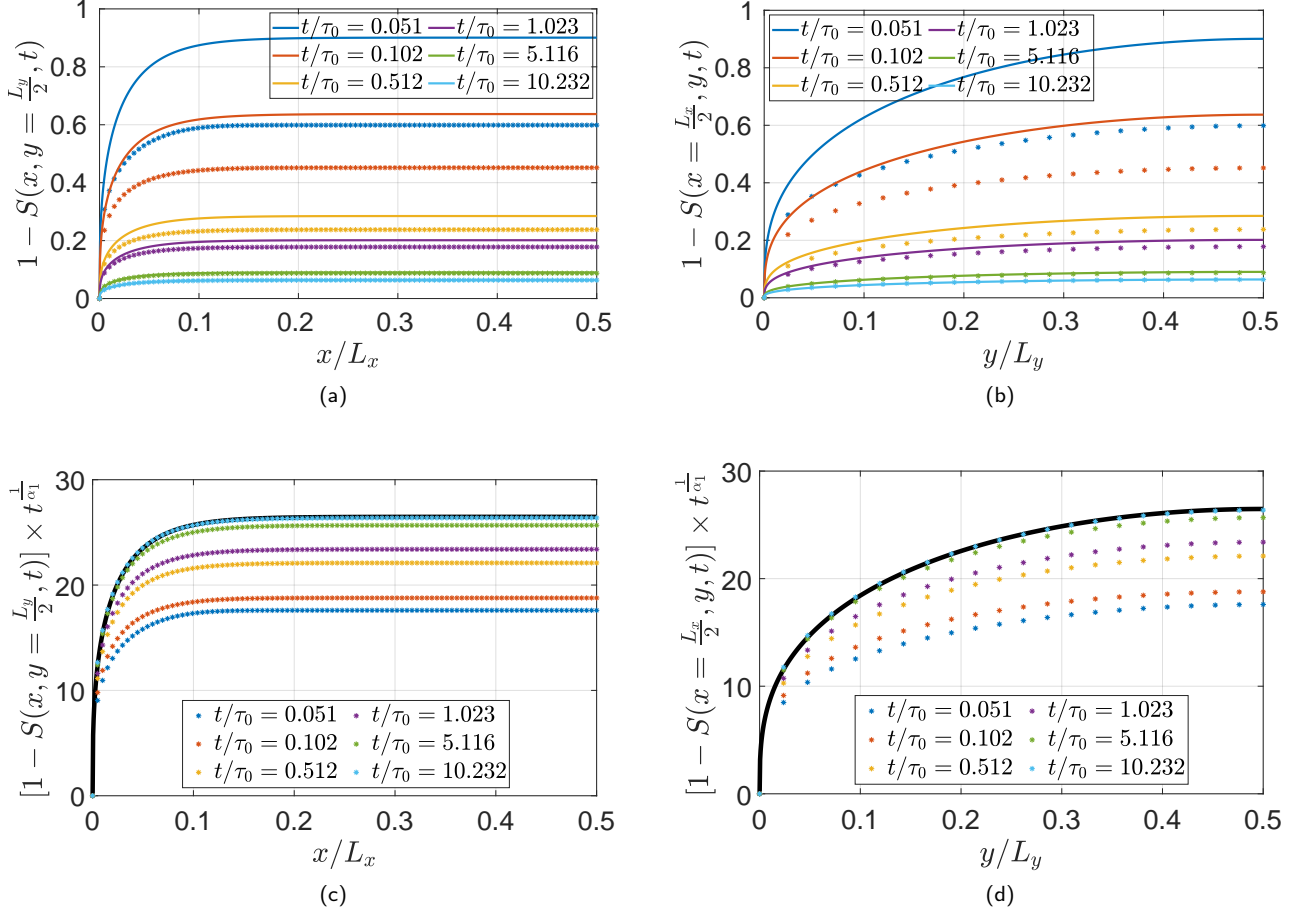


Figure 9: Rectangle — comparison of the fixed-point solution (solid lines, 2222 elements) with the numerical solution (symbols, $201 \times 41 = 8241$ cells). Timescale is normalized by the diffusion time $\tau_0 = L_c^2/(4D_0)$ with $L_c = L_x = 10 L_y = 1$ m (see Fig. 6).

928 is a boundary cell. As we use polyhedral mesh cells, it is the
 929 sum of several contributions if cell K has several boundary
 930 faces. For each boundary face $(\partial K)_j$, that contribution is
 931 $\Delta t |(\partial K)_j| / (|K| d_{K,(\partial K)_j})$ where $|(\partial K)_j|$ is the measure of
 932 the face and $d_{K,(\partial K)_j}$ the distance between the cell center
 933 and the face.

934 The updated numerical solution \mathbf{S}^n of this numerical
 935 scheme is the zero of the function \mathbf{Z}^n . As this function is
 936 non-linear because $G(S)$ non-linearly depends on saturation,
 937 a Newton-Raphson algorithm is required. Specifically, for
 938 each time step we construct a sequence $\mathbf{S}^{n,k}$ such that $\mathbf{Z}^{n,k} =$
 939 $\mathbf{Z}^n(\mathbf{S}^{n,k}) \xrightarrow{k \rightarrow \infty} 0$ and recursively defined by $\mathbf{S}^{n,k+1} = \mathbf{S}^{n,k} +$
 940 $\delta \mathbf{S}^{n,k}$ where $\delta \mathbf{S}^{n,k}$ denotes the increment vector given by

$$\delta \mathbf{S}^{n,k} = - [\nabla_{\mathbf{S}} \mathbf{Z}^{n,k}]^{-1} \mathbf{Z}^{n,k}, \quad (\text{A4})$$

941 where the matrix $[\nabla_{\mathbf{S}} \mathbf{Z}^{n,k}]^{-1}$ is the inverse of the Jacobian
 942 matrix $\nabla_{\mathbf{S}} \mathbf{Z}^{n,k} = \nabla_{\mathbf{S}} \mathbf{Z}^n(\mathbf{S}^{n,k})$. The increment vector $\delta \mathbf{S}^{n,k}$
 943 is thus determined by solving a linear system. The Jacobian
 944 matrix is sparse and its non-null terms are given by, for all

$K \in \mathcal{M}_h:$

$$\begin{aligned} (\nabla_{\mathbf{S}} \mathbf{Z}^{n,k})_{K,K} &= 1 - \left(\lambda_{K,\partial\Omega} + \sum_{L \in \mathcal{N}(K)} \lambda_{KL} \right) D_K^{n,k}, \\ (\nabla_{\mathbf{S}} \mathbf{Z}^{n,k})_{K,L} &= \lambda_{KL} D_L^{n,k}. \end{aligned} \quad (\text{A5})$$

946 In addition, as the function G is approximated numerically,
 947 we used the function G^h instead of G in relation (A3) to
 948 compute the Newton residue $\mathbf{Z}^{n,k}$. To end with, the Newton
 949 algorithm is performed until the L^2 norms of the residue
 950 $\|\mathbf{Z}^{n,k}\|_2$ and of the increment $\|\delta \mathbf{S}^{n,k}\|_2$ are sufficiently
 951 small.

A.1. Cylindrical geometry

952 For a cylindrical domain $\Omega = [0, R] \times [0, 2\pi] \times [0, H]$
 953 invariant by rotation and translation along the z -axis, the
 954 solution depends only on the radius $r \in [0, R]$. We therefore
 955 consider a uniform mesh composed of $N + 1$ embedded
 956 cylinder of first cell $K_0 = [0, \Delta r/2] \times [0, 2\pi] \times [0, H]$ and
 957 the following embedded $K_i = [(i - 1/2)\Delta r, (i + 1/2)\Delta r] \times$
 958 $[0, 2\pi] \times [0, H]$ for $1 \leq i \leq N_K$, where $\Delta r = R/(N_K + 1/2)$.
 959 After some straightforward calculations, the scheme writes
 960

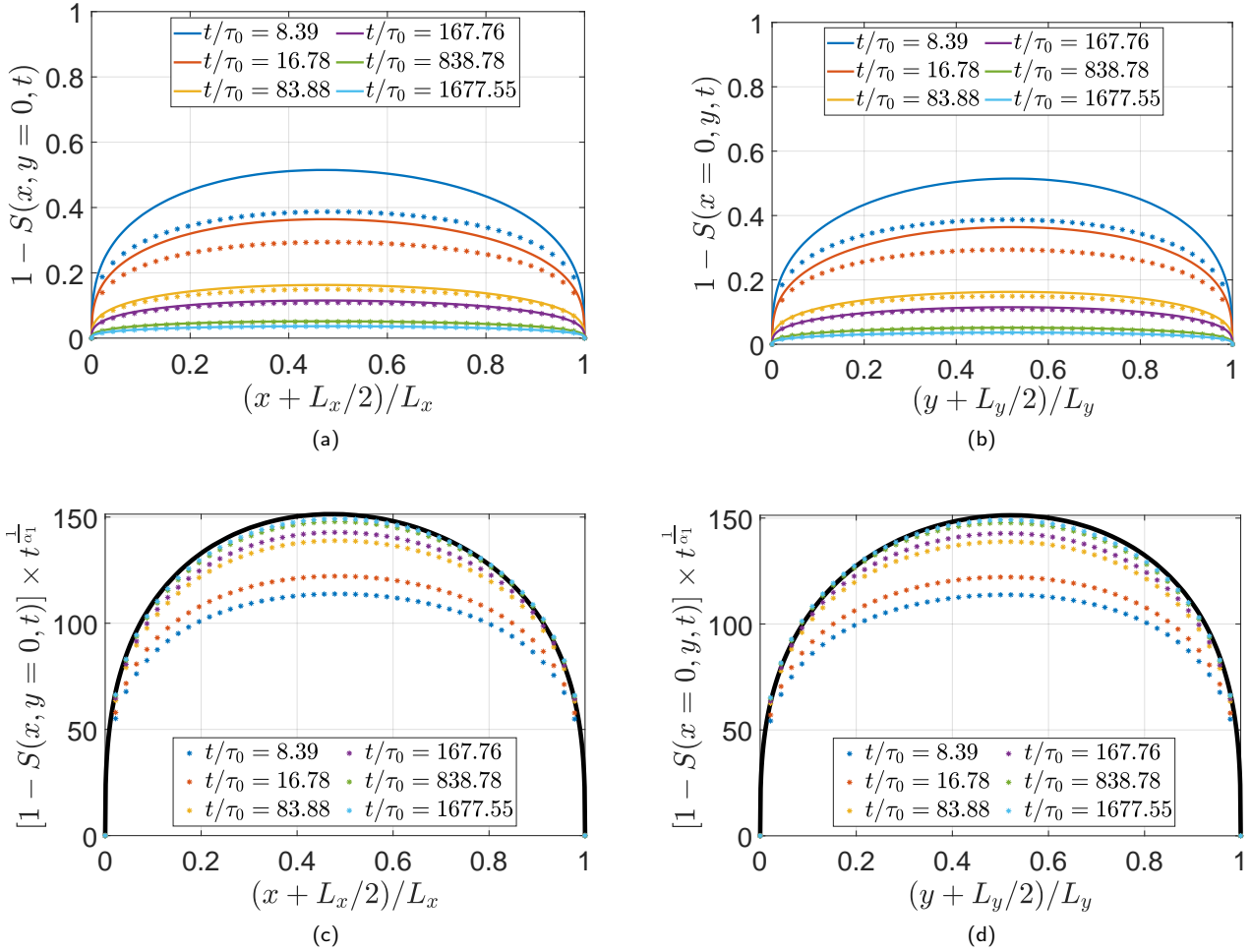


Figure 10: Quadrangle — comparison of the fixed-point solution (solid lines, 4 272 elements) with the numerical solution (symbols, $101 \times 101 = 10201$ cells). Timescale is normalized by the diffusion time $\tau_0 = L_c^2/(4D_0)$ with $L_c = L_x = L_y = 0.781$ m (see Fig. 6).

961 in the form (A3) with

$$\begin{aligned} \lambda_{K_0 K_1} &= \frac{4\Delta t}{(\Delta r)^2}, \\ \lambda_{K_i K_{i+1}} &= \left(1 + \frac{1}{2i}\right) \frac{\Delta t}{(\Delta r)^2} \quad \text{for } 1 \leq i \leq N_K, \quad (\text{A6}) \\ \lambda_{K_N, \partial\Omega} &= \frac{\Delta t}{\left[1 - \left(1 - \frac{\Delta r}{R}\right)^2\right] R \Delta r}. \end{aligned}$$

962 A.2. Spherical geometry

963 For a spherical domain $\Omega = [0, R]^3$ invariant by rotation,
964 we consider a uniform mesh composed of $N + 1$ embedded
965 sphere of first cell $K_0 = [0, \Delta r/2]^3$ and the following
966 embedded $K_i = [(i - 1/2)\Delta r, (i + 1/2)\Delta r]^3$ for $1 \leq i \leq N_K$,
967 with $\Delta r = R/(N_K + 1/2)$. In this case, the coefficients of
968 the numerical scheme (A3) are

$$\begin{aligned} \lambda_{K_0 K_1} &= \frac{6\Delta t}{(\Delta r)^2}, \\ \lambda_{K_i K_{i+1}} &= \frac{\left(i + \frac{1}{2}\right)^2}{\left(i + \frac{1}{2}\right)^3 - \left(i - \frac{1}{2}\right)^3} \frac{3\Delta t}{(\Delta r)^2} \quad \text{for } 1 \leq i \leq N_K, \quad (\text{A7}) \\ \lambda_{K_N, \partial\Omega} &= \frac{3\Delta t}{\left[1 - \left(1 - \frac{\Delta r}{R}\right)^3\right] R \Delta r}. \end{aligned}$$

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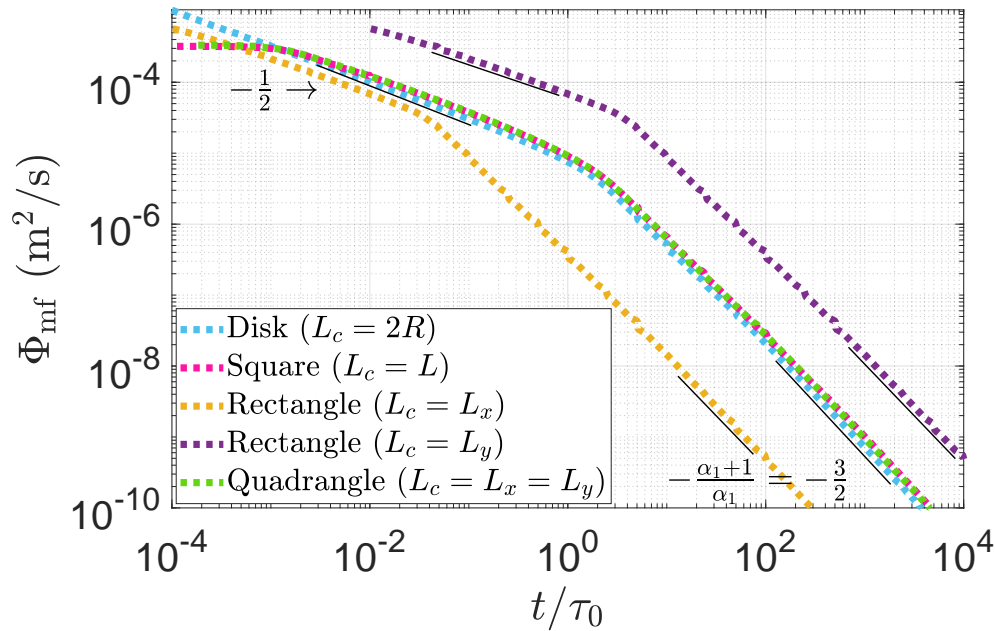


Figure 11: Time evolution of the simulated matrix-to-fracture flux for the considered two-dimensional geometries: disk, square, rectangle and quadrangle (see the text and Figs 7, 8, 9 and 10). Simulated early- and late-time slopes are in excellent agreement with the early- and late-time predictions (thin solid lines) $-\frac{1}{2}$ and $-\frac{\alpha_1+1}{\alpha_1} = -\frac{3}{2}$ for $\alpha_1 = 2$. Timescale is normalized by the diffusion time $\tau_0 = L_c^2/(4D_0)$ where the characteristic length L_c is set to $2R$, L or L_x or L_y depending on whether the medium is a disk (of radius R), square (of side L), rectangle (of sides L_x and L_y) or quadrangle (of medians L_x and L_y). The tested flow configuration is such that $\alpha_1 = 2$, $p = 3$, $M = 10$, $m = 2$, $k = 10$ mD, $P_e = 5$ bar, $\phi = 0.25$ and $S_{wi} = S_{orw} = 0$.

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1111 Nomenclature

1112 Dimensions of the quantities considered are given in paren-
1113 thesis with M = [mass], L = [length], T = [time],
1114 where [#] stands for dimension(s) of #

1115 Vectors are noted in bold

1116 D, mD Darcy, milli-Darcy (unit of permeability, L^2)

1117 1D, 2D, 3D One-/two-/three-dimensional

1118 NAPL Non-aqueous phase liquid

1119 t Time (T)

1120 $\tau_{0,m}$ Characteristic time (T)

1121 \mathbf{x} Position in space (L)

1122 x, y, z Cartesian space coordinates (L)

r	Radial coordinate (cylindrical/spherical geometries, L)	1123
ξ	Boltzmann variable $x/\sqrt{4D_0t}$ (dimensionless)	1125
∇	Gradient operator (L^{-1})	1126
∇^2	Laplacian operator (L^{-2})	1127
L_c	Characteristic length (L)	1128
$L, L_{x,y}$	Lengths (L)	1129
R	Radius (L)	1130
l_{cap}	Capillary length (L)	1131
ϕ	Porous medium porosity (dimensionless, fraction)	1132
k	Porous medium permeability (L^2)	1133
φ	Subscript denoting either water ($\varphi = w$) or NAPL ($\varphi = o$) phase	1134
ρ_φ	Density of phase φ (ML^{-3})	1136
$\Delta\rho$	Density difference $\rho_w - \rho_o$ between water and NAPL phases (ML^{-3})	1137
μ_φ	Dynamic viscosity of phase φ ($ML^{-1}T^{-1}$)	1139
γ	Interfacial tension (MT^{-2})	1140
θ	Contact angle between fluid and rock (dimensionless)	1141
S_φ	Saturation of phase φ (dimensionless, fraction)	1143
S_{wi}	Irreducible water saturation (dimensionless, fraction)	1144
S_{orw}	Residual NAPL saturation (dimensionless, fraction)	1146
S	Normalized water saturation $(S_w - S_{wi})/(1 - S_{orw} - S_{wi})$ (dimensionless, fraction)	1147
P_φ	Pressure in phase φ ($ML^{-1}T^{-2}$)	1149
P_c	Capillary pressure $P_o - P_w$ between water and NAPL phases ($ML^{-1}T^{-2}$)	1150
P_t	Global pressure ($ML^{-1}T^{-2}$)	1152
$k_{r\varphi}$	Relative permeability to phase φ (dimensionless)	1153
λ_φ	Mobility $k_{r\varphi}/\mu_\varphi$ of phase φ ($M^{-1}LT$)	1154
λ_t	Total mobility (sum of all considered phases mobilities, $M^{-1}LT$)	1155
\mathbf{u}_t	Total Darcy velocity (sum of all considered phases Darcy velocities, LT^{-1})	1157
κ_φ	Maximum relative permeability to phase φ (dimensionless)	1159
P_e	Entry capillary pressure ($ML^{-1}T^{-2}$)	1161
$p, q, r, m, \alpha_{0,1}, u, v$	Exponents (dimensionless)	1162
$D(S)$	Saturation S dependent diffusion function (L^2T^{-1})	1163
$D_{0,1}$	Constants that are homogeneous to a diffusion coefficient (L^2T^{-1})	1164
β, γ	Constants (dimensionless)	1166
M	End-point mobility ratio $(\mu_o\kappa_w)/(\mu_w\kappa_o)$ (dimensionless)	1167
$\langle \cdot \rangle$	Space averaging operator (dimensionless)	1169
Φ_{mf}	Matrix-to-fracture flux (LT^{-1})	1170
$A_{0,\infty}$	Matrix-to-fracture flux prefactors ($LT^{-1/2}$)	1171
$B(x; a, b)$	Euler incomplete beta function	1172
$B(x, y)$	Euler beta function	1173
$\Gamma(z)$	Euler gamma function	1174
σ	Shape factor (dimensionless)	1175
$\Omega, \partial\Omega, \mathbf{n}$	Domain, domain boundary, domain boundary outward normal unitary vector	1176
$ \Omega $	Measure of domain Ω (L^3, L^2 or L depending on whether Ω is a volume, a surface or a path)	1178

