Supporting information

Exact Analytical Solutions of Counter-Current Imbibition with both Capillary and Gravity Effects

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Appendix A

Referring to Equation (1) in the text as fundamental equation of continuity, for twophase immiscible, incompressible, isotherm flow through a rigid 3D porous medium, the fluid phase α satisfies P = $\rho_{\alpha} \dot{q}_{\alpha}$ and Equation (1) becomes:

$$\varphi \frac{\partial (\rho_{\alpha} S_{\alpha})}{\partial t} = -\nabla (\rho_{\alpha} q_{\alpha}), \qquad S_n + S_w = 1$$
(A-1)

 S_{α} is the phase saturation, and q_{α} is the phase velocity which is given by the extended Darcy equation [45]:

$$\stackrel{\mathbb{I}}{q}_{\alpha} = -\lambda_{\alpha}(S_{\alpha}, C) \stackrel{\mathbb{I}}{K} (\nabla p_{\alpha}(S_{\alpha}, C) - \rho_{\alpha} \stackrel{\mathbb{I}}{g})$$
(A-2)

Mobility $\lambda_{\alpha} = k_{r\alpha}(S_{\alpha}, C)/\mu_{\alpha}(C)$ is the ratio of relative permeability $k_{r\alpha}$ of phase α to its viscosity, μ_{α} . Mobility ratio describes the impairment of the flow of one phase by the other phase. K is the permeability tensor of the porous medium, p_{α} is the fluid pressure of phase α (which is defined through capillary pressure as $p_c = p_n - p_w$) and g is the gravitational acceleration vector.

It should be noted that Equations (1) and (A-1) can be derived from first principles [45], while equation (A-2) is an assumption.

The parameter C in Equation (A-2) represents chemical composition (water salinity, polymer, additives, tracers, etc.), with the following assumptions:

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- i. Components do not alter the porous medium (through chemical reactions);
- ii. They do not partition into the other phase;
- iii. Solute mass flux due to hydrodynamic dispersion within a phase is described by a Fickian model;
- iv. The volume fraction of the chemical composition is small compared to that of the wetting phase;
- v. The only chemical interaction between the rock and the components is equilibrium adsorption [46].

The flux of any component C_i includes contributions from advection and dispersion [47]:

$$\mathfrak{I}_{C_i} = C_i F_w - \varphi \mathfrak{S}_w \overset{\mathsf{d}}{D} \nabla C_i \tag{A-3}$$

where D is the hydrodynamic dispersion tensor [47]:

$$\overset{\mathsf{u}}{D} = \alpha_T \| \overset{\mathsf{u}}{q}_w \| \delta_{i,j} + (\alpha_L - \alpha_T) \overset{\mathsf{u}}{q}_{w,i} \overset{\mathsf{u}}{q}_{w,j} / \| \overset{\mathsf{u}}{q}_w \|$$
(A-4)

 $\delta_{i,j}$ is the Kronecker delta, α_L and α_T are longitudinal and transversal dispersivity, respectively, which are measured through experiments. D accounts for the effects of velocity fields, but the effect of molecular diffusion is ignored.

For a system of n components, there are n advection-dispersion-reaction (ADR) equations:

$$\phi \frac{\partial}{\partial t} (\rho_w C_i S_w) + \nabla \cdot (\rho_w C_i q_w) = \nabla \cdot (\phi S_w \rho_w D \nabla C_i) - \frac{\partial [(1-\phi)\rho_r A_{s,i}\rho_w]}{\partial t}$$
(A-5)

The rock density is ρ_{r_i} and the adsorption of C_i per unit mass of rock is presented by $A_{s,i}$. In addition, when water fluid properties stay constant, the tracer concentration will be constant (e.g., $\rho_w(C) = \rho_w = const$. and $\mu_w(C) = \mu_w = const$.). For chemical components (polymer or salts), empirical adsorption relations can be used [48] as below:

$$\Gamma \coloneqq \frac{(1-\varphi)}{\varphi} \rho_r A_s \tag{A-6}$$

At thermodynamic equilibrium, isotherm is a function of C only (i.e. $\Gamma = \Gamma(C)$). Furthermore, variations in density (due to the presence of chemical components) are considered in the gravity term only.

Equation (A-2) combined with the definition of capillary pressure results in a parabolic Partial differential Equation for water pressure [49]:

$$\varphi c_t \frac{\partial p_w}{\partial t} = \nabla \left(\overset{\mathbb{I}}{K} \lambda_t (S_w, C) \nabla p_w - \overset{\mathbb{I}}{K} \lambda_n (S_w, C) \nabla p_c - \left(\rho_w \lambda_w (S_w, C) + \rho_n \lambda_n (S_w, C) \right) \overset{\mathbb{I}}{g} \right) \quad (A-7)$$

where c_t is the total compressibility of the fluid-rock system, $\lambda_t = \lambda_w + \lambda_n$, and $\lambda_\alpha = k_{r\alpha}(S_\alpha, C)/\mu_\alpha(C), \alpha \in \{w, o\}.$

The fractional flow function is also introduced as:

$$f_{w}(S_{w},C) = \left(1 + \frac{k_{rn}(S_{w})\mu_{n}}{k_{rw}(S_{w},C)\mu_{w}(C)}\right)^{-1} = \left(\frac{\frac{k_{rw}}{\mu_{w}}}{\frac{k_{rw}}{\mu_{w}} + \frac{k_{rn}}{\mu_{n}}}\right)$$

$$f_{w}(S_{w},C) = \frac{\lambda_{w}}{\lambda_{w} + \lambda_{n}} = \frac{\lambda_{w}}{\lambda_{t}}$$
(A-8)

The wetting and non-wetting relative permeability functions depend on saturation (S_w) only, and numerous correlations have been proposed for them, already.

The total velocity is $\overset{u}{q}_{t} = \overset{u}{q}_{w} + \overset{u}{q}_{n}$. The set of equations is obtained as:

$$\begin{cases} \mathbf{r}_{w} = f\mathbf{q}_{t} - K \frac{fk_{ro}}{\mu_{n}} \nabla p_{c} + \frac{\lambda_{w}\lambda_{n}}{\lambda_{t}} (\rho_{w} - \rho_{n}) \mathbf{g} \\ \phi \frac{\partial S_{w}}{\partial t} = -\nabla \cdot (f\mathbf{q}_{t}) + \nabla \cdot \left(K \frac{fk_{ro}}{\mu_{n}} \nabla p_{c} \right) - \nabla \cdot \left(\frac{\lambda_{w}\lambda_{n}}{\lambda_{t}} (\rho_{w} - \rho_{n}) \mathbf{g} \right) + \frac{P}{\rho_{w}} \\ \phi \frac{\partial (CS_{w})}{\partial t} + \frac{\partial \Gamma}{\partial t} = -\nabla \cdot (Cf(S_{w}, C)\mathbf{q}_{t}) - CK \frac{fk_{ro}}{\mu_{n}} (S_{w}, C) \nabla p_{c} \\ + C \frac{\lambda_{w}\lambda_{n}}{\lambda_{t}} (S_{w}, C) K \nabla \cdot ((\rho_{w}(C) - \rho_{n})\mathbf{g}) + \nabla \cdot (\phi S_{w} D \nabla C) \end{cases}$$
(A-9)

When making the additional assumptions below for previously suggested models:

- i. No dissolved components exist, C = 0;
- ii. The porous medium is homogeneous;
- iii. The medium is one-dimensional and horizontal so that gravity can be ignored;
- iv. There is no sink or source term.

Applying these assumptions, the final equation form would be like below:

$$\varphi \frac{\partial S_w}{\partial t} = -q_t \frac{df}{dS_w} \frac{\partial S_w}{\partial x} + \frac{\partial}{\partial x} \left(D(S_w) \frac{dp_c}{dS_w} \frac{\partial S_w}{\partial x} \right)$$
(A-10)

 $D(S_w)$ has also the following equation form:

$$D(S_w) = -K \frac{\lambda_w \cdot \lambda_o}{\lambda_t} \frac{dP_c}{dS_w}$$
(A-11)

Appendix B

The net accumulated volume of wetting fluid is:

$$V_{w}(t) = \int_{0}^{t} (q_{0}(\tau) - f(S_{wi})q_{t})d\tau = \int_{0}^{t} q_{0}(\tau)(1 - f(S_{wi})R)d\tau = 2A(1 - f(S_{wi})R)t^{+1/2}$$
(B-1)

where R is the ratio of total flow to water flux at x = 0 ($R = q_t/q_0$). Clearly, for unidirectional displacement, $q_t = q_0$ and R = 1. For counter-current flow, total flux is equal to zero and R = 0.

The total imbibed volume of water would be:

$$Q_{w}(t) = \int_{0}^{t} q_{w}(x=0,t) dt = 2A\sqrt{t}$$
(B-2)

The magnitude of A determines the speed of SI, and it is a function of the rock-fluid system. The fractional flow functions for spontaneous imbibition are [1]:

$$F(x,t) = \frac{q_w/q_0 - f_i R}{1 - f_i R}$$
(B-3)

With R = 0 for counter-current imbibition and Equation (B-3) and Equation (11) turns into:

$$F(x,t) = \frac{q_w}{q_0} \Longrightarrow q_w = F \cdot q_0 = \frac{FA}{\sqrt{t}}$$
(B-4)

When considering Equation (A-2), the general 1D continuity equation simplifies to:

$$\varphi \frac{\partial S_w}{\partial t} = -\frac{\partial q_w}{\partial x} \tag{B-5}$$

Substituting Equation (B-4) into Equation (B-5), and using the similarity variable $\omega = x/\sqrt{t}$:

$$\varphi \frac{\partial S_{w}}{\partial \omega} \frac{\partial \omega}{\partial t} = -\left(\frac{A}{\sqrt{t}}\right) \frac{\partial F}{\partial S_{w}} \frac{\partial S_{w}}{\partial \omega} \frac{\partial \omega}{\partial x} \Rightarrow \varphi \frac{dS_{w}}{d\omega} \left(-\frac{\omega}{2t}\right) = -\left(\frac{A}{\sqrt{t}}\right) \frac{dF}{dS_{w}} \frac{dS_{w}}{d\omega} \left(\frac{1}{\sqrt{t}}\right)$$
$$\Rightarrow \omega = \frac{2A}{\varphi} \frac{dF}{dS_{w}}$$
(B-6)

The derivative of Equation (B-6) is:

$$\frac{d\omega}{dS_w} = \frac{2A}{\varphi} \frac{d^2 F}{dS_w^2}$$
(B-7)

Now consider Equation (A-9) for q_w (where $q_t = 0$):

$$q_{w} = fq_{t} - D(S_{w}) \xrightarrow{\partial S_{w}} \xrightarrow{q_{t}=0} q_{w} = -D(S_{w}) \frac{\partial S_{w}}{\partial x}$$
(B-8)

Combined with Equation (B-4):

$$q_{w} = \frac{F \cdot A}{\sqrt{t}} = -D(S_{w}) \frac{\partial S_{w}}{\partial \omega} \frac{\partial \omega}{\partial x} \Rightarrow \frac{F \cdot A}{\sqrt{t}} = -D(S_{w}) \frac{dS_{w}}{d\omega} \left(\frac{1}{\sqrt{t}}\right)$$

$$\Rightarrow \frac{dS_{w}}{d\omega} = -\frac{F \cdot A}{D(S_{w})} \quad \rightarrow \quad \frac{d\omega}{dS_{w}} = -\frac{D(S_{w})}{F \cdot A}$$
(B-9)

Inserting Equation (B-9) in Equation (B-7):

$$-\frac{D(S_w)}{F \cdot A} = \frac{2A}{\phi} \frac{d^2 F}{dS_w^2} \Longrightarrow \frac{d^2 F}{dS_w^2} = -\frac{\phi}{2A^2} \frac{D(S_w)}{F}$$
(B-10)

This is an Ordinary Differential Equation (ODE) with respect to F. The solution for Equation (B-10) can be obtained by integrating twice and applying initial and boundary conditions:

$$F(S_{w}) = 1 - \frac{\int_{S_{w}}^{S_{0}} \frac{(\beta - S_{w})D}{F} d\beta}{\int_{S_{i}}^{S_{0}} \frac{(S_{w} - S_{i})D}{F} dS_{w}}$$
(B-11)

Appendix C

Since F also depends on the constant A, iteration for A is required; in this case, an initial estimation of A is required. As F = 1 at the inlet, iteration of A is repeated until F(x=0,t)=1. The backward derivative of F" is as follows:

$$F''(S_w) \approx \frac{F(S_w + 2\Delta S_w) - 2F(S_w + \Delta S_w) + F(S_w)}{\Delta S_w^2}$$
(C-1)

Considering Equation (B-9) and rearranging for counter-current imbibition, we have:

$$F(S_w) = \left[F(S_w + \Delta S_w) - 0.5F(S_w + 2\Delta S_w)\right] + \sqrt{\left[F(S_w + \Delta S_w) - 0.5F(S_w + 2\Delta S_w)\right]^2 - \left(\frac{\varphi}{2A^2}\right)D(S_w)\Delta S_w^2}$$
(C-2)

When using the numerical form as in Equation (C-2), we can find an approximation of $F(S_w)$ knowing $F(S_w + \Delta S_w)$ and $F(S_w + 2\Delta S_w)$ beforehand, based on some starting value of $F(S_{w,Start})$. The value of F(x = 0, t) = 1 is considered as that starting value. The other required value of $F(S_{w,max} - \Delta S_w)$ is obtained by expansion into a Taylor series:

$$F\left(S_{w,\max} - \Delta S_{w}\right) = F\left(S_{w,\max}\right) - \Delta S_{w} \cdot F'\left(S_{w,\max}\right) = 1 - 0 = 1$$
(C-3)

It is assumed that the maximum saturation is always provided at the inlet and consequently $F'(S_{w,max}) = 0$.

Convergence of F is achieved by applying two criteria, firstly, as F is a fractional flow function and we have no flow at initial water saturation (S_{wir}) , then $F(S_{wir}) = 0$; secondly, due to the mass balance condition, the integrated saturation curve must equal the total imbibed pore volume, i.e.:

$$\int_{S_{wir}}^{S_{w,max}} x(S_w, t) dS_w = \frac{Q_w(t)}{\varphi} = \frac{2A\sqrt{t}}{\varphi}$$
(C-4)

Again, a numerical approximation is applied for Equation (C-4) to give:

$$\sum_{i=1}^{n} F'(S_w, i) \cdot \Delta S_w \approx \frac{Q_w(t)}{\varphi} = \frac{\varphi}{2A\sqrt{t}} = 1$$
(C-5)

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