

# Well-posedness of the single-cell transport problem for a two-phase flow with polymer

Xavier Raynaud, Knut-Andreas Lie  
Halvor Møll Nilsen, Atgeirr Rasmussen

October 14, 2014

## Abstract

A sequential splitting of the pressure and transport equations applied to a compressible two-phase flow with polymer leads to a considerable speed-up of the simulation, see [1]. To avoid excessive limitation on the size of the time step, the transport equations are solved implicitly. By using an iterative transport solver, in which the transport equations are solved cell-by-cell from upstream we can further decrease the computation time significantly. Such approach requires a robust single-cell transport solver. The single-cell problem consists of computing the saturation and the polymer concentration in a cell, given the total flux, the saturation, and the polymer concentration in the neighbouring cells. We derive a splitting and a discretization of the mass-conservation equations for which the single-cell problem is always well defined - for any time step size. We are now able to handle the compressible case, which requires a careful choice of the pressure equation and the segregation case, which requires to use a mixed upwind/downwind evaluation of the polymer concentration in the computation of the numerical flux.

- [1 ] Lie K.A, Nilsen H., Rasmussen A, Raynaud X. Fast simulation of polymer injection in heavy-oil reservoirs based on topological sorting and sequential splitting SPE J., 2014. DOI: 10.2118/163599-PA.

## Introduction

In reservoir with highly viscous oil, fingering effects lead to water penetrating easily the most permeable parts of the oil region. In this case, early water breakthrough letting a large fraction of the reservoir unswept implies a considerable loss in oil recovery. To avoid this situation, enhanced oil recovery strategies have been developed and polymer injection is one of the most important. The polymer is used to increase the water viscosity ratio and establish a more favorable mobility ratio between the two phases. Polymer are miscible in water but, in realistic reservoir models, the coarseness of the grid does not allow for a correct computation of the mixing zone. Instead, we have to resort to averaging models (see Booth (2008); Koval (1963)) and the most popular is the Todd-Longstaff mixing model (Todd and Longstaff, 1972), which is used in several commercial simulator, as Eclipse and CMG. In this paper, we consider a two phase flow problem with polymer injection. In addition to the Todd-Longstaff mixing model, we also include permeability reduction effects, polymer adsorption and dead pore space.

Fully implicit solvers are usually preferred for their robustness but may, in some context, be computationally too expensive. An alternative is to use splitting techniques (see e.g. Holden et al. (2010)). Here, we split the governing equations consisting of the mass conservation equations for water, oil and polymer into a pressure equation and two transport equations for water saturation and polymer concentration. The pressure and transport equations are then solved sequentially. In (Lie et al., 2013), the authors study an iterative implicit transport solver, where the nonlinear transport equations for saturation and concentration are solved cell by cell. The optimal configuration for such solver is when all the cells can be reordered following the direction of the flow, as the computations are done only once for each cell (Natvig and Lie, 2008). When using a two point flux approximation and in the absence of gravity, the pressure values in each cell directly provides us directly with an ordering of the cells so that, in this case, total reordering is always guaranteed. This aspect makes attractive a further splitting of the transport equation into a Darcy component and a gravity component. Moreover, from the physical point of view, polymer is used in heavy oil reservoir so that the effect of gravity segregation are not so strong. When the cells cannot be reordered, one can use a nonlinear Gauss-Seidel algorithm which iterates until convergence over each block of cells that are made inter-dependent by the orientation of the flux. When the cells can be reordered, the algorithm we use is still a nonlinear Gauss-Seidel algorithm, which converges in one iteration. In the case of a two phase flow problem (without polymer), it is shown in (Kwok and Tchelepi, 2008) that a non-linear Gauss-Seidel algorithm for the transport equation is globally convergent, that is, converges from any given starting point. In (Lie et al., 2013), we show the good scalability properties of this scheme and its computational efficiency.

The nonlinear Gauss-Seidel algorithm relies heavily on a robust solver for the single-cell problem. The single-cell problem consists of computing the saturation and concentration in a given cell, knowing the saturation and concentration in the neighboring cell and the total flux. The total flux is obtained in the pressure step where the pressure equation is solved. Mathematically, the single-cell equations is a set of two non-linear equations whose particular form depends on the time step, the geometry, the fluid and rock properties, the saturation and concentration values of the neighboring cells and the total flux. The main achievement in this paper is to show that the single cell problem is always well-posed and, in particular, for any time step length. By well-posedness, we mean that there exists a solution and that the solution is unique. In the case of the Darcy component, we extend the results obtained in (Lie et al., 2013) for the case with equal fluid compressibilities to the case with different compressibilities, assuming that one of the two phases is always more compressible than the other. This assumption is not too restrictive as oil is typically always more compressible than water. The well-posedness result relies on some properties of the pressure equation, which has to be chosen with care. For the gravity component, we use a standard phase upstreaming for saturation but for the polymer concentration, the problem is more delicate. At first, we consider the water upstream direction to evaluate the polymer concentration on the faces but this choice does not lead to the well-posedness of the solution. We present an alternative choice, mixing both upstream and downstream values, which guarantees well-posedness.

## Polymer Model

Our starting point is the mass conservation equations for oil, water, and polymer

$$\frac{\partial}{\partial t}(\rho_\alpha \phi S_\alpha) + \nabla \cdot (\rho_\alpha \vec{v}_\alpha) = 0, \quad \alpha \in \{w, o\}, \quad (1)$$

$$\frac{\partial}{\partial t}(\rho_w \phi S_w c) + \nabla \cdot (c \rho_w \vec{v}_{wp}) = 0. \quad (2)$$

Here,  $\rho_\alpha$ ,  $\vec{v}_\alpha$ , and  $S_\alpha$  denote density, velocity, and saturation of the phase  $\alpha$ . The porosity is denoted by  $\phi$  and is assumed to be a function  $\phi(p)$  of pressure only,  $c$  is the polymer concentration, and  $\vec{v}_{wp}$  the velocity of water containing diluted polymer. Sources and sinks may be included in a manner equivalent to boundary conditions, and are left out of the above equations.

To model the viscosity change of the mixture, we use the Todd–Longstaff model (Todd and Longstaff, 1972). This model introduces a mixing parameter  $\omega \in [0, 1]$  that takes into account the degree of mixing of polymer into water. Assuming that the viscosity  $\mu_m$  of a fully mixed polymer solution is a function of the concentration, the effective polymer viscosity is defined as

$$\mu_{p,\text{eff}} = \mu_m(c)^\omega \mu_p^{1-\omega} \quad \text{with} \quad \mu_p = \mu_m(c_{\max}). \quad (3)$$

The viscosity of the partially mixed water is given in a similar way by

$$\mu_{w,e} = \mu_m(c)^\omega \mu_w^{1-\omega}. \quad (4)$$

The effective water viscosity  $\mu_{w,\text{eff}}$  is defined by interpolating linearly between the inverse of the effective polymer viscosity and the partially mixed water viscosity

$$\frac{1}{\mu_{w,\text{eff}}} = \frac{1 - c/c_{\max}}{\mu_{w,e}} + \frac{c/c_{\max}}{\mu_{p,\text{eff}}}. \quad (5)$$

To take the incomplete mixing into account, we introduce the velocity of water that contains polymer, which we denote  $\vec{v}_{wp}$ . For this part of the water phase, the relative permeability is assumed to be equal to  $k_{rw}$  and the viscosity is equal to  $\mu_{p,\text{eff}}$ . We also consider the total water velocity, which we still denote  $\vec{v}_w$  and for which the viscosity is given by  $\mu_{w,\text{eff}}$ . Darcy's law then gives us

$$\vec{v}_w = -\frac{k_{rw}}{\mu_{w,\text{eff}} R_k(c^a)} \mathbf{K}(\nabla p - \rho_w g \nabla z), \quad (6)$$

$$\vec{v}_{wp} = -\frac{k_{rw}}{\mu_{p,\text{eff}} R_k(c^a)} \mathbf{K}(\nabla p - \rho_w g \nabla z) = m(c) \vec{v}_w, \quad (7)$$

as we assume that the presence of polymer does not affect the pressure and the density. The polymer mobility factor  $m(c)$  is defined as

$$m(c) = \frac{\mu_{w,\text{eff}}}{\mu_{p,\text{eff}}}$$

and, after some simplifications, we get

$$m(c) = \left[ \left(1 - \frac{c}{c_{\max}}\right) \left(\frac{\mu_p}{\mu_w}\right)^{1-\omega} + \frac{c}{c_{\max}} \right]^{-1}. \quad (8)$$

The function  $R_k(c^a)$  denotes the actual resistance factor and is a non-decreasing function which models the reduction of the permeability of the rock to the water phase due to the presence of absorbed polymer. The concentration of absorbed polymer is denoted by  $c^a$ . We introduce the total flux as  $\vec{v} = \vec{v}_w + \vec{v}_o$ . We have

$$\vec{v} = -(\lambda_w + \lambda_o) \mathbf{K} \nabla p + g(\lambda_w \rho_w + \lambda_o \rho_o) \mathbf{K} \nabla z$$

and, after some computation, we obtain the following expression of  $\vec{v}_\alpha$  as a function of  $\vec{v}$

$$\vec{v}_w = f_w \vec{v} + \vec{v}_g \quad \text{and} \quad \vec{v}_o = f_o \vec{v} - \vec{v}_g \quad (9)$$

with

$$\vec{v}_g = \frac{\lambda_w \lambda_o}{\lambda_w + \lambda_o} (\rho_w - \rho_o) g \mathbf{K} \nabla z. \quad (10)$$

Here,  $\lambda_\alpha$  denotes the mobility of phase  $\alpha$ , i.e.,

$$\lambda_w = \frac{k_{rw}}{\mu_{w,\text{eff}} R_k(c^a)} \quad \text{and} \quad \lambda_o = \frac{k_{ro}}{\mu_o},$$

and  $f_\alpha$  corresponds to the fractional flow,  $f_\alpha = \lambda_\alpha / (\lambda_w + \lambda_o)$ . The time scale of adsorption is much larger than that of mass transport and we will assume that adsorption takes place instantaneously so that  $c^a$  is a function of  $c$  only. The reference rock density is  $\rho_{r,\text{ref}}$  and the reference porosity  $\phi_{\text{ref}}$ . The adsorption of polymer is then taken into account by replacing (2) by

$$\frac{\partial}{\partial t} (\rho_w \phi S_w c) + \frac{\partial}{\partial t} (\rho_{r,\text{ref}} (1 - \phi_{\text{ref}}) c^a) + \nabla \cdot (c \rho_w \vec{v}_{wp}) = 0. \quad (11)$$

It is natural to assume that  $c^a$  is an increasing function of  $c$ . Polymer cannot reach the smallest pores and, as a result, the effective pore volume for the polymer solution is smaller than the pore volume of the rock. This effect can be modeled by replacing (11) with

$$\frac{\partial}{\partial t} (\rho_w \phi (1 - S_{\text{dpv}}) S_w c) + \frac{\partial}{\partial t} (\rho_{r,\text{ref}} (1 - \phi_{\text{ref}}) c^a) + \nabla \cdot (c \rho_w \vec{v}_{wp}) = 0. \quad (12)$$

where  $S_{\text{dpv}}$  denotes the fraction of the pore volume which is not accessible to polymer. The introduction of dead pore volume has the effect to increase the mobility of the polymer solution. However, the model equation (12) yields to instabilities because it allows polymer to go faster than its solvent (water). In a forthcoming article (!), it is shown that a consistent way to introduce dead pore volume is to replace the definition of the effective water viscosity given by (5) by

$$\frac{1}{\mu_{w,\text{eff}}} = \frac{1 - c/c_{\text{max}}}{\mu_{w,e}} + \frac{c/c_{\text{max}}}{(1 - S_{\text{dpv}}) \mu_{p,\text{eff}}}. \quad (13)$$

In this case, the polymer mobility factor becomes

$$m(c) = \frac{\mu_{w,\text{eff}}}{\mu_{p,\text{eff}}} = \left[ \left( 1 - \frac{c}{c_{\text{max}}} \right) \left( \frac{\mu_p}{\mu_w} \right)^{1-\omega} (1 - S_{\text{dpv}}) + \frac{c}{c_{\text{max}}} \right]^{-1}. \quad (14)$$

Finally the modeling equations are

$$\frac{\partial}{\partial t} (\rho_\alpha \phi S_\alpha) + \nabla \cdot (\rho_\alpha \vec{v}_\alpha) = 0, \quad (15a)$$

for  $\alpha \in \{w, o\}$ ,

$$\frac{\partial}{\partial t} (\rho_w \phi S_w c) + \frac{\partial}{\partial t} (\rho_{r,\text{ref}} (1 - \phi_{\text{ref}}) c^a) + \nabla \cdot (c \rho_w \vec{v}_{wp}) = 0. \quad (15b)$$

where  $\vec{v}_\alpha$  and  $\vec{v}_{wp}$  are defined in (6) and (7) using (3), (4) and (13).

### Discretization and Splitting of the equations

Simple PVT behaviour is modeled through the formation-volume factors  $b_\alpha = b_\alpha(p)$ , defined by  $\rho_\alpha = b_\alpha \rho_\alpha^S$ , where  $\rho_\alpha^S$  is the surface density of the phase  $\alpha \in \{w, o\}$ . Inserting this into (15), the system can be simplified by dividing each equation with the appropriate surface density  $\rho_\alpha^S$ ,

$$\frac{\partial}{\partial t} (b_\alpha \phi S_\alpha) + \nabla \cdot (b_\alpha \vec{v}_\alpha) = 0, \quad (16a)$$

$$\frac{\partial}{\partial t} (b_w \phi S_w c) + \frac{\partial}{\partial t} ((1 - \phi_{\text{ref}}) \hat{c}^a) + \nabla \cdot (b_w c \vec{v}_{wp}) = 0, \quad (16b)$$

where, for convenience, we have introduced the short-hand  $\hat{c}^a = c^a \rho_{r,\text{ref}} / \rho_w^S$ . To discretize (16), we introduce a grid consisting of cells  $\{C_i\}$  with a bulk volume  $V_i$ , integrate over each cell in space, and apply a standard implicit method for the temporal derivative. This gives the discrete transport equations

$$R_{\alpha,i} = (b_{\alpha,i} \phi_i S_{\alpha,i})^{n+1} - (b_{\alpha,i} \phi_i S_{\alpha,i})^n + \frac{\Delta t}{V_i} \sum_j (b_{\alpha,ij} v_{\alpha,ij})^{n+1} = 0, \quad (17a)$$

for  $\alpha \in \{w, o\}$  and

$$R_{c,i} = \left( b_{w,i} \phi_i S_{w,i} c_i + (1 - \phi_{\text{ref},i}) \hat{c}_i^a \right)^{n+1} - \left( b_{w,i} \phi_i S_{w,i} c_i + (1 - \phi_{\text{ref},i}) \hat{c}_i^a \right)^n + \frac{\Delta t}{V_i} \sum_j (b_{w,ij} c_{ij} v_{wp,ij})^{n+1} = 0. \quad (17b)$$

Here, subscripts  $i$  denote quantities associated with cell  $C_i$  and subscripts  $ij$  denote quantities associated with the interface between cells  $C_i$  and  $C_j$ . Superscripts denote time steps. To derive a discrete pressure equation, we sum the two phase equations, (17), using (9) and the condition  $S_w + S_o = 1$  to obtain the pressure residual equation

$$R_{p,i} = \phi_i^{n+1} - \phi_i^n \sum_{\alpha \in \{w,o\}} \left( \frac{b_{\alpha,i}^n}{b_{\alpha,i}^{n+1}} S_{\alpha,i}^n \right) + \frac{\Delta t}{V_i} \sum_j \sum_{\alpha \in \{w,o\}} \frac{b_{\alpha,ij}^{n+1}}{b_{\alpha,i}^{n+1}} (f_{\alpha,ij} v_{ij} + g_{\alpha,ij})^{n+1} = 0. \quad (18)$$

Here,  $v_{ij}$  is a discretisation of the total flux. We use a two point flux approximation and we obtain a relation of the form

$$v_{ij} = -T_{ij}(p_j - p_i) + g_{ij}. \quad (19)$$

The transmissibility  $T_{ij}$  depends on saturation and concentration and  $g_{ij}$  is a discretization of the gravity term  $\int_{C_i \cap C_j} g(\lambda_w \rho_w + \lambda_o \rho_o) \mathbf{K} \nabla z \cdot \mathbf{n} dA$ . The exact form of  $g_{ij}$  does not matter on the results which are presented here. The terms  $g_{w,ij}$  and  $g_{o,ij}$  correspond to discretizations of  $\int_{C_i \cap C_j} \vec{v}_g \cdot \mathbf{n} dA$ . Again, the exact form of  $g_{\alpha,ij}$  does not matter here for the pressure equation (for the transport equation, we will handle this term with care, see Section ) but we require that  $g_{w,ij} = -g_{o,ij}$ . Our overall system will consist of a pressure equation, (18), and two transport equations, (17a) with  $\alpha = w$  for the water saturation and (17b) for the polymer concentration. To solve this coupled system, we use a standard sequential solution procedure that separates and solves the pressure and transport equations in consecutive steps. We also split the transport equations (18) into a Darcy and gravitation component. The Darcy step consists of solving

$$(b_{w,i} \phi_i S_{w,i}^*)^{n+1} - (b_{w,i} \phi_i S_{w,i})^n + \frac{\Delta t}{V_i} \sum_j (b_{w,ij} f_w v_{ij})^{n+1} = 0, \quad (20)$$

and the corresponding residual equation for polymer (see (24)) to obtain the intermediate saturation and concentration values  $S_i^{*,n+1}$  and  $c_i^{*,n+1}$  which are used in the segregation step given by

$$(b_{w,i} \phi_i (S_i - S_i^*))^{n+1} + \frac{\Delta t}{V_i} \sum_j (b_{w,ij} g_{w,ij})_{ij}^{n+1} = 0, \quad (21)$$

and the corresponding residual equation for polymer (see (40b)) to update  $S^{n+1}$  and  $c^{n+1}$ . In the two following sections, we show that under certain conditions this sequential splitting is unconditionally stable in the sense that solutions to (20) and (21) exist and are unique without any restriction on the time step  $\Delta t$ .

### Unconditional Stability: Darcy component.

We use an upstream evaluation of the fractional flow  $f_\alpha$  and the pressure equation rewrites

$$R_{p,i} = \phi_i^{n+1} - \phi_i^n \sum_{\alpha \in \{w,o\}} \left( \frac{b_{\alpha,i}^n}{b_{\alpha,i}^{n+1}} S_{\alpha,i}^n \right) + \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} \sum_{\alpha \in \{w,o\}} \left( f_\alpha(S_j^n, c_j^n) \frac{b_{\alpha,i,j}^{n+1}}{b_{\alpha,i}^{n+1}} v_{ij}^{n+1} \right) \quad (22)$$

$$+ \frac{\Delta t}{V_i} \sum_{\alpha \in \{w,o\}} \left( f_\alpha(S_i^{n+1}, c_i^{n+1}) \sum_{\{j|v_{i,j}^{n+1} > 0\}} (b_{ij} v_{ij})^{n+1} \right) + \frac{\Delta t}{V_i} \sum_j g_{o,ij} \left( \frac{b_{o,ij}^{n+1}}{b_{o,i}^{n+1}} - \frac{b_{w,ij}^{n+1}}{b_{w,i}^{n+1}} \right) = 0,$$

where  $v_{ij}$  is given by (19) for values of saturation and concentration taken at the step  $n$ . The value of the surface volume factors have to be evaluated at the interface. To do so, we consider an approximation of the interface pressure,  $p_{ij}$ , for example given by the average  $p_{ij} = \frac{1}{2}(p_i + p_j)$ , then it is natural to set  $b_{\alpha,i,j}^{n+1} = b_\alpha(p_{ij}^{n+1})$  but, in order to obtain uniform stability of the Darcy component, we will see we need a different approximation, as detailed below. Once the coefficient  $b_{ij}$  are defined, we can compute the solution  $p^{n+1}$  and  $v_{ij}^{n+1}$  of the pressure equation (22) and proceed with the transport step. The Darcy component for the water residual equation is

$$R_{w,i}(S^{n+1}, c^{n+1}) = (b_i \phi_i S_i)^{n+1} - (b_i \phi_i S_i)^n + \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} (f_w(S_j, c_j) b_{ij} v_{ij})^{n+1} \quad (23)$$

$$+ \frac{\Delta t}{V_i} f_w(S_i, c_i)^{n+1} \sum_{\{j|v_{i,j}^{n+1} > 0\}} (b_{ij} v_{ij})^{n+1} = 0,$$

while, for the polymer residual equation, it is given by

$$R_{c,i}(S^{n+1}, c^{n+1}) = [b_i \phi_i S_i c_i + \hat{c}^a(c_i)(1 - \phi_{\text{ref},i})]^{n+1} - [b_i \phi_i S_i c_i + \hat{c}^a(c_i)(1 - \phi_{\text{ref},i})]^n \quad (24)$$

$$+ \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} (m(c_j) c_j f_w(S_j, c_j) b_{ij} v_{ij})^{n+1} + (m(c_i) c_i f_w(S_i, c_i))^{n+1} \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} > 0\}} (b_{ij} v_{ij})^{n+1} = 0.$$

Note that the polymer concentrations are also evaluated upstream. The single-cell problem for cell  $C_i$  consists of solving

$$R_{w,i}(S, c) = 0 \quad \text{and} \quad R_{c,i}(S, c) = 0 \quad (25)$$

where, slightly abusing notation, we denote by  $(S, c)$  the unknown  $(S_i, c_i)$ . The other values  $S_j$ , for  $j \neq i$  are assumed to be known. The pressure  $p^{n+1}$  is the solution of (22). In the following, we will show that the solution to (25) exists and is unique for all time steps  $\Delta t$ . First, we start by proving that, for any  $c \in [0, c_{\max}]$ , there exists a unique  $S$ , which we will denote  $S(c)$ , such that  $R_{w,i}(S(c), c) = 0$ . Given  $c$ , let us compute the values of the water residual at the endpoints, that is, for  $S = 0$  and  $S = 1$ . For  $S = 0$ , we have

$$R_{w,i}(0, c) = -(b_{w,i} \phi_i S_i)^n + \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} (f_w(S_j, c_j) b_{w,ij} v_{i,j})^{n+1} \leq 0. \quad (26)$$

For  $S = 1$ , after using (22), we have

$$\frac{1}{b_{w,i}} R_{w,i}(1, c) = \frac{1}{b_{w,i}} R_{w,i}(1, c) - R_{p,i} = A + B + C + D,$$

where

$$A = (\phi_i - \frac{b_{w,i}^n}{b_{w,i}} \phi_i^n S_{w,i}^n) - (\phi_i S_{w,i} - \frac{b_{w,i}^n}{b_{w,i}} \phi_i^n S_{w,i}^n) - (\phi_i S_{o,i} - \frac{b_{o,i}^n}{b_{o,i}} \phi_i^n S_{o,i}^n),$$

$$B = \frac{\Delta t}{V_i} \sum_{\{v_{i,j} < 0\}} v_{i,j} \left( f_w(S_j, c_j) \frac{b_{w,ij}}{b_{w,i}} - f_w(S_j^n, c_j^n) \frac{b_{w,ij}}{b_{w,i}} - f_o(S_j^n, c_j^n) \frac{b_{o,ij}}{b_{o,i}} \right),$$

$$C = \frac{\Delta t}{V_i} \sum_{\{v_{i,j} > 0\}} v_{i,j} \left( f_w(1, c) \frac{b_{w,ij}}{b_{w,i}} - f_w(S_i^n, c_i^n) \frac{b_{w,ij}}{b_{w,i}} - f_o(S_i^n, c_i^n) \frac{b_{o,ij}}{b_{o,i}} \right).$$

and

$$D = \frac{\Delta t}{V_i} \sum_j g_{o,ij} \left( \frac{b_{w,ij}}{b_{w,i}} - \frac{b_{o,ij}}{b_{o,i}} \right).$$

In the expressions above, the superscript is omitted when it corresponds to  $n+1$ . We will follow this convention in the rest of the article. These expressions simplify as follows. We have

$$A = \frac{b_{o,i}^n}{b_{o,i}} \phi_i^n S_{o,i}^n,$$

so that  $A \geq 0$  and

$$B = \frac{\Delta t}{V_i} \sum_{\{v_{i,j} < 0\}} v_{i,j} \left( f_o(S_j^n, c_j^n) \left( \frac{b_{w,ij}}{b_{w,i}} - \frac{b_{o,ij}}{b_{o,i}} \right) - \frac{b_{w,ij}}{b_{w,i}} (1 - f_w(S_j, c_j)) \right),$$

$$C = \frac{\Delta t}{V_i} \sum_{\{v_{i,j} > 0\}} v_{i,j} f_o(S_i^n, c_i^n) \left( \frac{b_{w,ij}}{b_{w,i}} - \frac{b_{o,ij}}{b_{o,i}} \right)$$

In the case where oil and water compressibilities are equal, that is,  $b_o = b_w$ , we have  $C = D = 0$  and  $B \geq 0$ . In the general case, a sufficient condition to obtain that  $R(1, c) \geq 0$  is that the quantity  $E$ , defined as

$$E = \sum_{\{v_{i,j} > 0\}} (f_o(S_i^n, c_i^n) v_{i,j} + g_{o,ij}) \left( \frac{b_{w,ij}}{b_{w,i}} - \frac{b_{o,ij}}{b_{o,i}} \right) + \sum_{\{v_{i,j} < 0\}} (f_o(S_j^n, c_j^n) v_{i,j} + g_{o,ij}) \left( \frac{b_{w,ij}}{b_{w,i}} - \frac{b_{o,ij}}{b_{o,i}} \right)$$

is positive. We assume that the water compressibility is smaller than the oil compressibility in the pressure range we are interested in, that is,

$$c_w(p) \leq c_o(p) \text{ for all } p \in [p_{\min}, p_{\max}], \quad (27)$$

then we can show that, for any  $p_1, p_2 \in [p_{\min}, p_{\max}]$ , if  $p_1 \leq p_2$ , then

$$\frac{b_{o,ij}}{b_{o,i}} - \frac{b_{w,ij}}{b_{w,i}} = \frac{b_o(p_2)}{b_o(p_1)} - \frac{b_w(p_2)}{b_w(p_1)} \geq 0. \quad (28)$$

By definition, we have  $\frac{1}{b_\alpha} \frac{db_\alpha}{dp} = c_\alpha(p)$ . Hence,  $d(\ln(b_o)) = c_o(p) dp$ . After integrating and using that  $p_i < p_j$ , we obtain

$$\ln \left( \frac{b_w(p_2)}{b_w(p_1)} \right) = \int_{p_1}^{p_2} c_w(p) dp \leq \int_{p_1}^{p_2} c_o(p) dp = \ln \left( \frac{b_o(p_2)}{b_o(p_1)} \right)$$

so that (28) holds. Now, we define the surface volume factor  $b_{ij}$  at the interface in a way which guarantees that  $E$  remains positive in all cases. As mentioned earlier, an optimal choice in term of accuracy would be to take  $b_{\alpha,ij} = b_\alpha(p_{ij}^{n+1})$ , where  $p_{ij}^{n+1}$  is an approximation of the pressure at the interface. But this choice does not guarantee negative values for  $E$ . By defining  $b_{\alpha,ij}$  as

$$b_{\alpha,ij} = \begin{cases} b_\alpha(p_i) & \text{if } (f_o(S_i^n, c_i^n) v_{i,j} + g_{o,ij})(p_i - p_j) \leq 0, \\ b_\alpha(p_j) & \text{if } (f_o(S_i^n, c_i^n) v_{i,j} + g_{o,ij})(p_i - p_j) \geq 0, \end{cases} \quad (29)$$

when  $v_{i,j} \geq 0$ , the identity (28) implies that the coefficient  $E$  always remains positive. Note that  $f_o v_{i,j} + g_{o,ij}$  is an approximation of the oil flux  $\vec{v} \cdot \mathbf{n}$  so that the condition (29) can be rephrased as follows: If the flux of the most compressible phase is in the same direction as the inverse pressure gradient, then we evaluate the densities by taking the pressure value downwind; otherwise, we use the upwind value. To prove that  $R_w(S, c)$  admits a unique solution in  $S$  for a given  $c$ , it remains to prove that the function  $S \mapsto R_w(S, c)$  is strictly increasing. We simplify the notations and rewrite the residuals in the cell  $C_i$  as

$$R_w(S, c) = \sigma_1 S + \sigma_2 f_w(S, c) + \sigma_3 \quad (30a)$$

$$R_c(S, c) = \sigma_1 S c + \sigma_4 \hat{c}^a(c) + \sigma_2 m(c) c f_w(S, c) \quad (30b)$$

where  $\{\sigma_i\}_{i=1}^4$  are constants whose definition can be inferred from the definition of the residuals. These constants depend only on the values of  $S_j$  and  $c_j$  of previous time steps or other cells than  $C_i$ , they are positive and  $\sigma_1 > 0$ . We have

$$\frac{\partial R_w}{\partial S} = \sigma_1 + \sigma_2 \frac{\partial f_w}{\partial S}$$

As expected, the fractional flow is an increasing function of saturation. Indeed, we have

$$\frac{\partial f_w}{\partial S} = \left( \frac{\partial \lambda_w}{\partial S} \lambda_o - \lambda_w \frac{\partial \lambda_o}{\partial S} \right) (\lambda_w + \lambda_o)^{-2} \geq 0,$$

because  $\frac{\partial \lambda_w}{\partial S} \geq 0$ ,  $\frac{\partial \lambda_o}{\partial S} \leq 0$ . Since

$$R_w(0, c) \leq 0, \quad R_w(1, c) \geq 0 \quad \text{and} \quad \frac{\partial R_w}{\partial S} > 0,$$

there exists a unique solution  $S(c)$  to  $R_w(S(c), c) = 0$  for any given  $c \in [0, c_{\max}]$ . Let us now prove that  $R_c(S(c), c)$  admits a unique solution in  $c \in [0, c_{\max}]$ . As earlier, we start by checking the endpoints  $c = 0$  and  $c = c_{\max}$ . We have

$$R_c(S(0), 0) = -\left(b_i \phi_i S_i c_i + \hat{c}^a(c_i)(1 - \phi_{\text{ref},i})\right)^n + \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} \left(m(c_j) c_j f(S_j, c_j) b_{ij} v_{ij}\right)^{n+1}$$

so that  $R_c(S(0), 0) \leq 0$ . For  $c = c_{\max}$ , we have

$$R_c(S(c_{\max}), c_{\max}) = R_c(S(c_{\max}), c_{\max}) - c_{\max} R_w(S(c_{\max}), c_{\max}) = A + B + C,$$

where

$$A = (\hat{c}^a(c_{\max}) - \hat{c}^a(c_i))(1 - \phi_{\text{ref},i}) + (c_{\max} - c_i) b_{w,i}^n \phi_i^n S_i^n,$$

$$B = \frac{\Delta t}{V_i} \sum_{\{j|v_{i,j}^{n+1} < 0\}} (m(c_j^{n+1}) c_j^{n+1} - c_{\max}) f_w(S_j^{n+1}, c_j^{n+1}) b_{ij}^{n+1} v_{ij}^{n+1}$$

and

$$C = \frac{\Delta t}{V_i} (m(c_{\max}) c_{\max} - c_{\max}) f_w(S(c_{\max}), c_{\max}) \sum_{\{j|v_{i,j}^{n+1} > 0\}} b_{ij}^{n+1} v_{ij}^{n+1}.$$

Since  $m(c_{\max}) = 1$ , we have  $C = 0$ . To prove that  $B \geq 0$ , we are going to establish that  $m(c)c$  is non-decreasing so that

$$\sup_{c \in [0, c_{\max}]} m(c)c = m(c_{\max})c_{\max} = c_{\max}. \quad (31)$$

Let us denote

$$\kappa = \left( \frac{\mu_p}{\mu_w} \right)^{1-\omega} (1 - S_{\text{dpv}}) > 0,$$

and we rewrite (14) as

$$\frac{1}{m(c)} = \kappa + (1 - \kappa) \frac{c}{c_{\max}}. \quad (32)$$



After some computation, we get that

$$\frac{d}{dc}(m(c)c) = m(c)\left[1 - \frac{m(c)c}{c_{\max}}(1 - \kappa)\right],$$

which can be rewritten, using (32), as

$$\frac{d}{dc}(m(c)c) = \kappa m^2. \quad (33)$$

Hence,  $m(c)c$  is non-decreasing. From (31) it follows that  $B \geq 0$  and  $R_w(S(c_{\max}), c_{\max}) \geq 0$ . Let us now prove that the function  $c \mapsto R_c(S(c), c)$  is a non-decreasing function. We have

$$dR_c = \sigma_1(c dS + S dc) + \sigma_4 d\hat{c}^a + \sigma_2 f d(m(c)c) + \sigma_2 m(c)c df \quad (34)$$

Since  $S(c)$  is solution of  $R_w(S(c), c) = 0$ , we have

$$\sigma_1 dS + \sigma_2 df = 0, \quad (35)$$

from (30a). Plugging this result into (34), we get

$$dR_c = \sigma_1 c(1 - m(c)) dS + \sigma_1 S dc + \sigma_4 d\hat{c}^a + \sigma_2 f d(m(c)c). \quad (36)$$

From (35), we obtain that

$$(\sigma_1 + \sigma_2 \frac{\partial f}{\partial S}) dS = -\sigma_2 \frac{\partial f}{\partial c} dc,$$

which yields, by (36),

$$\begin{aligned} (\sigma_1 + \sigma_2 \frac{\partial f}{\partial S}) \frac{dR_c}{dc} &= \sigma_1^2 S + \sigma_2^2 \frac{\partial f}{\partial S} \kappa f m^2 + \sigma_4 (\sigma_1 + \sigma_2 \frac{\partial f}{\partial S}) \frac{d\hat{c}^a}{dc} \\ &\quad + \sigma_1 \sigma_2 (S \frac{\partial f}{\partial S} + \kappa f m^2 - c(1 - m) \frac{\partial f}{\partial c}). \end{aligned} \quad (37)$$

Since  $\frac{\partial f}{\partial S} \geq 0$  and, as we will show,  $\frac{\partial f}{\partial c} \leq 0$ , we can conclude that  $\frac{dR_c}{dc} \geq 0$  if the condition that

$$S \frac{\partial f}{\partial S} + \kappa f m^2 - c(1 - m) \frac{\partial f}{\partial c} \geq 0 \quad (38)$$

is fulfilled. In the appendix we explain why condition (38) is necessary if we want the system of equations given by (16a) (for  $\alpha = w$ ) and (16b) to produce waves which both travel in the same direction of the total flux. Otherwise, we can still assume that  $\kappa > 1$ , which directly implies that the polymer mobility factor  $m(c)$  is a non-decreasing function of the concentration  $c$ . This assumption is physically reasonable and easy to check. In this case,  $m(c) \leq m(c_{\max}) = 1$ . Let us compute  $\frac{\partial f}{\partial c}$ . Since,  $\lambda_w = k_{rw}/\mu_{w,\text{eff}} R_k$ , we have

$$\frac{\partial f}{\partial c} = \frac{\partial \lambda_w}{\partial c} \frac{\lambda_o}{(\lambda_w + \lambda_o)^2} = -\frac{\partial}{\partial c} (\mu_{w,\text{eff}} R_k) \frac{k_{rw}}{\mu_{w,\text{eff}}^2 R_k^2} \frac{\lambda_o}{(\lambda_w + \lambda_o)^2} \leq 0. \quad (39)$$

Indeed, by assumption,  $\frac{\partial R_k}{\partial c} \geq 0$  and we expect that  $\frac{\partial \mu_{w,\text{eff}}}{\partial c} \geq 0$  because the effect of polymer is to increase the effective viscosity of water. Let us check that directly. As  $\mu_{w,\text{eff}} = m(c)\mu_{p,\text{eff}}$  and  $\mu_{p,\text{eff}} = \mu_m(c)^\omega \mu_p^{1-\omega}$ , we have

$$\frac{d}{dc} \ln(\mu_{w,\text{eff}}) = \frac{d}{dc} \ln(m) + \omega \frac{d}{dc} \ln(\mu_m).$$

Since  $m$  and, by assumption,  $\mu_m$  are non-decreasing functions, we can conclude that  $\mu_{w,\text{eff}}$  is non-decreasing. Since

$$R_c(S(0), 0) \leq 0 \text{ and } R_c(S(c_{\max}), c_{\max}) \geq 0,$$

there exists at least one solution  $c$  to  $R_c(S(c), c) = 0$  in  $[0, c_{\max}]$ . If, for this solution,  $S(c) > 0$ , then  $\frac{dR_c}{dc} > 0$  because  $\sigma_1 > 0$  and the solution is unique. Otherwise, if there exists  $c_0$  such that  $S(c_0) = 0$  and  $R_c(S(c_0), c_0) = 0$ , then we have, by (26), that  $S_i^n = 0$  and  $f_w(S_j, c_j) = 0$  for all  $j$  such that  $v_{i,j} < 0$ . The polymer residual becomes

$$R_c(S, c) = \hat{c}^a(c) - \hat{c}^a(c_i^n).$$

In the absence of adsorption, it yields  $R_c(S, c) = 0$  for all  $c \in [0, c_{\max}]$ . Thus, the solution is not unique. This property simply reflects the fact that concentration is not a well defined quantity in the absence of water. Note that adsorption will have a stabilizing effect. Indeed, assuming that  $\hat{c}^a$  is a strictly increasing function, we recover uniqueness.

### Unconditional Stability: Segregation component.

To take into account the gravity segregation effects, we introduce an additional operator splitting for the transport equations as described at the end of Section . This operator splitting method was first introduced within streamline simulation (Gmelig Meyling, 1990, 1991; Bratvedt et al., 1996), but can also offer certain benefits for finite-volume methods, e.g., as discussed in (Lie et al., 2012a). The segregation residual equations for water and polymer are given by

$$(b_w \phi(S - S^*))_i^{n+1} + \frac{\Delta t}{V_i} \sum_j (b_w \lambda_o f(S, c) (\rho_w - \rho_o) g K \nabla z)_{ij}^{n+1} = 0, \quad (40a)$$

and

$$\begin{aligned} & \left( b_w \phi c(S - S^*) + (1 - \phi_{\text{ref}})(c^a(c) - c^a(c^*)) \right)_i^{n+1} \\ & + \frac{\Delta t}{V_i} \sum_j (b_w m(c) c \lambda_o f(S, c) (\rho_w - \rho_o) g K \nabla z)_{ij}^{n+1} = 0. \end{aligned} \quad (40b)$$

We use a two point flux approximation to discretize the term  $\int_{C_i \cap C_j} b_w (\rho_w - \rho_o) g K \nabla z \cdot \mathbf{n} dA$ . For a face  $C_i \cap C_j$ , the flux of the gradient of a function  $\psi$  of the form

$$\int_{C_i \cap C_j} h K \nabla \psi \cdot \mathbf{n}_{i,j} dA,$$

for some given function  $h$ , is approximated by

$$\int_{C_i \cap C_j} h K \nabla \psi \cdot \mathbf{n}_{i,j} dA, \approx T_{i,j} (\psi_j - \psi_i)$$

where

$$T_{i,j} = \left( \frac{h_i}{t_{i,j}} + \frac{h_j}{t_{j,i}} \right)^{-1}$$

and  $t_{i,j}$  denotes the one sided transmissibility coefficient of the cell  $C_i$  with respect to the face  $C_i \cap C_j$ , defined as

$$t_{i,j} = \frac{1}{|\mathbf{c}_{i,j}|^2} \mathbf{n}_{ij} \cdot \mathbf{K} \mathbf{c}_{i,j},$$

where  $\mathbf{c}_{i,j}$  denotes the vector joining the cell centroid of  $C_i$  to the face centroid of  $C_i \cap C_j$ , see (Lie et al., 2012b) for more details. For the gravitation segregation equation, we take

$$\psi = z \quad \text{and} \quad h = b_w - b_o \frac{\rho_o^s}{\rho_w^s}.$$

In the case of a corner-point grid consisting of strictly vertical pillars, the gravitation flux vanishes on all vertical faces of a cell. Let us consider a column which consists of vertically aligned cells  $C_i$

( $i = 1, \dots, N$ ) and denote by  $z_i$  the vertical coordinate of the centroid of  $C_i$ . The discretization of (40) in a column yields, after denoting  $\mathbf{u} = (S, c)$ , the following residual equations

$$b_{w,i}\phi_i(S_i - S_i^*) + \frac{g\Delta t}{V_i} \left( F(\mathbf{u}_i, \mathbf{u}_{i+1})T_{i,i+1}(z_{i+1} - z_i) - F(\mathbf{u}_{i-1}, \mathbf{u}_i)T_{i-1,i}(z_i - z_{i-1}) \right) = 0$$

and

$$b_{w,i}\phi_i(S_i c_i - S_i^* c_i^*) + (1 - \phi_{\text{ref}})(c^a(c_i) - c^a(c_i^*)) + \frac{g\Delta t}{V_i} \left( G(\mathbf{u}_i, \mathbf{u}_{i+1})T_{i,i+1}(z_{i+1} - z_i) - G(\mathbf{u}_{i-1}, \mathbf{u}_i)T_{i-1,i}(z_i - z_{i-1}) \right) = 0$$

where  $F(\mathbf{u}_u, \mathbf{u}_l)$  and  $G(\mathbf{u}_u, \mathbf{u}_l)$  denote approximations of the flux (For a given face,  $\mathbf{u}_u$  and  $\mathbf{u}_l$  are the values  $\mathbf{u}$  of the cell above and below the face). As earlier, we drop the superscript  $n+1$  in the notation. For the numerical flux  $F$ , we use a phase upwind mobility approximation,

$$F(\mathbf{u}_u, \mathbf{u}_l) = \frac{\lambda_w(S_u, c_u)\lambda_o(1 - S_l)}{\lambda_w(S_u, c_u) + \lambda_o(1 - S_l)}. \quad (41)$$

Note that the value of the polymer concentration in the mobility term  $\lambda_w$  is taken from the cell above, that is, we use upwinding from the water phase. For the numerical flux  $G$ , it seems at first natural to take the value of the polymer concentration from the cell above, because polymer is transported by the water phase to which it belongs. Then, we obtain the following expression for the numerical flux

$$G(\mathbf{u}_u, \mathbf{u}_l) = m(c_u)c_u \frac{\lambda_w(S_u, c_u)\lambda_o(1 - S_l)}{\lambda_w(S_u, c_u) + \lambda_o(1 - S_l)}. \quad (42)$$

But this choice does not lead to well-posedness and we have to consider instead

$$G(\mathbf{u}_u, \mathbf{u}_l) = m(c_u)c_u \frac{\lambda_w(S_u, c_l)\lambda_o(1 - S_l)}{\lambda_w(S_u, c_l) + \lambda_o(1 - S_l)}. \quad (43)$$

where the value of the polymer concentration in the mobility term  $\lambda_w$  is taken from the cell below, which corresponds to upwinding from the oil phase. In the rest of this section, we prove that the single cell problem for the segregation problem is unconditionally stable. In the case of a column, the spatial dimension is reduced to one and we start this section by introducing the general setting in one dimension and investigate what are the requirements on the numerical flux function to obtain an unconditionally stable single cell problem. Let us consider the scalar conservation law

$$u_t + f(u)_z = 0,$$

which we discretised using an implicit Euler scheme, that is,

$$u_i^{n+1} - u_i^n + \frac{\Delta t}{\Delta z} (F(u_i^{n+1}, u_{i+1}^{n+1}) - F(u_{i-1}^{n+1}, u_i^{n+1})) = 0,$$

where the function  $F(u_u, u_l)$  is a discrete approximation of the flux between two cells. For compatibility reason, we require

$$F(u, u) = f(u).$$

The single cell problem consists of finding the solution of  $R(u) = 0$ , where

$$R(u) = u - u_i^n + \frac{\Delta t}{\Delta z} (F(u, u_{i+1}) - F(u_{i-1}, u))$$

and  $u_i^n$ ,  $u_{i-1}$ ,  $u_{i+1}$  are known. We want to find the conditions for which this scalar equation admits a unique solution for any given  $u_i^n$ ,  $u_{i-1}$ ,  $u_{i+1}$  and  $\Delta t$ . Since  $u_i^n$  is arbitrary, we must have that  $R$  is

monotone. By taking  $\Delta t$  small, we obtain that, if  $R$  is monotone, it can only be increasing. By taking  $\Delta t$  very large, we see that we must have  $\frac{d}{du}(F(u, u_{i+1}) - F(u_{i-1}, u))$  positive. Since this must hold for any  $u_{i+1}$  and  $u_{i-1}$ , we end up with the following monotonicity conditions for  $F$ ,

$$\frac{\partial F}{\partial u_u} \geq 0 \quad \text{and} \quad \frac{\partial F}{\partial u_l} \leq 0. \quad (44)$$

The Engquist-Osher flux (add reference)

$$F(u_u, u_l) = \int_0^{u_u} \max(0, f'(u)) du + \int_0^{u_l} \min(0, f'(u)) du + f(0)$$

satisfies this condition. In the case of gravity segregation, a phase upwind mobility numerical flux is commonly used and we are going to check that it also satisfies the monotonicity condition (44). For the case without polymer, the discretization of the segregation equation is given by

$$R(S) = b_i^{n+1} \phi^{n+1} (S_i^{n+1} - S_i^n) + \frac{\Delta t}{V_i} (F(S_i^{n+1}, S_{i+1}^{n+1}) g T_{i,i+1}(z_i - z_{i-1}) - F(S_{i-1}^{n+1}, S_i^{n+1}) g T_{i-1,i}(z_i - z_{i-1})) = 0,$$

where  $F(S_u, S_l)$  is a discrete numerical flux which has to satisfy the compatibility condition

$$F(S, S) = \frac{\lambda_w(S) \lambda_o(1 - S)}{\lambda_w(S) + \lambda_o(1 - S)}.$$

In a segregation process where oil is lighter than water, the oil phase travels upwards and the water phase downwards. Thus, an upstream mobility weighting for the flux is given by

$$F(S_u, S_l) = \frac{\lambda_w(S_u) \lambda_o(1 - S_l)}{\lambda_w(S_u) + \lambda_o(1 - S_l)}.$$

Since  $\lambda_w$  and  $\lambda_o$  are non decreasing functions, we can check that this discrete flux satisfies the condition (44) which is necessary for the solution of  $R(u) = 0$  to exist for any  $\Delta t$ .

Let us now turn our attention to the segregation case in the presence of polymer. In this case we have to solve a system of two equations. The water and polymer gravity residuals,  $R_w^g(\mathbf{u})$  and  $R_c^g(\mathbf{u})$  in the cell  $C_i$  are given by

$$R_w^g(\mathbf{u}) = b_{w,i} \phi_i (S - S_i^*) + \frac{g \Delta t}{V_i} (F(\mathbf{u}, \mathbf{u}_{i+1}) T_{i,i+1}(z_{i+1} - z_i) - F(\mathbf{u}_{i-1}, \mathbf{u}) T_{i-1,i}(z_i - z_{i-1})) \quad (45)$$

and

$$R_c^g(\mathbf{u}) = b_{w,i} \phi_i (S c - S_i^* c_i^*) + (1 - \phi_{\text{ref}}) (c^a(c) - c^a(c_i^*)) + \frac{g \Delta t}{V_i} (G(\mathbf{u}, \mathbf{u}_{i+1}) T_{i,i+1}(z_{i+1} - z_i) - G(\mathbf{u}_{i-1}, \mathbf{u}) T_{i-1,i}(z_i - z_{i-1})). \quad (46)$$

The discrete flux  $F$  and  $G$  have to satisfy the compatibility conditions

$$F(\mathbf{u}, \mathbf{u}) = \frac{\lambda_w(S, c) \lambda_o(1 - S)}{\lambda_w(S, c) + \lambda_o(1 - S)} \quad (47a)$$

and

$$G(\mathbf{u}, \mathbf{u}) = m(c) c \frac{\lambda_w(S, c) \lambda_o(1 - S)}{\lambda_w(S, c) + \lambda_o(1 - S)}. \quad (47b)$$

We choose the numerical fluxes given by

$$F(\mathbf{u}_u, \mathbf{u}_l) = \frac{\lambda_w(S_u, c_u) \lambda_o(1 - S_l)}{\lambda_w(S_u, c_u) + \lambda_o(1 - S_l)}$$

and

$$G(\mathbf{u}_u, \mathbf{u}_l) = m(c_u)c_u \frac{\lambda_w(S_u, c_l)\lambda_o(1 - S_l)}{\lambda_w(S_u, c_l) + \lambda_o(1 - S_l)},$$

which satisfy (47) and enjoy the following monotonicity properties

$$\frac{\partial F}{\partial S_u} \geq 0, \quad \frac{\partial F}{\partial c_u} \leq 0, \quad \frac{\partial F}{\partial S_l} \leq 0, \quad \frac{\partial F}{\partial c_l} = 0 \geq 0 \quad (48a)$$

and

$$\frac{\partial G}{\partial S_u} \geq 0, \quad \frac{\partial G}{\partial c_u} \geq 0, \quad \frac{\partial G}{\partial S_l} \leq 0, \quad \frac{\partial G}{\partial c_l} \leq 0. \quad (48b)$$

The proofs of (48) follow from the chain rule and the fact that  $\frac{d(mc)}{dc} \geq 0$  (see (33)),  $\frac{\partial \lambda_w}{\partial c} \leq 0$  (see (39)) and  $\frac{\partial \lambda_w}{\partial S_w} \geq 0$ ,  $\frac{\partial \lambda_o}{\partial S_o} \geq 0$  (by assumption). Let us prove that for this choice of numerical flux, there exists a unique solution  $(S, c)$  of  $R_w^g(S, c) = 0$  and  $R_c^g(S, c) = 0$  for any  $\Delta t$ . To simplify the notation, we rewrite (45) and (46) as

$$R_w^g(\mathbf{u}) = \alpha(S - S_i^*) + \beta F(\mathbf{u}, \mathbf{u}_{i+1}) - \gamma F(\mathbf{u}_{i-1}, \mathbf{u}) \quad (49)$$

and

$$R_c^g(\mathbf{u}) = \alpha(Sc - S_i^*c_i^*) + \delta(c^a(c) - c^a(c_i^*)) + \beta G(\mathbf{u}, \mathbf{u}_{i+1}) - \gamma G(\mathbf{u}_{i-1}, \mathbf{u}). \quad (50)$$

Note that  $\alpha, \beta, \gamma, \delta$  are positive and  $\alpha > 0$ . We differentiate these expressions and obtain

$$dR_w^g = (\alpha + \beta \frac{\partial F}{\partial S_u} - \gamma \frac{\partial F}{\partial S_l})dS + (\beta \frac{\partial F}{\partial c_u} - \gamma \frac{\partial F}{\partial c_l})dc \quad (51a)$$

and

$$dR_c^g = (\alpha c + \beta \frac{\partial G}{\partial S_u} - \gamma \frac{\partial G}{\partial S_l})dS + (\alpha S + \delta \frac{c^a}{dc} + \beta \frac{\partial G}{\partial c_u} - \gamma \frac{\partial G}{\partial c_l})dc. \quad (51b)$$

We proceed as in the previous section. First, we prove that, for any given  $c \in [0, c_{\max}]$ , there exists a unique  $S \in [0, 1]$ , which we denote  $S(c)$ , such that  $R_w^g(S(c), c) = 0$ . For  $\mathbf{u} = [0, c]$ , we have  $F(\mathbf{u}, \mathbf{u}_{i+1}) = 0$  so that  $R_w^g(0, c) = -\alpha S_i^* - \gamma F(\mathbf{u}_{i-1}, \mathbf{u}) \leq 0$ . For  $\mathbf{u} = [1, c]$ , we have  $F(\mathbf{u}_{i-1}, \mathbf{u}) = 0$  and  $R_w^g(1, c) = \alpha(1 - S_i^*) + \beta F(\mathbf{u}, \mathbf{u}_{i+1}) \geq 0$ . Moreover, we have

$$\frac{\partial R_w^g}{\partial S} = \alpha + \beta \frac{\partial F}{\partial S_u} - \gamma \frac{\partial F}{\partial S_l} > 0$$

so that the function is  $S \mapsto R_w^g(S, c)$  is strictly increasing for a given  $c$ . Therefore there exists a unique solution  $S(c)$  in  $[0, 1]$  to  $R_w^g(S, c) = 0$ . Let us now prove that the equation  $R_c^g(S(c), c) = 0$  admits a unique solution. For  $c = 0$ , that is,  $\mathbf{u} = [S(0), 0]$ , we have  $G(\mathbf{u}, \mathbf{u}_{i+1}) = 0$  and  $R_c^g(S(0), 0) = -\alpha S_i^*c_i^* + \delta(c^a(0) - c^a(c_i^*)) - \gamma G(\mathbf{u}_{i-1}, \mathbf{u}) \leq 0$ . Since  $R_w^g(\mathbf{u}) = 0$ , for  $\mathbf{u}_{\max} = [c_{\max}, S(c_{\max})]$ , we obtain from (50) that

$$\begin{aligned} R_c^g(\mathbf{u}_{\max}) &= R_c^g(\mathbf{u}_{\max}) - c_{\max}R_w^g(\mathbf{u}_{\max}) \\ &= \alpha S_i^*(c_{\max} - c_i^*) + \delta(c^a(c_{\max}) - c^a(c_i^*)) + \beta(G(\mathbf{u}_{\max}, \mathbf{u}_{i+1}) - cF(\mathbf{u}_{\max}, \mathbf{u}_{i+1})) \\ &\quad - \gamma(G(\mathbf{u}_{i-1}, \mathbf{u}_{\max}) - cF(\mathbf{u}_{i-1}, \mathbf{u}_{\max})). \end{aligned}$$

We have

$$\begin{aligned} G(\mathbf{u}_{\max}, \mathbf{u}_{i+1}) - c_{\max}F(\mathbf{u}_{\max}, \mathbf{u}_{i+1}) &= c_{\max} \left( \frac{\lambda_w(S(c_{\max}), c_{i+1})\lambda_o(1 - S_{i+1})}{\lambda_w(S(c_{\max}), c_{i+1}) + \lambda_o(1 - S_{i+1})} \right. \\ &\quad \left. - \frac{\lambda_w(S(c_{\max}), c_{\max})\lambda_o(1 - S_{i+1})}{\lambda_w(S(c_{\max}), c_{\max}) + \lambda_o(1 - S_{i+1})} \right) \geq 0 \end{aligned}$$

because the function

$$c \mapsto \frac{\lambda_w(S(c_{\max}), c)\lambda_o(1 - S_{i+1})}{\lambda_w(S(c_{\max}), c) + \lambda_o(1 - S_{i+1})}$$

is non-increasing as  $\frac{\partial \lambda_w}{\partial c} \leq 0$ . Similarly,

$$\begin{aligned}
 & G(\mathbf{u}_{i-1}, \mathbf{u}_{\max}) - c_{\max} F(\mathbf{u}_{i-1}, \mathbf{u}_{\max}) \\
 &= m(c_{i-1})c_{i-1} \frac{\lambda_w(S_{i-1}, c_{\max})\lambda_o(1-S(c_{\max}))}{\lambda_w(S_{i-1}, c_{\max}) + \lambda_o(1-S(c_{\max}))} - c_{\max} \frac{\lambda_w(S_{i-1}, c_{i-1})\lambda_o(1-S(c_{\max}))}{\lambda_w(S_{i-1}, c_{i-1}) + \lambda_o(1-S(c_{\max}))} \\
 &= c_{\max} \left( \frac{\lambda_w(S_{i-1}, c_{\max})\lambda_o(1-S(c_{\max}))}{\lambda_w(S_{i-1}, c_{\max}) + \lambda_o(1-S(c_{\max}))} - \frac{\lambda_w(S_{i-1}, c_{i-1})\lambda_o(1-S(c_{\max}))}{\lambda_w(S_{i-1}, c_{i-1}) + \lambda_o(1-S(c_{\max}))} \right) \\
 &\quad + (m(c_{i-1})c_{i-1} - c_{\max}) \frac{\lambda_w(S_{i-1}, c_{\max})\lambda_o(1-S(c_{\max}))}{\lambda_w(S_{i-1}, c_{\max}) + \lambda_o(1-S(c_{\max}))} \\
 &\leq 0,
 \end{aligned}$$

because of (31). Hence,  $R_c(S(c_{\max}), c_{\max}) \geq 0$ . From (51a) and (48), we have that

$$\frac{dS}{dc} \geq 0$$

so that, by (51b) and again (48), we get

$$\frac{d}{dc} R_c(S(c), c) = \left( \alpha c + \beta \frac{\partial G}{\partial S_u} - \gamma \frac{\partial G}{\partial S_l} \right) \frac{dS}{dc} + \left( \alpha S + \delta \frac{dc^a}{dc} + \beta \frac{\partial G}{\partial c_u} - \gamma \frac{\partial G}{\partial c_l} \right) \geq 0.$$

Therefore the function  $R_c(S(c), c)$  is non-decreasing and there exists a solution  $c \in [0, c_{\max}]$ . The solution  $(S, c)$  is unique if  $S > 0$  because the function  $R_c(S(c), c)$  is then strictly increasing at  $c$ . In the case where the equation has a solution of  $(S, c)$  such that  $S = 0$ , the conclusion is the same as at the end of the previous section, that is, the solution is unique only if the adsorption function is strictly increasing.

## Appendix

In section , we observe that the condition (38) naturally shows up when we want to show the well-posedness of the single cell problem. Here, we show that this condition is necessary from a physical perspective. We consider the transport equations (16a) (for  $\alpha = w$ ) and (16b) in one space dimension. To simplify the computation, we assume that  $b = 1$  and the total flux  $v$  is constant and equal to one. The equations can then be rewritten as

$$\begin{aligned}
 \frac{\partial S}{\partial t} + \frac{\partial}{\partial x}(f(S, c)) &= 0, \\
 \frac{\partial Sc}{\partial t} + \frac{\partial}{\partial x}(m(c)cf(S, c)) &= 0.
 \end{aligned}$$

We introduce the variable  $z = Sc$  so that the system can be rewritten as a system of conservation laws, namely,

$$\begin{aligned}
 \frac{\partial S}{\partial t} + \frac{\partial}{\partial x}(\bar{f}(S, z)) &= 0, \\
 \frac{\partial z}{\partial t} + \frac{\partial}{\partial x}(\bar{m}(S, z)\bar{c}(S, z)\bar{f}(S, z)) &= 0,
 \end{aligned}$$

where

$$\bar{m}(S, z) = m(S, c), \quad \bar{c}(S, z) = c \quad \text{and} \quad \bar{f}(S, z) = f(S, c).$$

The properties of the system are related to the eigenvalues of the linearized flux given by

$$F = \begin{pmatrix} \frac{\partial \bar{f}}{\partial S} & \frac{\partial \bar{f}}{\partial z} \\ \bar{m}\bar{c}\frac{\partial \bar{f}}{\partial S} & \frac{\partial}{\partial z}(\bar{m}\bar{c}\bar{f}) \end{pmatrix} \quad (52)$$

We have an hyperbolic system if the eigenvalues of  $F$  are real. We do not investigate this condition here and assume it is fulfilled. Instead we consider the sign of the eigenvalues. Each eigenvalue  $\lambda_i, i = \{1, 2\}$ ,

corresponds to the speed of the wave of the corresponding Riemann problem. It is natural to require that the wave travels in the same direction as the flux, from left to right, and thus we want to impose  $\lambda_1 \geq 0$  and  $\lambda_2 \geq 0$ . These sign conditions are equivalent to

$$\det(F) \geq 0 \text{ and } \operatorname{tr}(F) \geq 0.$$

For a generic function  $\bar{g}(S, z) = g(S, c)$ , the change of variables formulas are

$$\frac{\partial \bar{g}}{\partial S} = \frac{\partial g}{\partial S} - \frac{c}{S} \frac{\partial g}{\partial c} \quad \text{and} \quad \frac{\partial \bar{g}}{\partial z} = \frac{1}{S} \frac{\partial g}{\partial c}.$$

Thus, we can compute  $\det(F)$  and we get

$$\begin{aligned} \det(F) &= \bar{f} \frac{\partial \bar{f}}{\partial S} \frac{\partial(\bar{m}\bar{c})}{\partial z} \\ &= \left( S \frac{\partial f}{\partial S} - c \frac{\partial f}{\partial c} \right) \frac{f}{S} \frac{\partial(mc)}{\partial c} \\ &\geq 0 \end{aligned}$$

because, as shown in Section ,  $\frac{\partial f}{\partial S} \geq 0$  and  $\frac{\partial f}{\partial c} \leq 0$ . For the trace of  $F$ , we have

$$\begin{aligned} \operatorname{tr}(F) &= \frac{\partial \bar{f}}{\partial S} + \frac{\partial(\bar{m}c\bar{f})}{\partial z} \\ &= \frac{\partial f}{\partial S} - \frac{c}{S} \frac{\partial f}{\partial c} + \frac{1}{S} \frac{\partial(mc f)}{\partial c} \\ &= \frac{1}{S} \left( S \frac{\partial f}{\partial S} + \kappa m^2 f + c(m-1) \frac{\partial f}{\partial c} \right). \end{aligned}$$

Hence, the condition (38) is fulfilled if and only if  $\operatorname{tr}(F) \geq 0$ , that is, if and only if the speeds of both waves are positive.

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