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A mathematical model for Biogrout

Bacterial placement and soil reinforcement

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Abstract We present a mathematical model for Biogrout, which is a technique for soil reinforcement that is based on microbially induced carbonate precipitation. The model deals with the entire process, consisting of fixation of bacteria, as well as of the subsequent soil reinforcement. The paper deals with the coupling of two earlier models for bacterial placement and reinforcement, where the construction of the model is discussed, as well as numerical results. Further, we present analytical solutions for the constant flow velocity case. The model is based on the assumption that the porous medium is stiff.

Keywords Advection–dispersion–reaction equation · Microbially induced carbonate precipitation (MICP) · Soil improvement · Numerical modelling · Analytical solution

1 Introduction

Biogrout is a soil improvement method that is based on microbially induced carbonate precipitation (MICP). The Biogrout process consists of two parts: the placement of the microbes and the precipitation of calcium carbonate. Some examples of applications are as follows:

- prevention of liquefaction [2, 3],
- bore hole stabilisation [4], and
- slope stabilisation [2].

The first step in the Biogrout process is the injection of bacteria. The bacteria will adsorb onto the porous matrix. That gives retardation. To fixate the bacteria onto the porous matrix, a fixation fluid is injected. This fixation fluid is a solution with a high salinity, and it will overtake the weakly adsorbed bacteria and strongly fix them onto the solid matrix. In [7], a model has been derived to describe the placement of the bacteria.

The second part in the Biogrout process is the injection of reactants. Urea ($CO(NH_2)_2$) and calcium chloride ($CaCl_2$) are injected into the soil. The bacteria catalyse the hydrolysis of urea, and ammonium (NH_4^+) and carbonate (CO_3^{2-}) are formed. In the presence of calcium ions (Ca^{2+}), the carbonate precipitates as calcium carbonate ($CaCO_3$). In [9], the reaction equations are discussed in more detail. The hydrolysis reaction is given by

$$CO(NH_2)_2 + 2H_2O \xrightarrow{\text{bacteria}} 2NH_4^+ + CO_3^{2-}. \tag{1}$$

The precipitation of calcium carbonate happens in several steps, depending on the pH. The overall reaction equation for the precipitation is given by

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3(s). \tag{2}$$

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Combining the hydrolysis reaction equation (1) and the reaction equation for the precipitation of calcium carbonate (2) gives the overall Biogrout reaction equation:

$$CO(NH_2)_2 + Ca^{2+} + 2H_2O(l) \rightarrow 2NH_4^+ + CaCO_3(s). \tag{3}$$

The side-product ammonium chloride (NH_4Cl) has to be removed. The solid calcium carbonate forms bridges between the sand grains. These bridges cause an increase in the strength and stiffness of the soil.

In [5] and [6], a model has been proposed for the transport and reaction of the reactants and the formation of calcium carbonate, based on the biochemical reaction Eq. 3. In [5, 6], a homogeneous bacterial activity was assumed, which is probably not realistic. Therefore, in [7], a model was derived that describes the placement of the bacteria. Solving the model equations gives the distribution of the bacteria. In this paper, these two models are combined to end up with a model that describes the placement of bacteria as well as the transport of the reactants and the formation of calcium carbonate. In Section 2, the (partial differential) equations are given for both models and shortly discussed. Further, analytical solutions are presented that are valid under idealised conditions. In Section 3, it is described which numerical methods are used to solve the model equations. Some results of the numerical simulations with the combined model are presented in Section 4 as well as a comparison with the model in which a homogeneous bacterial activity was assumed. Some discussion and conclusions can be found in Section 5.

2 Mathematical model

In this section, the model equations are given for the Biogrout process. In Section 2.1, the (partial differential) equations are given for the placement of the bacteria, whereas the equations for the precipitation of calcium carbonate are presented in Section 2.2. Finally, some analytical solutions are derived.

2.1 Model equations for the placement of the bacteria

The first step in the Biogrout process is the injection of bacteria. These bacteria will partly adsorb onto the solid matrix. This adsorption gives retardation. Next, a fluid with high salinity is injected into the subsoil. This solution acts as a fixation fluid to the bacteria. This fluid will overtake the adsorbed bacteria and strongly fixate them onto the matrix of the porous media. When, later on, the suspended bacteria are flushed away, the fixated bacteria stay in place and will play an important role in the precipitation of calcium carbonate, which is the second part in the Biogrout process. For completeness, we give the model equations for the placement of bacteria, as derived in [7].

The model for the placement of bacteria contains three phases of bacteria: bacteria in suspension, adsorbed bacteria and fixated bacteria. Concentration C^{bac} is defined as the bacterial concentration in suspension, concentration $\overline{C}^{\text{bac}}$ is the concentration of adsorbed bacteria and S^{bac} is the concentration of fixated bacteria. Note that, for convenience, the concentrations $\overline{C}^{\text{bac}}$ and S^{bac} have the same unit as C^{bac}, although the adsorbed and fixated bacteria are no longer in suspension but adsorbed or fixated onto the porous matrix. The following differential equations are derived for the concentrations of the bacteria:

$$\frac{\partial\left(\theta C^{\text{bac}}\right)}{\partial t} = \nabla\cdot\left(\mathbf{D}_{\text{bac}}\theta\nabla C^{\text{bac}}\right) - \nabla\cdot\left(\mathbf{q}C^{\text{bac}}\right) - \theta r_{\text{ads}} + \theta r_{\text{des}}, \tag{4}$$

$$\frac{\partial\left(\theta\overline{C}^{\text{bac}}\right)}{\partial t} = \theta r_{\text{ads}} - \theta r_{\text{des}} - \theta r_{\text{fix}}, \tag{5}$$

$$\frac{\partial\left(\theta S^{\text{bac}}\right)}{\partial t} = \theta r_{\text{fix}}, \tag{6}$$

In these equations, θ is the porosity, $\mathbf{D}_{\text{bac}}$ is the dispersion tensor, $\mathbf{q}$ is the Darcy flow velocity which relates to the pore water flow velocity $\mathbf{v}$ as $\mathbf{q} = \mathbf{v}\theta$, r_{ads} is the adsorption reaction rate, r_{des} is the desorption reaction rate and r_{fix} is the fixation reaction rate. The left-hand side of Eqs. 4, 5 and 6 models accumulation, the first term at the right-hand side of Eq. 4 represents the dispersion and diffusion of the bacteria and the second term is the advection term. Since the adsorbed and fixated bacteria can not be transported, there are no dispersion/diffusion and advection terms in Eqs. 5 and 6. The other terms in Eqs. 4, 5 and 6 stand for the adsorption, desorption and fixation reactions. These equations show that it is assumed that only adsorbed bacteria are fixated (only Eqs. 5 and 6 contain a fixation reaction term).

In the case of an equilibrium-controlled adsorption, the concentration of the adsorbed species tend to the adsorption isotherm. In the Biogrout process, there are both temporarily adsorbed and permanently adsorbed (fixed) bacteria. The adsorption isotherm $\varphi(C^{\text{bac}})$ depends on the concentration of bacterial cells in suspension (C^{bac}) and may also depend on properties of the

microorganisms, the porous medium and the pH. It has been assumed that the equilibrium of the permanently adsorbed bacteria is equal to $\beta\varphi(C^{\text{bac}})$ and that the equilibrium of the temporarily adsorbed bacteria is equal to $(1-\beta)\varphi(C^{\text{bac}})$. The fraction β ranges between 0 and 1. Its value depends on the concentration of the fixation fluid C^{fix} but may also depend on properties of the microorganisms, the pH and the porous medium.

As a driving force for the adsorption reaction, the difference between the adsorption isotherm and the concentration of the adsorbed (temporarily or permanently) bacteria is used. Adsorption only takes place when the adsorption isotherm is larger than the concentration of the adsorbed and fixated bacteria. That gives the following adsorption reaction rate:

$$r_{\text{ads}} = k_{\text{ads}} \left(\varphi(C^{\text{bac}}) - \left(\overline{C}^{\text{bac}} + S^{\text{bac}} \right) \right)_+ , \tag{7}$$

where k_{ads} is the adsorption reaction rate constant. The notation $(.)_+$ considers the positive part of an expression and has been defined as $(.)_+ := \max(0, .)$.

In the same way, the driving force for the fixation reaction is the difference between concentration S and its equilibrium $\beta\varphi(C^{\text{bac}})$ and fixation only takes place if S^{bac} is smaller than its equilibrium. We multiply this driving force by the concentration of adsorbed bacteria $\overline{C}^{\text{bac}}$ to guarantee that bacteria only can be fixated if there are adsorbed bacteria present, hence

$$r_{\text{fix}} = k_{\text{fix}} \overline{C}^{\text{bac}} \left(\beta\varphi \left(C^{\text{bac}} \right) - S^{\text{bac}} \right)_+ . \tag{8}$$

In this equation, k_{fix} is the fixation reaction constant.

As a reaction rate for desorption (the opposite phenomenon of adsorption), the following equation was derived:

$$r_{\text{des}} = k_{\text{des}} \left(\left(\overline{C}^{\text{bac}} - (1-\beta)\varphi \left(C^{\text{bac}} \right) \right) + \left(S^{\text{bac}} - \beta\varphi \left(C^{\text{bac}} \right) \right)_- \right)_+ , \tag{9}$$

where k_{des} is the desorption reaction rate. The notation $(.)_-$ has been defined as $(.)_- := \min(0, .)$, which implies that only the negative part of an expression is considered. Again, the driving force is the difference between the concentration of temporarily and permanently adsorbed bacteria (respectively, $\overline{C}^{\text{bac}}$ and S^{bac}) and their equilibria (respectively, being $(1-\beta)\varphi(C^{\text{bac}})$ and $\beta\varphi(C^{\text{bac}})$). Desorption only takes place if the difference is positive. Otherwise, it would be *ad*sorption. The term $(S^{\text{bac}} - \beta\varphi(C^{\text{bac}}))$ is only considered if it is negative for the following reason: Consider the case that S^{bac} is larger than its equilibrium, $S^{\text{bac}} > \beta\varphi(C^{\text{bac}})$ (which can happen, if C^{bac} is decreasing) and that $\overline{C}^{\text{bac}}$ is smaller than its equilibrium, $\overline{C}^{\text{bac}} < (1-\beta)\varphi(C^{\text{bac}})$, while the sum of adsorbed bacteria is larger than the adsorption isotherm, $\overline{C}^{\text{bac}} + S^{\text{bac}} > (1-\beta)\varphi(C^{\text{bac}}) + \beta\varphi(C^{\text{bac}}) = \varphi(C^{\text{bac}})$. The latter implies that there is a driving term for desorption and that the concentration of temporarily adsorbed bacteria $\overline{C}^{\text{bac}}$ will decrease. However, this temporarily adsorbed bacteria concentration is already smaller than its equilibrium, which would give adsorption rather than desorption. Hence, if S^{bac} is larger than its equilibrium, the difference should not contribute to desorption. That explains why this difference is only taken into account if it is negative.

The ratio β depends on the concentration of fixation fluid C^{fix}. As a relation between β and the concentration of fixation fluid, the following Monod equation is used:

$$\beta = \beta_0 \frac{C^{\text{fix}}}{K_{\text{m,fix}} + C^{\text{fix}}}, \tag{10}$$

for some positive constant β_0.

For this concentration of fixation fluid, the following partial differential equation is derived:

$$\frac{\partial \left(\theta C^{\text{fix}} \right)}{\partial t} = \nabla \cdot \left(\mathbf{D}_{\text{fix}} \theta \nabla C^{\text{fix}} \right) - \nabla \cdot \left(\mathbf{q} C^{\text{fix}} \right), \tag{11}$$

where $\mathbf{D}_{\text{fix}}$ is the dispersion tensor. The left-hand side of this equation models accumulation, the first term at the right-hand side stands for dispersion and diffusion and the last term is the advection term.

For the simulations in this paper, a Langmuir adsorption isotherm is used, as given in [10]:

$$\varphi(C^{\text{bac}}) = \frac{K_{\text{l}} \overline{C}_{\max} C^{\text{bac}}}{1 + K_{\text{l}} C^{\text{bac}}}, \tag{12}$$

where the positive constant K_{l} denotes the Langmuir constant and $\overline{C}_{\max}$ is the maximum adsorption capacity.

A differential equation for the flow is given in the next subsection.

2.2 Model equations for the precipitation of calcium carbonate

After the placement of bacteria, urea ($CO(NH_2)_2$) and calcium chloride ($CaCl_2$) are injected into the soil. The bacteria provide the hydrolysis of urea according to reaction (1). Carbonate (CO_3^{2-}) and ammonium (NH_4^+) are formed. The carbonate precipitates with the calcium (Ca^{2+}) as calcium carbonate ($CaCO_3$), see

precipitation reaction (2). The solid calcium carbonate causes a (slight) decrease in porosity and in permeability, which has an influence on the flow and the pressure. The hydrolysis and precipitation reactions influence the density of the solution. In [5, 6], partial differential equations are given for the concentration of urea, calcium chloride, ammonium chloride and calcium carbonate, for the pressure and the flow, as well as relations for the porosity, permeability and density of the fluid. In this subsection, we repeat them and shortly discuss them.

We start with the partial differential equations for the aqueous species:

$$\frac{\partial(\theta C^i)}{\partial t} = \nabla \cdot (\theta \mathbf{D} \cdot \nabla C^i) - \nabla \cdot (\mathbf{q} C^i) + n_i \theta r_{\text{hp}}. \tag{13}$$

In this equation, θ is the porosity, C^i is the dissolved concentration of species i, $i \in \{\text{urea}, Ca^{2+}, NH_4^+\}$ with M (equal to kilomoles per cubic meter) as a unit, $\mathbf{D}$ is the dispersion tensor, $\mathbf{q}$ is the Darcy velocity, n_i is a constant that deals with the stoichiometry in the biochemical reaction Eq. 3 and r_{hp} is the reaction rate of the production of calcium carbonate, which is a function of the urea concentration and the bacterial concentrations. From the stoichiometry of reaction (3), the values of n_i for the various aqueous species are given by $n_{\text{urea}} = -1$, $n_{Ca^{2+}} = -1$ and $n_{NH_4^+} = 2$.

The left-hand side of Eq. 13 stands for the accumulation. In the right-hand side, we have terms for dispersion/diffusion, for the advection and for the biochemical reaction (3).

For the reaction rate r_{hp} of Eq. 3, the following relation has been used:

$$r_{\text{hp}} = v_{\max} \frac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}} \left(C^{\text{bac}} + \overline{C}^{\text{bac}} + S^{\text{bac}} \right). \tag{14}$$

Here, $v_{\max}$ is the bacterial conversion rate constant and $K_{\text{m,urea}} \geq 0$ is the saturation constant.

For the concentration of the solid calcium carbonate C^{CaCO_3}, we have the following differential equation:

$$\frac{\partial C^{\text{CaCO}_3}}{\partial t} = m_{\text{CaCO}_3} \theta r_{\text{hp}}. \tag{15}$$

In this equation, m_{CaCO_3} is the molar mass of calcium carbonate, which is used to convert moles into mass. Since it has been assumed that the calcium carbonate is not transported, there are no transport terms in the differential equation. Hence, Eq. 15 only contains an accumulation term and a reaction term.

The solid calcium carbonate that is formed in the pores causes a decrease in porosity. The difference $(C^{\text{CaCO}_3}(t) - C^{\text{CaCO}_3}(0))$ gives the amount of calcium carbonate that has been formed per unit of volume. Division by the density of calcium carbonate ρ_{CaCO_3} gives the decrease in pore volume per unit of volume. That leads to the following relation between the calcium carbonate concentration and the porosity:

$$\theta(t) = \theta(0) - \frac{C^{\text{CaCO}_3}(t) - C^{\text{CaCO}_3}(0)}{\rho_{CaCO_3}}. \tag{16}$$

For the flow, we use the continuity equation that was derived in [8], which is an adaptation of the differential equation derived in [6].

$$\nabla \cdot \mathbf{q} = K \theta r_{\text{hp}}. \tag{17}$$

The constant K has been defined as

$$K := \left(\frac{m_{CaCO_3}}{\rho_{CaCO_3}} - (1 - V_s) \right). \tag{18}$$

In this definition, $1 - V_s$ is the decrease of liquid volume per number of converted urea particals according to the biochemical reaction (3). In [8], we compared Eq. 17 to another differential equation for the flow:

$$\frac{\partial(\rho\theta)}{\partial t} = -\nabla \cdot (\rho \mathbf{q}) - m_{CaCO_3} \theta r_{\text{hp}}. \tag{19}$$

The results were very similar, at least in 1-D, but Eq. 17 turned out to be more stable than Eq. 19. We also note that Eq. 17 is consistent with the Oberbeck–Boussinesq approximation as $r_{\text{hp}} \to 0$, i.e. in absence of the reaction.

As a relation between the flow and the pressure p, Darcy's law is used [10]:

$$q_{\text{x}} = -\frac{k_{\text{x}}}{\mu} \frac{\partial p}{\partial x}, \tag{20}$$

$$q_{\text{y}} = -\frac{k_{\text{y}}}{\mu} \frac{\partial p}{\partial y}, \tag{21}$$

$$q_{\text{z}} = -\frac{k_{\text{z}}}{\mu} \left(\frac{\partial p}{\partial z} + \rho_{\text{l}} g \right). \tag{22}$$

In Darcy's law, k_i is the intrinsic permeability in the various coordinate directions, $i \in \{x, y, z\}$, μ is the viscosity of the fluid, ρ_{l} is the density of the fluid and g is the gravitational constant.

The Kozeny–Carman equation is used to determine the intrinsic permeability. This equation is an empirical relation between the intrinsic permeability and the porosity that is commonly used in ground water flow modelling (see [1]):

$$k = k_{\text{x}} = k_{\text{y}} = k_{\text{z}} = \frac{(d_{\text{m}})^2}{180} \frac{\theta^3}{(1-\theta)^2}. \tag{23}$$

In this relation, d_{m} is the mean particle size of the sand.

Table 1 Boundary conditions for the various concentrations and the flow for the one-dimensional configuration

	Γ_1			Γ_2
	Phase 1	Phase 2	Phase 3	Phase 1–3
C^{bac}	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = q_{\text{in}}c_{\text{in}}$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$\frac{\partial C}{\partial n} = 0$
C^{fix}	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = q_{\text{in}}c_{\text{in}}$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$\frac{\partial C}{\partial n} = 0$
C^{urea}	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = q_{\text{in}}c_{\text{in}}$	$\frac{\partial C}{\partial n} = 0$
$C^{\text{Ca}^{2+}}$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = q_{\text{in}}c_{\text{in}}$	$\frac{\partial C}{\partial n} = 0$
$C^{\text{NH}_4^+}$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$(D\theta\nabla C - \mathbf{q}C)\cdot\mathbf{n} = 0$	$\frac{\partial C}{\partial n} = 0$
q	$q = q_{\text{in}}$	$q = q_{\text{in}}$	$q = q_{\text{in}}$	

For the fluid density, the empirical relation that is given in [6] is used:

$$\rho_{\text{l}} = 1000 + 15.4996C^{\text{urea}} + 86.7338C^{\text{Ca}^{2+}} + 15.8991C^{\text{NH}_4^+}. \tag{24}$$

The bacteria hardly influence the density. Hence, they are not taken into account in the density calculation.

Substituting Eqs. 20, 21 and 22 into Eq. 17, using relation (23), gives a partial differential equation for the pressure, which can be used to solve for the flow pattern if the boundary conditions are given in terms of pressure, or if density differences influence the flow.

$$\nabla\cdot\mathbf{q} = \nabla\cdot\left(-\frac{k}{\mu}(\nabla p + \rho_{\text{l}}g\mathbf{e_z})\right) = K\theta r_{\text{hp}}. \tag{25}$$

Here, $\mathbf{e_z}$ is the unit vector in vertical direction, taken positive upwards.

2.3 Boundary conditions and initial conditions

We consider a one-dimensional configuration, which corresponds to a column. We take a line with a length of 1 m, with inflow at the left-hand side (Γ_1) and outflow at the right-hand side (Γ_2).

The injection strategy exists of three phases. During phase 1, from time $t = T_0 = 0$ h until time $t = T_1$, bacteria are injected. During phase 2, from time $t = T_1$ until time $t = T_2$, a fixation fluid is injected. The third phase is from time $t = T_2$ until time $t = T_{\text{end}}$. During this phase, urea and calcium chloride are injected.

Table 1 gives the boundary conditions for the one-dimensional configuration.

Initially, all concentrations are equal to zero. The initial porosity is equal to some constant θ_0. Since the partial differential equations for the concentration of urea and the concentration of calcium ions are the same, as well as the initial and boundary conditions, both concentration–distributions are identical. Therefore, we only consider the concentration of urea. We use the parameter values given in Table 2.

2.4 Analytical solution

In this subsection, an analytical solution is derived for a simplified version of system (4)–(18). For the analytical solution, we restrict ourselves to one dimension. Furthermore, the reaction constants are infinitely large: $k_{\text{ads}}, k_{\text{des}}, k_{\text{fix}} \to \infty$ and dispersion and diffusion are neglected: $\mathbf{D_i} = 0$ for $i \in \{\text{bac, fix, urea, Ca}^{2+}, \text{NH}_4^+\}$. The decrease of the porosity and the change of liquid volume as a result of the reaction are also neglected: $\theta(x,t) = \theta_0$ and $K = 0$. As we consider one- dimensional flow without sinks and sources and a constant porosity, the pore water velocity v is constant.

The analytical solution for the concentration of suspended bacteria C^{bac}, the concentration of temporarily adsorbed bacteria $\overline{C}^{\text{bac}}$ and the concentration of fixated

Table 2 Values that are taken for the various parameters

$\alpha_{\text{bac}} = 0.001$ m,	$\alpha_{\text{fix}} = 0.001$ m,	$\alpha_{\text{urea}} = 0.001$ m,
$\alpha_{\text{Ca}^{2+}} = 0.001$ m,	$\alpha_{\text{NH}_4^+} = 0.001$ m,	$\mathbf{D}_{\text{m,bac}} = 10^{-9}$ m^2/s,
$\mathbf{D}_{\text{m,fix}} = 10^{-9}$ m^2/s,	$\mathbf{D}_{\text{m,urea}} = 10^{-9}$ m^2/s,	$\mathbf{D}_{\text{m,Ca}^{2+}} = 10^{-9}$ m^2/s,
$\mathbf{D}_{\text{m,NH}_4^+} = 10^{-9}$ m^2/s,	$K_{\text{l}} = 0.5$ [1],	$\overline{C}_{\text{max}} = 1$ [1],
$\beta_0 = 0.505$ [1],	$K_{\text{m,urea}} = 0.01$ kmol/m^3,	$K_{\text{m,fix}} = 0.01$ kmol/m^3,
$m_{\text{CaCO}_3} = 100.1$ kg/kmol,	$\rho_{\text{CaCO}_3} = 2710$ kg/m^3,	$V_{\text{s}} = 0.97035$ m^3/kmol,
$d_{\text{m}} = 200$ μm,	$g = 9.81$ m/s^2,	$\theta_0 = 0.35$ [1],
$q_{\text{in}} = 0.35$ m/h,	$v_{\text{max}} = 0.72$ kmol/m^3/h,	$c_{\text{in}} = 1$ [1] or kmol/m^3,
$T_1 = 0.5$ h,	$T_2 = 1.0$ h,	$T_{\text{end}} = 2.0$ h.

bacteria S^{bac} are derived in [7]. We give the analytical solution for C^{bac}:

$$C^{\text{bac}} = \begin{cases} 1 & \text{for } (t,x) \in (0,T_1)\times(0,s(t)) \cup (T_1,T_3)\times(x_R(t),s(t)); \\ 0 & \text{for } (t,x) \in (T_1,\infty)\times(0,x_L(t)) \cup \mathbb{R}^+ \times (s(t),\infty); \\ \dfrac{1}{K_1}\left(\sqrt{\dfrac{(1-\beta(1))K_1\overline{C}_{\max}\frac{x}{t-T_1}}{v-\frac{x}{t-T_1}}}-1\right) & \text{for } (t,x) \in (T_1,\infty)\times(x_L(t),\min(x_R(t),s(t))), \end{cases} \tag{26}$$

where the shock speed $s(t)$ is given by

$$s(t) = \begin{cases} \dfrac{vt}{1+\varphi(1)} & \text{for } t < T_3; \\ \dfrac{v(1+K_1)T_3}{1+K_1+K_1\overline{C}_{\max}} + \displaystyle\int_{T_3}^{t} \frac{v\sqrt{(1-\beta(1))K_1\overline{C}_{\max}(\bar{t}-T_1)}}{\sqrt{(1-\beta(1))K_1\overline{C}_{\max}(\bar{t}-T_1)}+K_1\overline{C}_{\max}\sqrt{s(\bar{t})-(\bar{t}-T_1)}}\,d\bar{t} & \text{for } t > T_3, \end{cases} \tag{27}$$

and the location of the endpoints of the constant states are determined by

$$x_L = \frac{v(t-T_1)}{1+(1-\beta(1))K_1\overline{C}_{\max}}; \tag{28}$$

$$x_R = \frac{v(t-T_1)}{1+\frac{(1-\beta(1))K_1\overline{C}_{\max}}{(1+K_1)^2}}. \tag{29}$$

Time T_3 is the time at which the shock speed of the bacteria changes, which is calculated from

$$T_3 = \frac{\frac{(1+K_1)^2}{K_1\overline{C}_{\max}}+(1+K_1)}{K_1+\beta(1)}T_1. \tag{30}$$

The concentrations of temporarily adsorbed bacteria $\overline{C}^{\text{bac}}$ and fixated bacteria S^{bac} are determined as a function of the concentration of suspended bacteria C^{bac}:

$$\overline{C}^{\text{bac}} = \left(1-\beta\left(c^{\text{fix}}\right)\right)\frac{K_1\overline{C}_{\max}C^{\text{bac}}}{1+K_1C^{\text{bac}}}, \tag{31}$$

$$S^{\text{bac}} = \max_{0\le\tilde{t}\le t}\left\{\beta\left(c^{\text{fix}}\right)\frac{K_1\overline{C}_{\max}C^{\text{bac}}}{1+K_1C^{\text{bac}}}\right\}, \tag{32}$$

see [7] for a derivation.

In [7], the ratio β is given by $\beta(C^{\text{fix}}) = \beta_0 C^{\text{fix}}$. In this article, we use the more complex but also more physical relation $\beta(C^{\text{fix}}) = \beta_0\frac{C^{\text{fix}}}{K_{\text{m,fix}}+C^{\text{fix}}}$, see relation (10). Here, β_0 has a somewhat larger value, such that the value of $\beta(1)$ is the same as for the case in [7].

In this article, the fixation fluid is only injected for a finite time, while in [7], the injection of fixation fluid is never stopped. In this article, a solution containing Ca^{2+} is being injected after the injection of fixation fluid. The Ca^{2+} is needed for the precipitation reaction but also acts as a fixation fluid due to its high salinity. Hence, the solution in [7] is still valid for this study.

The analytical solution for the concentration of urea is constructed with the method of characteristics. Along characteristics, we have

$$\begin{aligned}\frac{d}{dt}C^{\text{urea}}(t,x(t)) &= C^{\text{urea}}_t + C^{\text{urea}}_{\text{x}}x'(t) \\ &= -v_{\max}\frac{C^{\text{urea}}}{K_{\text{m,urea}}+C^{\text{urea}}} \\ &\quad\times\left(C^{\text{bac}}+\overline{C}^{\text{bac}}+S^{\text{bac}}\right),\end{aligned} \tag{33}$$

with

$$x'(t) = v. \tag{34}$$

The injection of the urea starts at time T_2. This time has been chosen in such a way that the urea does not overtake the non-fixated bacteria within the domain. We define x_{fu} as the position of the urea front. If x_{fu}, $x_{\text{L}} < L$, it should hold that

$$x_{\text{fu}} = v(t-T_2) < x_{\text{L}}. \tag{35}$$

Hence, everywhere in the domain where the urea concentration is non-zero, only fixated bacteria are left. The length of the domain L has been chosen such that finally a constant concentration of fixated bacteria is reached, which is the case if $L < s(T_3)$. Therefore, on

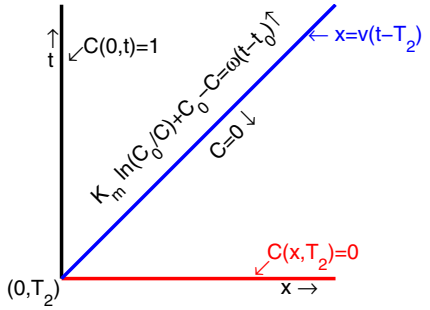

Fig. 1 The $(x$-$t)$ diagram for the concentration of urea. In this diagram, we have that $C = C^{\text{urea}}$, $C_0 = C_0^{\text{urea}}$ and $\omega = v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}$

the locations where the concentration of urea is non-negative, it holds that

$$C^{\text{bac}} + \overline{C}^{\text{bac}} + S^{\text{bac}} = S^{\text{bac}} = \beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}, \tag{36}$$

which is a constant. This constant is substituted into Eq. 33.

A solution to Eq. 33 is the trivial solution:

$$C^{\text{urea}}(x,t) = 0. \tag{37}$$

The non-trivial solution can be found by application of separation of variables on Eq. 33 to give the following implicit solution:

$$K_{\text{m,urea}} \ln\left(\frac{C_0^{\text{urea}}}{C^{\text{urea}}}\right) + C_0^{\text{urea}} - C^{\text{urea}} = v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}(t-t_0), \tag{38}$$

with C_0^{urea} the concentration at time t_0.

Figure 1 displays the $(x$-$t)$-diagram for the concentration of urea.

The factors that determine the concentration of urea at time t and location x for the non-trivial case are the initial concentration and the time difference between time t and the starting point of the characteristic on the t-axis t_0. With Eq. 34, we find that this time difference equals $t - t_0 = x/v$ and hence

$$\begin{aligned} &K_{\text{m,urea}} \ln\left(\frac{C_0^{\text{urea}}}{C^{\text{urea}}}\right) + C_0^{\text{urea}} - C^{\text{urea}} \\ &\quad = v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}(t-t_0) \\ &\quad = v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}\frac{x}{v}. \end{aligned} \tag{39}$$

That implies that the concentration of urea has a fixed value on a fixed position x for a time $t > T_2 + x/v$. Further, $C^{\text{urea}} = 0$ for $t < T_2 + \frac{x}{v}$. These identities will be used in the construction of the solution for the calcium carbonate concentration.

The partial differential equation for the concentration of calcium carbonate is given in Eq. 15, which contains reaction rate r_{hp}. This reaction rate is given in Eq. 14. Substituting relation (36) in this rate gives the following differential equation for the concentration of calcium carbonate:

$$\begin{aligned} \frac{\partial C^{\text{CaCO}_3}}{\partial t} &= m_{\text{CaCO}_3}\theta r_{\text{hp}} \\ &= m_{\text{CaCO}_3}\theta v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}\frac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}}. \end{aligned} \tag{40}$$

Integrating this equation leads to

$$\begin{aligned} C^{\text{CaCO}_3}(x,t) &= \int_0^t \frac{\partial C^{\text{CaCO}_3}}{\partial \bar{t}}\,d\bar{t} \\ &= m_{\text{CaCO}_3}\theta v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1} \\ &\quad \times \int_0^t \frac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}}\,d\bar{t}. \end{aligned} \tag{41}$$

Since it holds that $C^{\text{urea}} = 0$ for $0 \le \bar{t} < T_2 + x/v$ and since C^{urea} is equal to a constant for $\bar{t} \ge T_2 + x/v$ on a fixed position x, Eq. 41 becomes

$$C^{\text{CaCO}_3}(x,t) = \begin{cases} 0 & \text{for } t < T_2 + \dfrac{x}{v}; \\ m_{\text{CaCO}_3}\theta v_{\max}\beta(1)\dfrac{K_1\overline{C}_{\max}}{1+K_1}\displaystyle\int_{T_2+\frac{x}{v}}^{t} \frac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}}\,d\bar{t}; & \text{for } t \ge T_2 + \dfrac{x}{v}. \end{cases} \tag{42}$$

$$= \begin{cases} 0 & \text{for } t < T_2 + \dfrac{x}{v}; \\ m_{\text{CaCO}_3}\theta v_{\max}\beta(1)\dfrac{K_1\overline{C}_{\max}}{1+K_1}\dfrac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}}\left(t - T_2 - \dfrac{x}{v}\right); & \text{for } t \ge T_2 + \dfrac{x}{v}. \end{cases} \tag{43}$$

$$= m_{\text{CaCO}_3}\theta v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}\frac{C^{\text{urea}}}{K_{\text{m,urea}} + C^{\text{urea}}}\left(t - T_2 - \frac{x}{v}\right)_+. \tag{44}$$

In the derivation of these analytical solutions, we substituted relation (36) into rate (14). According to this equation, the reaction rate is related to the concentration of urea via a Monod equation. For completeness, we consider the case that the rate is linear in the urea concentration. Then, we have

$$r_{\mathrm{hp}} = v_{\max}\beta(1)\frac{K_1\overline{C}_{\max}}{1+K_1}C^{\mathrm{urea}}. \tag{45}$$

Then, the analytical solution for the concentration of urea and calcium carbonate reads as follows:

$$C^{\mathrm{urea}}(x,t) = C_0^{\mathrm{urea}}(x-vt)\exp\left\{-v_{\max}\frac{K_1\overline{C}_{\max}}{1+K_1}t\right\}, \tag{46}$$

$$= \begin{cases} 0 & \text{for } t < T_2 + x/v; \\ \exp\left\{-v_{\max}\dfrac{K_1\overline{C}_{\max}}{1+K_1}\dfrac{x}{v}\right\} & \text{for } t \geq T_2 + x/v, \end{cases} \tag{47}$$

$$C^{\mathrm{CaCO_3}}(x,t) = \begin{cases} 0 & \text{for } t < T_2 + \dfrac{x}{v}; \\ (t - T_2 - x/v)v_{\max}\beta(1)\dfrac{K_1\overline{C}_{\max}}{1+K_1}\theta\exp\left\{-v_{\max}\dfrac{K_1\overline{C}_{\max}}{1+K_1}\dfrac{x}{v}\right\} & \text{for } t \geq T_2 + \dfrac{x}{v}. \end{cases} \tag{48}$$

3 Numerical methods

The differential equations for the pressure, the flow and the concentrations of fixation fluid, bacteria and the aqueous species are solved by the standard Galerkin finite element method. The weak formulations have been derived by multiplication by a test function $\eta \in H^1(\Omega)$ and integration over the domain Ω. The Newton–Cotes quadrature rules are used for the development of the element matrices and vectors. Furthermore, line elements are used, as well as linear basis functions. For the time integration, the Euler backward method is used.

The differential equations for the concentrations of bacteria (Eqs. 4, 5 and 6) are coupled due to the reaction terms r_{ads} (Eq. 7), r_{des} (Eq. 9) and r_{fix} (Eq. 8). Due to the Langmuir isotherm (Eq. 12), the differential equations are non-linear in the concentration of suspended bacteria C^{bac}. Hence, Newton's method is used to solve for the differential equations for the concentrations of bacteria. By doing so, the three various concentrations of bacteria come together in one matrix-vector system.

Since the differential equation for the concentration of urea is also non-linear in the concentration, due to the reaction term, Newton's method is used to calculate the concentration of urea.

The partial differential equation for the concentration of calcium carbonate, Eq. 15, can be considered as an ordinary differential equation in each grid point. To calculate the concentration of calcium carbonate, the following scheme is used:

$$\left(C^{\mathrm{CaCO_3}}\right)^{\mathrm{n+1}} = \left(C^{\mathrm{CaCO_3}}\right)^{\mathrm{n}} + \Delta t m^{\mathrm{CaCO_3}}\theta^{\mathrm{n}} r_{\mathrm{hp}}^{\mathrm{n+1}}, \tag{49}$$

which uses the porosity θ from the previous time step and the reaction rate r_{hp} (Eq. 14) from the current time step.

As a step size for the time integration is taken $\Delta t = \frac{1}{640}$ h and as the length of an element is taken $\Delta x = \frac{1}{640}$ m. For a more detailed description of the numerical methods, see [5–7].

At each time step, the equations are solved sequentially in the following order: first, the flow is calculated. This can be done by solving the differential equation for the pressure (25), and from this pressure, the flow is calculated with Darcy's law, Eqs. 20, 21 and 22. Since the pressure is not involved in the boundary terms for the flow in this case, the flow can be calculated directly from Eq. 17. Subsequently, the partial differential equation for the concentration of the fixation fluid Eq. 11 is solved. Then, the equations for the concentrations of bacteria Eqs. 4, 5 and 6 are solved as a coupled system, applying Newton's method. These concentrations partly determine the reaction rate r_{hp} (Eq. 14) of the biochemical reaction, given by Eq. 3. The partial differential equation for the urea concentration Eq. 13 is solved, again, using Newton's method, and the reaction rate r_{hp} Eq. 14 is updated. Usually, the Newton method converges in approximately three iterations. Finally, the concentration of ammonium ($C^{\mathrm{NH_4^+}}$) and calcium carbonate ($C^{\mathrm{CaCO_3}}$) are calculated sequentially and the porosity (θ), intrinsic permeability (k) and fluid density (ρ_{l}) are updated by using Eqs. 13, 15, 16, 23 and 24, respectively.

4 Results

In this section, some analytical and numerical results are shown, as well as a comparison of the current model and the model with a homogeneous bacterial activity that was used in [5, 6]. The numerical results are in Section 4.1, the comparison of the two models is in Section 4.2, the analytical results are in Section 4.3 and the comparison of the numerical and analytical solutions is in Section 4.4.

4.1 Numerical results

We start with the one-dimensional configuration. Numerical simulations have been done for two different values of the adsorption, desorption and fixation reaction rate constant. The first value is $K_{\text{bac}} := k_{\text{ads}} = k_{\text{des}} = k_{\text{fix}} = 10\ \text{h}^{-1}$. The results are displayed in the left graphs of Fig. 2. As a second value has been chosen $K_{\text{bac}} = k_{\text{ads}} = k_{\text{des}} = k_{\text{fix}} = 1{,}000\ \text{h}^{-1}$. The results for that value are displayed in the right graphs of Fig. 2. A small reaction constant means that the process is slow. The larger the reaction constant is, the more the result tends to the equilibrium.

The top graphs of Fig. 2 show a situation in the first phase, in which bacteria are injected. The graphs show a non-zero concentration of suspended bacteria (C^{bac}) and adsorbed bacteria ($\overline{C}^{\text{bac}}$, in the legend called C^{bar}). The concentration of adsorbed bacteria in the equilibrium case is a function of the concentration of suspended bacteria as described by the Langmuir isotherm (Eq. 12). The bacteria enter the domain with a steep front, somewhat smoothened by dispersion and diffusion. Retardation of the front takes place due to the adsorption process. The top right graph of Fig. 2 shows a situation that is close to equilibrium. The top left graph, where the adsorption process is slow compared to the top right graph, has a very smooth front. Since, in this phase, fixation fluid is not yet being injected, there are no fixated bacteria.

The second row of Fig. 2 shows a situation in the second phase, where fixation fluid is injected. The concentration of fixated bacteria, S^{bac}, is no longer zero. Since in the right graph, the fixation rate constant is larger than in the left graph and the concentration of fixated bacteria is also higher there. In the left graph, the concentration of adsorbed bacteria is larger than the concentration of suspended bacteria in a part of the domain. The reason is the slow desorption process.

The bottom four graphs of Fig. 2 display some shots of phase 3, where urea and calcium chloride are injected. Note that the calcium carbonate concentration is scaled, such that the range is comparable to the range of the other graphs. Since the concentration of fixated bacteria in the left graphs is lower than in the right graphs, the concentration of calcium carbonate is lower as well. An exception to this situation is the zone around $x = 0.6$ m in the bottom graphs. Although the concentration of fixated bacteria is smaller for a smaller K_{bac}-value, there are still adsorbed and suspended bacteria left in that zone, which also contribute to the hydrolysis of urea and hence to a higher calcium carbonate concentration. The calcium carbonate concentration in the left graphs has its maximum somewhere in the middle of the domain, whereas in the right graph, the maximum is close to the injection point.

Both for the calculation of the concentration of the bacteria and the concentration of urea, Newton iterations are performed. As long as the concentrations are constant, only one iteration is needed for convergence. Else, for the calculation of the urea concentration, approximately three iterations are needed for convergence and approximately three or four iterations are needed for the calculation of the bacteria. Although the number of iterations that is needed for convergence is almost similar, the CPU time per iteration differs significantly. It takes nine times as much CPU time per Newton iteration to calculate the (three) concentrations of bacteria as to calculate the urea concentration. The reason is that the matrix that is built for the calculation of the concentrations of the bacteria is nine times as large as the matrix for the calculation of the urea concentration since the concentrations of bacteria are solved from one matrix-vector system.

4.2 Results of the comparison between the current model and the model with a homogeneous distribution of bacteria

In this section, the current model is compared to the previous model, where the previous model assumes a homogeneous distribution of bacteria. As in the previous subsections, this comparison is carried out for two K_{bac}-values: $K_{\text{bac}} = 10\ \text{h}^{-1}$ and $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$. To be able to make a good comparison, the average of the concentration of fixated bacteria in the current model is used as a value for the (constant) concentration of fixated bacteria in the previous model. Some results of this comparison are shown in Fig. 3. The left graphs display the results for the low K_{bac}-value, $K_{\text{bac}} = 10\ \text{h}^{-1}$, and the right graphs show the results for the high K_{bac}-value, $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$.

The top graphs show the concentration of fixated bacteria at time $t = 2$ h. The right plot, which displays the situation for a high K_{bac}-value, shows two graphs that are almost similar. Only at the inflow boundary that a large difference is visible. That has the following reason. First, a pulse with bacteria is injected, without injection of fixation fluid. There is no fixation fluid in the domain, so there are only non-fixated bacteria. Then a pulse with fixation fluid is injected. Bacteria are only fixated at that location where both bacteria and a

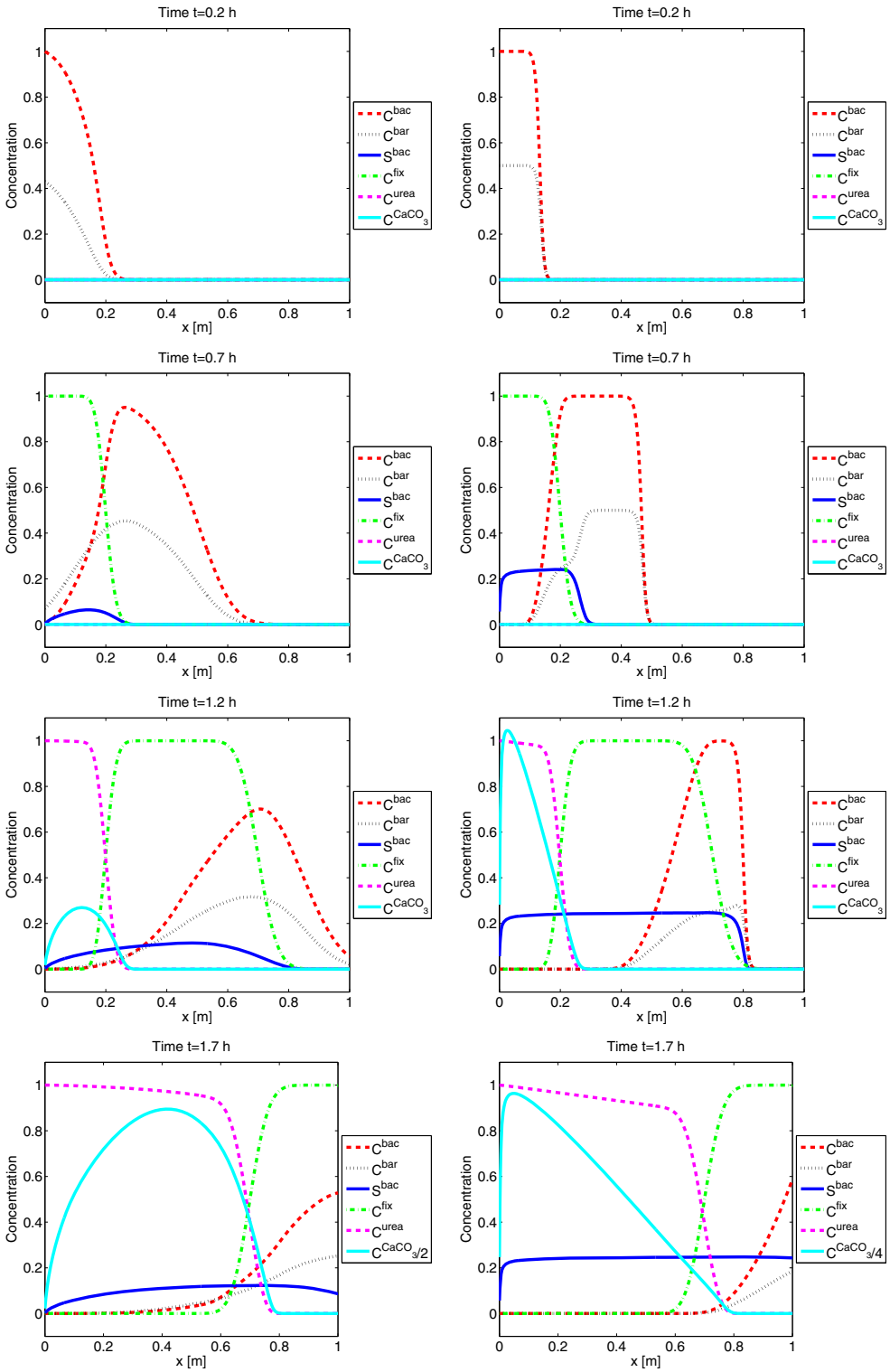

Fig. 2 Numerical solution for the concentration of suspended, temporarily adsorbed and fixated bacteria and the concentration of fixation fluid, urea and $CaCO_3$ as a function of location at several times ($t = 0.2$ h, $t = 0.7$ h, $t = 1.2$ h and $t = 1.7$ h) for $K_{\text{bac}} = 10\ \text{h}^{-1}$ (*left graphs*) and $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$ (*right graphs*)

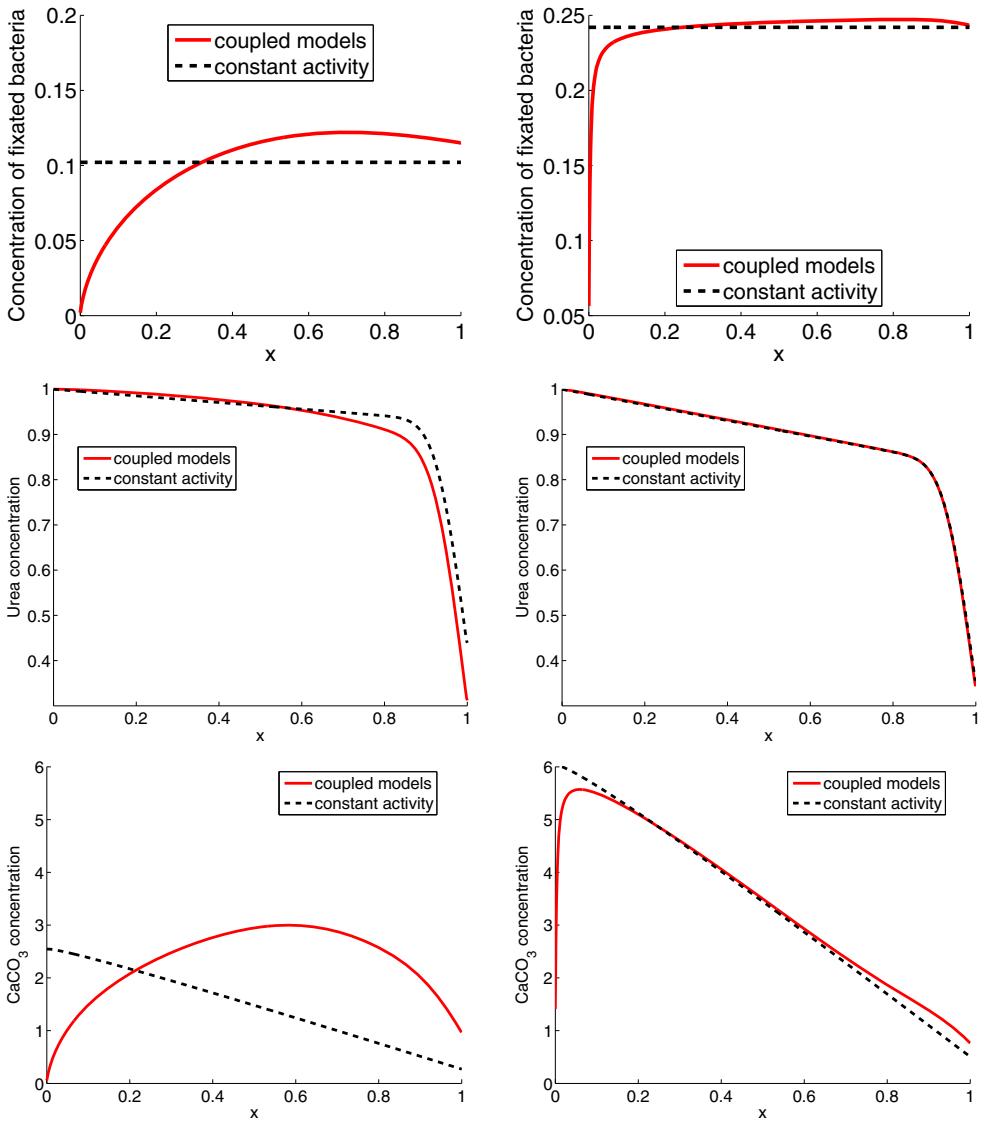

Fig. 3 Several concentrations as a function of location at time $t = 2$ h, for $K_{bac} = 10\ h^{-1}$ (*left graphs*) and $K_{bac} = 1{,}000\ h^{-1}$ (*right graphs*). *Top graphs* concentration of fixated bacteria, *middle graphs* urea concentration and *bottom graphs* $CaCO_3$ concentration

fixation fluid are present. The latter can only happen where the fixation fluid is overtaking the bacteria since they are injected after each other. The reason that they are not injected together is that this would result in clogging in the injection filter, that leads to stoppage of the filter. Hence, the injection point is a critical point, where (almost) no bacteria are fixated.

The middle graphs of Fig. 3 show the urea concentration at time $t = 2$ h. For the high K_{bac}-value, there is visually no difference. But also in the left plot, there is only a small difference between the graphs.

The bottom graphs display the concentration of calcium carbonate. Again, the concentrations from the high K_{bac}-value are similar except near the injection point. The concentrations, calculated with the low K_{bac}-value, however, show a large difference.

It can be concluded that, if the concentration of fixated bacteria is similar for both models, the calcium carbonate profile is similar as well. A high reaction constant leads to a homogeneous bacterial distribution, at least for the first part of the domain, except for the region around the injection point. A low reaction rate constant, corresponding to slow adsorption, desorption and fixation processes, leads to a non-homogeneous bacterial distribution and hence to a different calcium carbonate profile. An instantaneous equilibrium, however, is not a guarantee that the bacterial distribution will be homogeneous. The concentration of fixated bacteria does not depend on the length of the domain. Since only a finite amount of bacteria is injected, the domain can be chosen so large that only around the injection boundary bacteria are fixated and that there

are no bacteria in the rest of the domain. This can also be seen from the analytical solution for the instantaneous equilibrium, Eqs. 26, 31 and 32. In order to get a homogeneous distribution of bacteria in this case, more bacteria should be injected, possibly via multiple injection points.

4.3 Analytical results

The analytical solution for the equilibrium case, while dispersion, diffusion, decrease of the porosity and the change of liquid volume are neglected, is shown in Fig. 4, as a reaction rate has been taken Eq. 14. This figure shows the analytical solutions at the same times as the numerical solutions are shown (Fig. 2). The top left graph shows a situation of the first phase where only bacteria are injected. The top right graphs displays a shot of the second phase, in which fixation fluid is injected. Where both bacteria and fixation fluid are present, bacteria are fixated. The bottom graphs show two shots of the last phase in which calcium carbonate is formed.

4.4 Comparison of the numerical solutions to the analytical solutions

In this subsection, we compare the numerical and analytical solution for the concentration of urea and calcium carbonate. The comparison for the concentrations of bacteria for the bacterial injection model has been made in [7]. In order to make a valid comparison, we redo our numerical simulations for $\mathbf{D}_{\text{bac}} = \mathbf{D}_{\text{fix}} = \mathbf{D}_{\text{urea}} = \mathbf{D}_{Ca^{2+}} = \mathbf{D}_{NH_4^+} = \mathbf{0}$ and for a constant porosity and flow rate. We do take a finite K_{bac}-value, however, namely $K_{\text{bac}} = 10\ \text{h}^{-1}$ and $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$.

Figure 5 shows the numerical and analytical solution of the concentration of urea and calcium carbonate. The figures display the situation at times $t = 1.2$ h and $t = 1.7$ h, as in Figs. 2 and 4. The results at time $t = 0.2$ h and $t = 0.7$ h are not shown since the urea and calcium carbonate concentration are zero then. Again, the left graphs show the situation for $K_{\text{bac}} = 10\ \text{h}^{-1}$ and the right graphs for $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$.

In all the graphs of Fig. 5, the analytical solution of the urea concentration corresponds well with the numerical solution. The front of the numerical solution is less steep than the front of the analytical solution due to numerical diffusion. For the low K_{bac}-value (left graphs), the numerical urea concentration is higher than the analytical urea concentration in the first part of the domain. The reason is that not so much urea is consumed due to the low concentration of fixated bacteria, which is a consequence of the low K_{bac}-value.

The analytical solution of the calcium carbonate concentration is not similar to the numerical solution for $K_{\text{bac}} = 10\ \text{h}^{-1}$. The reason is that the analytical solution has been constructed for an infinite K_{bac}-value. The numerical solution for the high K_{bac}-value, $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$, is similar to the analytical solution, except close to the inlet. Although the graphs are similar, the difference is not equal to zero. This time, the reason is not the difference in K_{bac}-value but the numerical

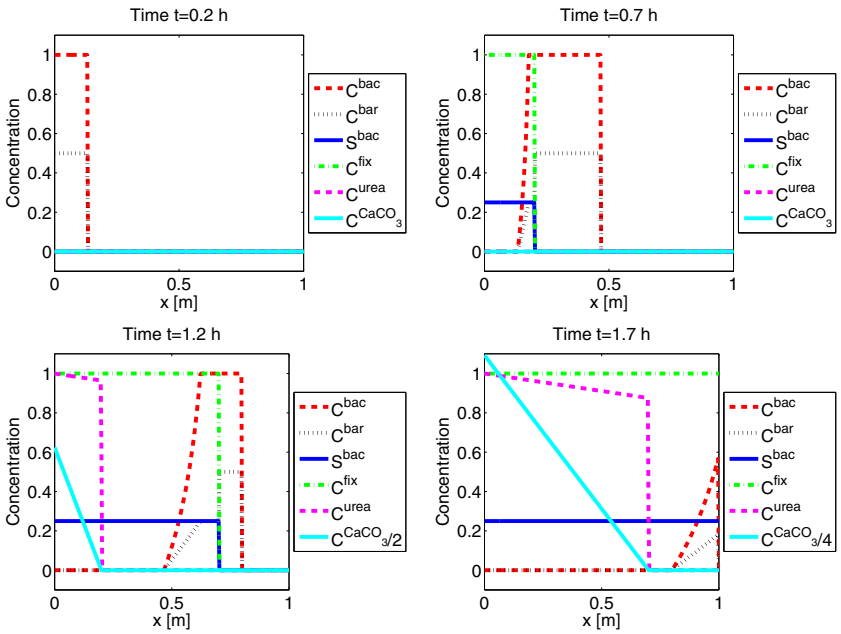

Fig. 4 Analytical solution for the concentration of suspended, temporarily adsorbed and fixated bacteria and the concentration of fixation fluid, urea and $CaCO_3$ as a function of location at several times ($t = 0.2$ h, $t = 0.7$ h, $t = 1.2$ h and $t = 1.7$ h)

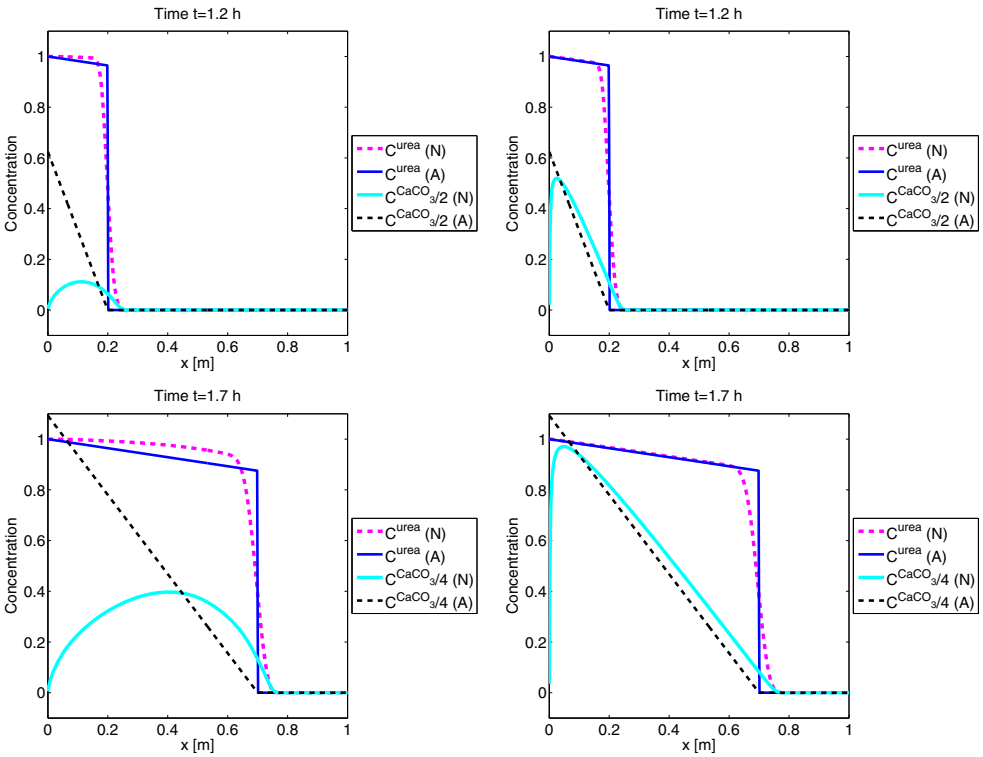

Fig. 5 Numerical and analytical solution of the urea and calcium carbonate concentration at times $t = 1.2$ h and $t = 1.7$ h for $K_{\text{bac}} = 10\ \text{h}^{-1}$ (*left graphs*) and $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$ (*right graphs*). The numerical solutions are marked with N and the analytical solutions are marked with A. In both the analytical and numerical solutions, dispersion and diffusion are neglected, as well as the effect of the reaction on the porosity and flow rate

diffusion. Due to the numerical diffusion, the numerical solution to the urea concentration has a less steep front and the urea penetrates a little further in the column. Although the concentration is small, reaction (3) can happen and calcium carbonate is formed. Hence, the numerical solution to the calcium carbonate concentration is somewhat larger that the analytical solution.

In Fig. 5, rate (14) has been taken as a reaction rate for reaction (3). The urea concentration is related to the reaction rate via a Monod equation. This article also provides an analytical solution for a reaction rate that is linear in the urea concentration, Eq. 45. For this case, the analytical solutions are given in Eqs. 47 and 48. Figure 6 shows the comparison between the analytical and numerical solution for both reaction rates at time $t = 1.7$ h. In the left graph, the urea concentration is related to the hydrolysis reaction rate via a Monod equation (see Eq. 14). In the right graph, the reaction rate is linear in the urea concentration (see Eq. 45).

The left graph of Fig. 6 is equal to the bottom right graph of Fig. 5. In both graphs of Fig. 6, the numerical solution to the urea concentration corresponds well

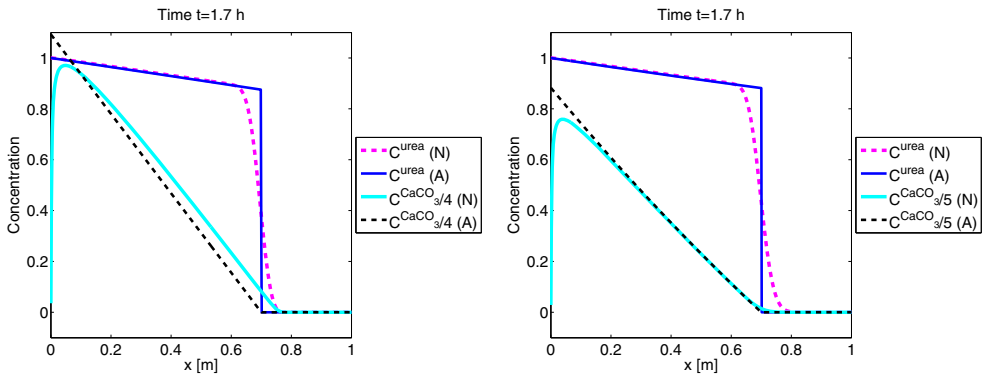

Fig. 6 Numerical and analytical solution of the urea and calcium carbonate concentration at time $t = 1.7$ h for $K_{\text{bac}} = 1{,}000\ \text{h}^{-1}$. The numerical solutions are marked with N and the analytical solutions are marked with A. *Left graph* the urea concentration is related to the hydrolysis reaction rate via a Monod Eq. 14. *Right graph* the hydrolysis reaction rate is linear in the urea concentration, see Eq. 45

with the analytical one, as well for rate (14) (left graph) as for rate (45) (right graph).

In the right graph, which is calculated with rate (45), the numerical solution for the calcium carbonate concentration is closer to the analytical solution than in the left graph. Due to numerical diffusion, the numerical urea concentration approximates zero at a location further away from the inlet than the analytical urea concentration does. Since for a small urea concentration

$$r_{\mathrm{hp}} = v_{\max}\left(C^{\mathrm{bac}} + \overline{C}^{\mathrm{bac}} + S^{\mathrm{bac}}\right)\frac{C^{\mathrm{urea}}}{K_{\mathrm{m,urea}} + C^{\mathrm{urea}}} \sim v_{\max}\left(C^{\mathrm{bac}} + \overline{C}^{\mathrm{bac}} + S^{\mathrm{bac}}\right)\frac{C^{\mathrm{urea}}}{K_{\mathrm{m,urea}}}, \tag{50}$$

and since $K_{\mathrm{m,urea}} = 0.01$ (see Table 2), the rate as determined by Eq. 14 is 100 times larger as the rate as computed by Eq. 45. This implies that the numerical calcium carbonate concentration has a larger increase in the case of use of Eq. 14. Therefore, the difference between the analytical and numerical solution is larger if Eq. 14 is used.

5 Discussion and conclusions

In this article, the model for the placement of bacteria and the model for the hydrolysis of urea and the precipitation of calcium carbonate are coupled. These two models were introduced and discussed in [6, 7]. We shortly mention some of the discussion points.

It is crucial to find a good relation for ratio β. This ratio determines the amount of fixated bacteria. These bacteria eventually provide the production of calcium carbonate, which is the aim of Biogrout. Laboratory experiments need to be carried out to find such a relation. Whether the Langmuir isotherm is a good choice for an adsorption isotherm, as well as the values of the various constants in this isotherm, should also follow from these experiments. Furthermore, experiments need to be done to find the right values for the adsorption, desorption and fixation reaction constants. Another important effect that should be investigated is the possible wash-out of bacteria as a result of a high pore-water velocity. This wash-out violates the present model assumption that fixated bacteria will always stay sticked to the sand grains.

The precipitation model is based on the biochemical reaction Eq. 3. In reality, this reaction happens in several steps. Some of these steps are equilibrium reactions that depend on the pH. The differential equation for the calcium carbonate concentration does not contain a transport term as it has been assumed that calcium carbonate precipitates locally and will not be transported. Calcium carbonate can precipitate in several ways. It can attach to sand grains but can also form crystals. Especially when these crystals are small, they can be transported before they will stick in the pore throats. The retardation of urea, calcium, ammonium and fixation fluid is neglected for the moment. Especially when the particles are charged, there can be retardation.

In this article, the two models are coupled. It is possible that the parameters in both models will influence each other. For example, the bacteria can be encapsulated by the calcium carbonate. Then, the urea can no longer reach these bacteria and therefore, these bacteria can not contribute to the hydrolysis of urea any more.

In the biochemical reaction rate r_{hp} (Eq. 14), the concentration of bacteria is used, multiplied by the maximal bacterial activity $v_{\max}$. What actually provides the hydrolysis of urea are some enzymes in the bacteria. These enzymes can be released from the bacteria and flow with the water. Consequently, the activity of the bacteria decreases. It would be better to use the activity of the bacteria in the reaction rate (14). From experiments, it is known that the reaction rate decreases [9]. For a good estimation of the hydrolysis reaction rate, it is necessary to know what the reasons are and how they influence the rate.

We succeeded in coupling the model for the placement of bacteria with the precipitation model. From the numerical simulations with the coupled model, it can be concluded that, when the adsorption, desorption and fixation processes are fast and hence the K_{bac}-value is large, the calcium carbonate concentration has its maximum close to the injection point. When the K_{bac}-value is small, less calcium carbonate is formed, and its maximum lies further away from the injection point.

Furthermore, a Newton iteration to calculate the concentrations of bacteria costs nine times as much CPU time as a Newton iteration to calculate the concentration of urea.

A high reaction constant leads to a homogeneous bacterial distribution, at least for the first part of the domain, except for the region around the injection point. Furthermore, the calcium carbonate content that is calculated from the coupled model is similar to the calcium carbonate that is calculated from the model where a homogeneous bacterial distribution is assumed. A low reaction rate constant, corresponding to slow adsorption, desorption and fixation processes, leads to a non-homogeneous bacterial distribution and hence to a different calcium carbonate profile. High

reaction rates, however, do not guarantee that the bacterial distribution will be homogeneous. This can also be seen from the analytical solution for the instantaneous equilibrium, Eqs. 25, 31 and 32. The reason is that the concentration of fixated bacteria does not depend on the length of the domain. Since only a finite amount of bacteria is injected, the domain can be chosen so large that only bacteria close to the inlet are fixated and that there are no bacteria in the rest of the domain.

An analytical solution has been constructed for the coupled model for the case that dispersion, diffusion, decrease of the porosity and the change of liquid volume are neglected and the concentrations of suspended, adsorbed and fixated bacteria are in equilibrium. Although these phenomena are neglected, the analytical solution of the calcium carbonate concentration is similar to the numerical solution with a high K_{bac}-value (see Figs. 2 and 4). Hence, in real life applications that can be modelled through a 1-D model, the analytical solution can be used as a first estimate for engineering purposes. If the sorption and fixation processes are close to equilibrium, the analytical solution might be as good as the numerical solution since the numerical solution also includes some error as a result of the error in the estimation of the various parameters.

We further think that the models can be extended with the following features:

- The model for the placement of bacteria contains the most important phenomena of the transport of bacteria: advection, dispersion, adsorption, desorption and fixation. Other phenomena, like decay, growth and systematic motion of bacteria can be included.
- The fixation of bacteria will cause a decrease in porosity and permeability. This has not yet been added to the model.
- As a function for the ratio β, a Monod equation is used. In this paper, the ratio is only a function of the fixation fluid, whereas it may also depend on the properties of the microorganisms, the pH and the porous medium.
- The (saturated) flow can be extended to unsaturated flow to be able to model also the unsaturated zones.

Appendix: List of symbols

C^{bac} normalised concentration of suspended bacteria, [1];

$\overline{C}^{\text{bac}}$ normalised concentration of temporarily adsorbed bacteria, [1];

S^{bac} normalised concentration of fixated bacteria, [1];

C^i concentration of fixation fluid, urea, Ca^{2+} and NH_4^+ (i = fix, urea, Ca^{2+}, NH_4^+), [kmol/m^3];

C^{CaCO_3} concentration of calcium carbonate molecules, [kg/m^3];

θ porosity, [1];

θ_0 initial porosity, [1];

$\mathbf{D_i}$ hydrodynamic dispersion tensor of bacteria, fixation fluid, urea, Ca^{2+} and NH_4^+ (i = bac, fix, urea, Ca^{2+}, NH_4^+), [m^2/s];

α_i dispersion length of bacteria, fixation fluid, urea, Ca^{2+} and NH_4^+ (i = bac, fix, urea, Ca^{2+}, NH_4^+), [m];

$D_{m,i}$ diffusion coefficient of bacteria, fixation fluid, urea, Ca^{2+} and NH_4^+ (i = bac, fix, urea, Ca^{2+}, NH_4^+), [m^2/s];

r_i reaction rate of adsorption, desorption and fixation (i = ads, des, fix), [1/s];

r_{hp} reaction rate of the hydrolysis of urea and precipitation of $CaCO_3$, [kmol/m^3/s];

k_i reaction rate constant of adsorption, desorption and fixation (i = ads, des, fix), [1/s];

$v_{\max}$ reaction rate constant of the hydrolysis of urea and precipitation of $CaCO_3$, [kmol/m^3/s];

φ adsorption isotherm, [1];

K_{l} Langmuir constant, [1];

$\overline{C}_{\max}$ maximum adsorption capacity, [1];

β factor that describes which part of the adsorbed bacteria are fixated, [1];

β_0 factor that describes which part of the adsorbed bacteria are fixated, [1];

q_i Darcy flow velocity in the respective coordinate directions (i = x, y, z), [m/s];

v_i pore water flow velocity in the respective coordinate directions (i = x, y, z), [m/s];

K constant in the differential equation for the flow, [m^3/kmol];

$1 - V_s$ volume that disappears per number of converted particles, [m^3/kmol];

$K_{\text{m,urea}}$ saturation constant of urea and calcium chloride, [kmol/m^3];

$K_{\text{m,fix}}$ saturation constant of fixation fluid, [kmol/m^3];

m_{CaCO_3} molecular mass of calcium carbonate, [kg/kmol];

ρ_{CaCO_3} density of calcium carbonate, [kg/m^3];

k_i intrinsic permeability in the respective coordinate directions (i = x, y, z), [m^2];

d_m mean particle size of the subsurface medium, [m];
μ dynamic viscosity of the fluid, [Pa·s];
p pressure, [Pa];
g gravitation constant, [m/s^2];
ρ_l density of the fluid, [kg/m^3];
T_1 time at which the injection of bacteria is stopped and the injection of fixation fluid is started, [h];
T_2 time at which the injection of fixation fluid is stopped and the injection of precipitation fluid is started, [h];
T_3 time at which the shock speed of the bacteria changes, [h];
T_{end} time at which the injection of precipitation fluid is stopped, [h].

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