

CHAPTER IV

NUMERICAL IMPLEMENTATION

The objective of this chapter is to describe the numerical implementation of CLOG. The task consisted of integrating two existing codes and adding some specific modules. The codes are:

- CODE_BRIGHT (OLIVELLA & GARCÍA-MOLINA, 1996). It is the acronym of *CO*upled *DE*formation, *transport of BR*ine, *Gas and Heat Transport*.
- RETRASO (SAALTINK ET AL., 1997), acronym of *RE*active *TR*ansport of *SO*lutes.

The specific clogging modules included:

- (1) Modifying the input of data
- (2) Generalising kinetic subroutines to account for suspended particles and bacteria. *Deposited particles* and *attached bacteria*, together with the *chemical or true minerals* that were already treated by RETRASO, form the so-called generic minerals.
- (3) Updating porosity, whose changes are driven by variations in the concentration of the generic minerals.
- (4) Updating intrinsic permeability through the Kozeny-Carman equation, which related current permeability and porosity to the initial values.

The general layout of the code is shown in Figure 4.1. Next sections are devoted to describing the capabilities of the code and its structure. This description is based on PÉREZ-PARICIO ET AL. (2000).

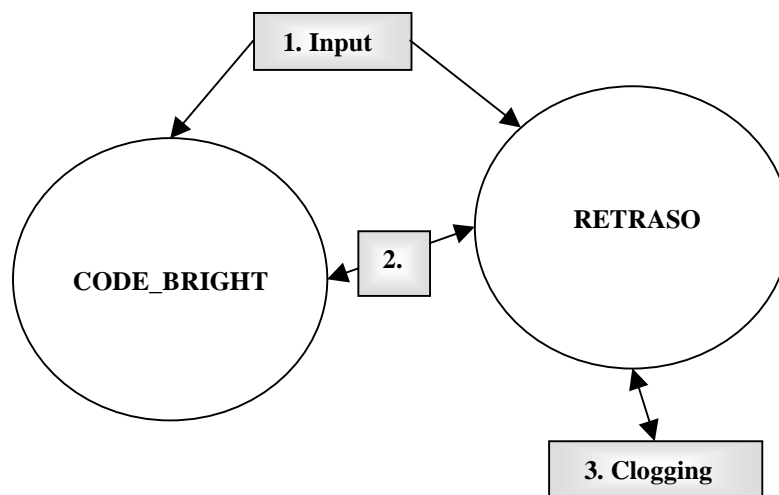


Figure 4.1. Schematic diagram of CLOG where the main modules are shown. Basic changes needed to obtain this code involved three areas:
 1=changes at the input level due to adding clogging processes,
 2=integration between the two existing codes, CODE_BRIGHT and RETRASO,
 3=clogging specific subroutines.

4.1. MULTIPHASE FLOW: CODE_BRIGHT

CODE_BRIGHT, acronym of COupled DEformation with BRIne, Gas and Heat Transport, was developed at the *Technical University of Catalonia (UPC)* with the original aim of dealing with saline environments. However, it has evolved towards a generic coupled deformation, multiphase flow model. CODE_BRIGHT is a 3-D finite element code for unsaturated flow of both liquid and gas phases (OLIVELLA ET AL., 1994). Table 4.1. summarises the basic balances and constituents.

Table 4.1. Summary of CODE_BRIGHT (after OLIVELLA ET AL., 1994).

The Problem			
Phase / Species	Water	Air	Halite (solid)
Liquid	x	x	x
Gas	x	x	-
Solid	x	-	x
Equation	Balance	State variables	
I	Mass of water	Liquid pressure	
II	Mass of air	Gas pressure	
III	Mass of halite	Porosity	
IV	Mass of water in inclusions	Mass fraction of water in solid	
V	Momentum	Solid velocity	
VI	Internal energy	Temperature	
Constitutive equations			
Darcy's law		Advective flux of liquid and gas	
Fick's law		Non-advective flux of vapour and salt	
Inclusion migration law		Non-advective flux of inclusions	
Fourier's law		Conductive heat flux	
Retention curve		Liquid degree of saturation	
Mechanic constitutive model		Stress tensor	
Phase density		Liquid density	
Liquid viscosity		Liquid viscosity	
Gases law		Gas density	
Equilibrium restrictions			
Solubility		Mass fraction of dissolved salt	
Henry's law		Mass fraction of dissolved air	
Psychrometric law		Mass fraction of vapour	

As for the numerical solution of the system of equations, CODE_BRIGHT adopts a Newton-Raphson scheme to update the state variables (Table 4.1.) at each node and time step. After the spatial and temporal discretization, it is possible to write the following system of equations:

$$\mathbf{r}(\mathbf{X}^{k+1}) = \frac{\mathbf{d}(\mathbf{X}^{k+1}) - \mathbf{d}(\mathbf{X}^k)}{\Delta t^k} + \mathbf{A}(\mathbf{X}^{k+\varepsilon}) \mathbf{X}^{k+\theta} + \mathbf{b}(\mathbf{X}^{k+\varepsilon}, \mathbf{X}^{k+\theta}) = \mathbf{0} \quad [4.1.]$$

where \mathbf{r} is the vector of residuals at time $k+1$ that depends on the vector of state variables, \mathbf{X} , both at times t^k (previous step) and t^{k+1} ; \mathbf{d} includes the storage terms, \mathbf{A} the conductance terms, and \mathbf{b} is the sink/source term plus boundary conditions. Two intermediate points, $t^{k+\varepsilon}$ and $t^{k+\theta}$, are taken

between the initial and the final times within a given time step to evaluate the conductance and boundary terms and the vector of unknowns.

The Newton-Raphson scheme for this non-linear set of equations yields:

$$\frac{\partial \mathbf{r}(\mathbf{X}^{k+1})}{\partial \mathbf{X}^{k+1}} (\mathbf{X}^{k+1, it+1} - \mathbf{X}^{k+1, it}) = -\mathbf{r}(\mathbf{X}^{k+1, it}) \quad [4.2.]$$

where *it* indicates iteration during the calculation of the residuals at time t^{k+1} .

4.2. REACTIVE TRANSPORT: RETRASO

RETRASO, acronym of REactive TRAnsport of SOLutes, was also developed by the *Technical University of Catalonia* (SAALTINK ET AL., 1997) in order to cope with equilibrium and kinetic reactions for non-conservative dissolved species. This was originally a 2-D finite element code for saturated conditions, as represented in Table 4.2.

Table 4.2. Summary of chemical reactions as originally considered by RETRASO. (E=equilibrium; K=kinetics).

Reaction type	Type	Approach	Comments / Models
Homogeneous	-Aqueous complexation	E	
	-Acid-base	E	
	-Redox	E	
Heterogeneous	-Precipitation/dissolution	E, K	
	-Gas dissolution	E	
Others	a) Electrostatic	E	-surface electric potential
			-capacitance models
	-Adsorption	E	-triple layer model
			-diffuse layer model
b) Non-electrostatic	E	-constant capacitance	
		-linear isotherm	
-Cation exchange	E	-non-linear (Freundlich)	
		-Langmuir model	
			-Gaines-Thomas convention
			-Gapon convention

RETRASO is based on the direct substitution approach (DSA) to solve the coupled transport processes and chemical reactions. The chemical equations are substituted as a sink/source term in the transport equations, which are solved simultaneously. The solution of these non-linear partial differential equations is carried out by a Newton-Raphson iterative procedure, so that the chemical source term is updated after convergence. DSA is also called one-step and global implicit method.

In principle, DSA needs to solve for $N_{\text{comp}} \times N_{\text{node}}$ equations, that is, the number of components times the number of nodes. However, the mathematical formulation (SAALTINK ET AL., 1998a) allows for a reduction in the number of components (N_{comp}) by using mineral equilibrium reactions

(N_{eq}). By further removing constant activity species, the final system can be expressed as a function of the thermodynamic degrees of freedom or **reduced primary species**. The rest of species depends on the concentrations of the reduced primary species.

The whole systems of equations is omitted here for the sake of simplicity. The numerical formulation implemented in RETRASO requires solving the following final system:

$$\left(\frac{\partial \mathbf{f}_t^{it}}{\partial \mathbf{c}_t} + \frac{\partial \mathbf{f}_t^{it}}{\partial \mathbf{c}_c} \frac{\partial \mathbf{c}_c}{\partial \mathbf{c}_t} \right) \cdot (\mathbf{c}_t^{it+1} - \mathbf{c}_t^{it}) = -\mathbf{f}_t^{it} \quad [4.3.]$$

where \mathbf{c}_t is the vector of $N_{comp}-N_{min}$ concentrations, linked to transport equations; \mathbf{c}_c is the vector of $N_{aqu}+N_{ads}+N_{min}$ (number of equilibrium aqueous reactions, sorption reactions and mineral in equilibrium, respectively); it refers to the iteration number. Vectors \mathbf{f}_t and \mathbf{f}_c are defined as:

$$\mathbf{f}_t = \mathbf{E}\mathbf{U}_{aqu} \frac{\partial \mathbf{c}_{aqu}}{\partial t} + \mathbf{E}\mathbf{U}_{ads} \frac{\partial \mathbf{c}_{ads}}{\partial t} - \mathbf{E}\mathbf{U}_{aqu} \mathbf{L}(\mathbf{c}_{aqu}) - \mathbf{E}\mathbf{U}\mathbf{S}_{kin}^t \mathbf{r}_{kin}(\mathbf{c}_{aqu}) = \mathbf{0} \quad [4.4.]$$

$$\mathbf{f}_c = \begin{bmatrix} \mathbf{S}_{aqu} (\log \mathbf{c}_{pri} + \log \gamma_{pri}) - \log \mathbf{c}_{sec} - \log \gamma_{sec} - \log \mathbf{k}_{aqu} \\ \mathbf{S}_{ads} (\log \mathbf{c}_{pri} + \log \gamma_{pri}) - \log \mathbf{c}_{dis} - \log \gamma_{dis} - \log \mathbf{k}_{dis} \\ \mathbf{S}_{min} (\log \mathbf{c}_{pri} + \log \gamma_{pri}) - \log \mathbf{k}_{min} \end{bmatrix} = \mathbf{0}$$

where subscript *aqu* applies to aqueous equilibrium reactions, *ads* to sorption reactions, *min* to minerals in equilibrium, *pri* to the reduced primary species and *sec* to the secondary species. \mathbf{E} is the elimination matrix used to reduce the number of transport equations from N_{com} to $N_{comp}-N_{min}$; \mathbf{U} is the component matrix that relates total concentrations of components to the vector of concentrations; \mathbf{S} is the stoichiometric matrix and γ the activity coefficients.

In this approach, a set of equations with $N_{comp}-N_{min}$ unknowns has to be solved for all nodes simultaneously by means of a maximum relative convergence criterion. Prior to solving for \mathbf{c}_t^{it+1} , several calculations must be done. First, \mathbf{c}_c^{it+1} is obtained through the application of the Jacobian matrix:

$$\frac{\partial \mathbf{f}_c^{it}}{\partial \mathbf{c}_c} \cdot (\mathbf{c}_c^{it+1} - \mathbf{c}_c^{it}) = -\mathbf{f}_c^{it} \quad [4.5.]$$

Finally, computing the derivative of \mathbf{c}_c with respect to \mathbf{c}_t through the chain's rule allows solving equation [4.3.].

4.3. CLOG AND RCB (OR THE JOINT MODEL)

RCB (BENET ET AL., 1998) is the result of integrating both CODE_BRIGHT and RETRASO. In this context, CLOG is an extension of RCB, where the clogging-related changes have been included. CLOG is a research code that is being applied to synthetic and real data. It will be implemented in the final version of RCB.

In this section we present a brief summary of the modifications required by CLOG in order to incorporate the required calculations. As depicted in Figure 4.1., three main tasks had to be done:

1. Adapting input files and subroutines to accommodate the new options and variables.
2. Linking CODE_BRIGHT and RETRASO.
3. Adding the specific clogging subroutines, i.e. kinetic terms for particles and bacteria plus updating porosity and intrinsic permeability as a result of clogging processes.

The third tier will receive all the attention, as it condenses the essence of CLOG from the point of view of new developments. Next sections compile a brief summary of the numerics of clogging subroutines, based on the previous theoretical presentation. Before doing this, we enclose a succinct sequence of CLOG's calculations for time step $k+1$ (Figure 4.2.):

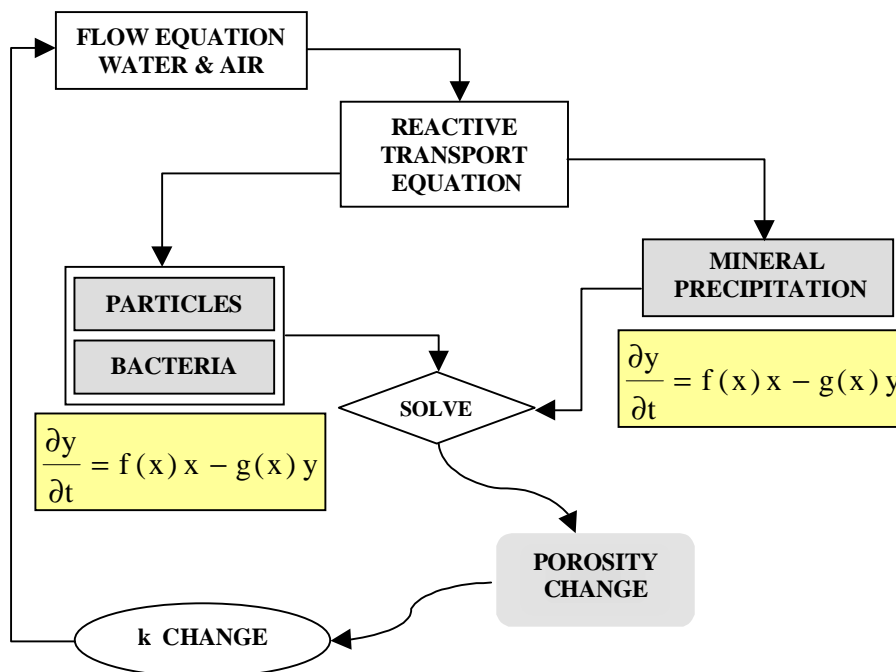


Figure 4.2. Schematic diagram of CLOG calculations.

Liquid and gas flow are solved by the CODE_BRIGHT module, whilst RETRASO dealt with reactive transport of solutes. In order to account for growth of bacteria and attachment of suspended particles, the transport module had to be modified (notice that kinetics is formally identical for physical, biological and chemical clogging, as represented by the equation: y refers to the concentration of generic minerals and x to that of aqueous species). Porosity changes and its impact on intrinsic permeability were also included to obtain the final code.

- A value for porosity at time step k is used for the calculations at time step $k+1$. This means that a time lag is adopted, because porosity is only updated at the end of the reactive transport calculations –when the new concentrations of retained particles, attached bacteria and precipitated minerals are known.

- Liquid pressure (and the gaseous pressure) is calculated by the CODE_BRIGHT module. Darcy's law yields the flow velocity field that is subsequently used by the RETRASO module. Temperature and matrix displacements can be obtained too by CODE_BRIGHT.
- Based on some chemical and mathematical constraints, RETRASO uses a base of primary reduced species at each node that constitutes the degrees of freedom of the system. The rest of species (non-reduced primary species, secondary species, gaseous species, minerals under equilibrium reactions, adsorbed species, retained particles and attached bacteria) can be written as certain functions of the primary reduced species.
- Through the DSA method, the linearised system of equations for reduced primary species is solved.
- If the convergence criterion is met, then all the concentrations are updated. Porosity is accordingly adjusted by means of equation [3.18.], and the intrinsic permeability is also corrected with equation [3.19.]. Finally, a new time step ($k+2$) begins.

4.3.1. THE KINETIC TERM

Equation [3.16.] is a linear first-order partial derivative equation, where the time dependency of $(w_s^m)^{k+1}$ can be approximated by a finite difference approach. The problem, though, consists of the presence of the generic mineral concentration, w_s^m , in the right hand side of the equation –how to weigh this variable in time. The Crank-Nicholson' approach for has been selected to express the concentration of generic mineral as a function of the previous (time step k) and unknown (time step $k+1$) concentrations:

$$(w_s^j)^\theta = \theta(w_s^j)^{k+1} + [1-\theta](w_s^j)^k \quad [4.6.]$$

The value of θ in (4.6.) will determine which scheme is being used. If $\theta=0$, we are assuming an explicit scheme for $(w_s^j)^{k+1}$; on the other hand, $\theta=1$ would imply a fully implicit scheme. With this approximation, it is straightforward to come to the final discretised expression for equation [3.21.] where the concentration of mobile species, w_1^i , is evaluated at time step $k+1$:

$$\frac{D w_s^m}{D t} = \frac{(w_s^m)^{k+1} - (w_s^m)^k}{\Delta t} = \left\{ \frac{1}{1 + \Psi_d \theta \Delta t} \right\} \left[\Psi_a - \Psi_d \cdot (w_s^m)^\theta \right] \quad [4.7.]$$

In this equation, the dependency of the generalised attachment and detachment terms (Ψ_a and Ψ_d , respectively) on the mobile species has been omitted to simplify the notation.

All the Jacobian components or derivatives can be directly obtained from Equation [4.7.], by deriving that expression with regards to the concentrations of the reduced primary species (SAALTINK ET AL., 1998). After rearranging all the terms, we come to the following expression:

$$J_{mj} = \frac{\partial r_{kin}^m}{\partial w_1^j} = \left\{ \frac{1}{1 + \Psi_d \theta \Delta t} \right\} \left[\frac{\partial \Psi_a}{\partial w_1^j} + \frac{\partial \Psi_d}{\partial w_1^j} \left(- (w_s^m)^k - \theta \Delta t r_{kin}^m \right) \right] \quad (4.8.)$$

The derivatives of Ψ_a and Ψ_d are quite complicated from the point of view of calculus, but they do not present serious conceptual inconveniences.

4.3.2. POROSITY UPDATE

Porosity is updated, at the end of a given time step, following equation [3.23.]. The resulting discretized formulae is:

$$\phi^{k+1} = \phi^k - \sum_m \Phi_m \left[(w_s^m)^{k+1} - (w_s^m)^k \right] \quad [4.8.]$$

4.3.3. INTRINSIC PERMEABILITY UPDATE

Prior to CODE_BRIGHT calculations, the intrinsic permeability tensor is adjusted with the use of equation [3.24.]. For each component of this tensor, the discretised equation is as follows:

$$k_i^{k+1} = k_{0i}^k \frac{(\phi^{k+1})^3}{(1 - \phi^{k+1})^2} \frac{(1 - \phi_0)^2}{\phi_0^3} \quad [4.9.]$$

where $i=1,N$ and N refers to the number of components of the tensor. As usual, the superscript k refers to the time step.

