

# PREDICTION OF MINERAL DIAGENESIS BY NUMERICAL MODELING

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## Summary

Diagenetic transformations in sedimentary basins result from the influence of several parameters. The governing processes can be coupled in a complex way. Multi-component reactive transport modeling is a way to better understand such a complexity. With simplification assumptions dimensionless numbers can be defined, which help in appraising the dominant mechanisms. Applications of a water-rock interaction software are shown in the context of subarkosic sandstone diagenesis. Problems relevant to clay minerals, silicification and shale / sand transfers are discussed.

## Introduction

Throughout its geological history a sediment experiences various situations of diagenesis, during which some minerals dissolve whereas other minerals precipitate. As diagenesis can impact considerably on the quality of the rock (as an aquifer, as a reservoir, as a mineral deposit, as a natural barrier for waste repository, *etc*), it is of economical interest to try to predict its effects accurately. Initial mineralogical composition, temperature evolution, hydrodynamism, and mechanical stress are the basic factors controlling the diagenetic transformations. These parameters act in the framework of processes that can be coupled in a complex way, difficult to appraise using only the analytical data sets obtained from the samples of a case study (petrographical, mineralogical, petrophysical and geochemical data). The objective of any deterministic geochemical modeling is to integrate such complex processes in numerical codes that can help to understand the data available from the minerals, and to derive guidelines for predicting the diagenetic heterogeneities in places where no information is available.

This presentation addresses two orders of questions. First, it gives insights on the way to select the important mechanisms to integrate in the modeling approach for a type of problem. Secondly, it provides a series of examples where the contribution of modeling can be illustrated. Because of authors' experience these examples are directly relevant to the quality of siliciclastic petroleum reservoirs.

To represent diagenetic phenomena, modeling choices must be considered with respect to five basic processes : (1) heterogenous reactions, *i.e.*, dissolution or precipitation of several coexisting mineral phases ; (2) reactions in the aqueous phase, between ionic species and complexes ; (3) advective and diffusive-dispersive transport of the aqueous species ; (4) feed-back of the mineral transformations on the petrophysical properties (and consequently on the transport term) ; (5) compaction, *i.e.*, mechanical compaction and pressure-solution, the latter relatively more active with increasing burial depth. Taking into account all these processes result in mathematical systems commonly composed of 10 to 15 partial derivative equations, which are the conservation equations of the considered elements (Si, Al, Ca, K, *etc*) (*e.g.*, Ortoleva, 1987a, 1987b ; Bethke, 1996 ; Lichtner *et al.*, Eds, 1997 ; Giles, 1997 ; Steefel and Van Cappellen, Eds, 1998). To these basic aspects are eventually added : (6) interaction with a multi-

component gas phase ; (7) interaction with a hydrocarbon phase ; (8) ion exchange between water and particular minerals.

Extremely few numerical codes, *e.g.*, CIRF.B developed by Ortoleva *et al* at Indiana University, are able today to integrate a comprehensive calculation of the six basic diagenetic processes in a basin modeling software. Though difficult, it is not an unattainable target of work in terms of numerical algorithms and computing capacities. Nevertheless, any practical use of such a numerical model should mean a tremendous effort for gathering enough appropriate data, able to constrain and to control the simulation results in all types of lithology. The software used for illustrating this presentation, DIAPHORE (Lé Gallo *et al.*, 1998; Brosse *et al.*, 2000), is adapted to the simulation of diagenetic "episodes", short enough to neglect compaction (process 5 listed above). It is preferentially applied to local systems (*e.g.*, a petroleum reservoir, or a part of an aquifer), using as boundary conditions the results of a basin modeling approach. Generally it is also on such local systems that investigations exist to provide the minimum amount of data required for controlling the simulation results.

In the first part of the talk it is examined if can be *a priori* determined, from the main characteristics of a system (dimensions, mineral composition, geological situation), from the type of diagenetic feature that is investigated, and from the scale of interest, a dominant behaviour or mechanism and if, according to that, a way exists to simplify the mathematical problem. Possibilities, but also limitations of such an approach are discussed in the case of complex, multi-components systems.

In the second part of the talk several examples are shown where the application of a modeling water-rock interaction software helps in testing hypotheses formulated, from analytical data, on the diagenetic evolution of reservoir siliciclastic sandstones :

- the formation of clay minerals, such as : (a) kaolinite, in aquifers exposed to efficient water infiltrations due to their structural position in a tilted block ; (b) illite, in more buried and confined contexts ;
- the silicification phenomenon, which is a long-term-process resulting from various mechanisms (dissolution of silicates in the sandstone, short-distance exchanges with other lithologies, pressure-solution, *etc*) ;
- the mineral exchanges likely to occur between shale and sand layers during their burial history, in the context of pure diffusive mineral transfers.

These examples are used to present the most common problems encountered when dealing with diagenetic modeling : choice of a relevant geochemical system ; thermodynamic parameters of clay minerals, which often are solid solutions ; correctly calibrated kinetics for the heterogenous reactions, *i.e.*, kinetics that integrate both experimental data and accurate surface areas ; constraints on the nature and the flow rate of the interstitial water ; mineralogical controls of the modeling results, which commonly show "zones" of mineral composition.

## General equation of the reactive transport phenomenon, and *a priori* analysis

The conservation equation of an aqueous species in the general case of the advective / dispersive transport throughout a porous unidimensional medium ( $x$  coordinate) is the following, written on a representative elementary volume (REV) under the hypotheses of isovolumic transport and of a production rate that depends linearly on concentration (Steefel and Yabusaki, 1996) :

$$\frac{\partial}{\partial t}(\Phi c) + u \frac{\partial c}{\partial x} + \frac{\partial}{\partial x} \left( -D \frac{\partial c}{\partial x} \right) = a \left( 1 - \frac{c}{c_{eq}} \right) \quad (1)$$

where  $\Phi$  represents the porosity of the rock,  $u$  ( $\text{m.s}^{-1}$ ) the Darcy's velocity of the water,  $D$  ( $\text{m}^2.\text{s}^{-1}$ ) the diffusion-dispersion coefficient of the considered species,  $c$  and  $c_{eq}$  ( $\text{mol.m}^{-3}_{\text{water}}$ ) its actual concentration and its concentration at equilibrium, respectively, and  $a$  ( $\text{mol.m}^{-3}_{\text{rock}}.\text{s}^{-1}$ ) a term that expresses its volumic production.

Considering a distance  $L$  that can be the scale of interest for the problem investigated (for instance,  $L = 100$  m in the case of a reservoir layer that a petroleum geologist has to characterize in terms of petrophysical heterogeneity), it is possible to make a variable change replacing  $x$  by  $X=x/L$ ,  $t$  by  $T=ut/\Phi L$  and  $c$  by  $C=c/c_{eq}$ . Doing this the equation (1) becomes, if  $\Phi$  is constant and if  $u$  and  $D$  are uniform :

$$\frac{\partial C}{\partial T} + \frac{\partial C}{\partial X} - \frac{1}{P_e} \frac{\partial^2 C}{\partial X^2} = D_a (1 - C) \quad (2)$$

where two characteristic numbers appear, the Peclet number  $P_e$  :

$$P_e = \frac{uL}{D} \quad (3)$$

and the Damköhler number  $D_a$  :

$$D_a = \frac{aL}{uc_{eq}} \quad (4)$$

These numbers give a way to express, in the general process of water-rock interaction, the relative importance of advection and diffusion (Peclet), and the relative importance of reaction and advection (Damköhler), for the considered length  $L$  (Knapp, 1989). If  $P_e \gg 1$  the diffusion mechanism has negligible effects compared with those of advection. If  $D_a \gg 1$  the kinetic effects are negligible compared to those of advection, in other words thermodynamic equilibrium can be assumed in order to calculate the evolution of the system.

A simple application of this approach is given, for instance by considering quartz precipitation in the 1 km-scale grid elements of a basin numerical model. Typical values of the  $D_a$  number at 25°C and 200°C are calculated with the following hypotheses : (a) water enough diluted to neglect the activity correction, so that the silica activity value is  $10^{-4} \text{ mol.kg}^{-1}_{\text{water}}$  at 25°C and  $10^{-2.43} \text{ mol.kg}^{-1}_{\text{water}}$  at 200°C ; (b) rock composed of pure quartz with spherical grains of 100  $\mu\text{m}$  diameter, with 25 % porosity at 25°C and 10 % porosity at 200°C ; (c) maximum envisaged flow rates of 1  $\text{m.yr}^{-1}$

at 25°C and  $10^{-3} \text{ m.yr}^{-1}$  at 200°C ; (d) quartz dissolution rate value of  $10^{-11}$  at 25°C and  $10^{-6}$  at 200°C (Dove, 1994). It comes out that the  $D_a$  value is 35,521 at 25°C and 114,790 at 200°C, and that under the mentioned hypotheses local equilibrium can be assumed for modeling the water-rock interactions. Nevertheless, another point of view on the Damköhler number in this case will be shown below, introducing kinetics of quartz precipitation deduced from observations of natural systems. When possible, the assumption of local equilibrium presents the great advantage to eliminate the tricky problem related to the badly constrained reactive surface values (process 4 listed above).

The  $P_e$  number concept can be used in the choice of the numerical scheme for space discretization of the mathematical problem. For example if  $P_e$  exceeds 10 it is decided that diffusion is neglected, and in the case of a 1-dimensional system this leads to a strong simplification of the numerical resolution, with correlative high efficiency of the computation.

An important limitation to this type of analysis comes from the expression of the kinetic rate. In usual multi-mineral systems, the production rate of a species is not a direct function of the concentration  $c$  as in equation (1), but is the sum of several terms corresponding to the various minerals that produce this species during dissolution or consume it during precipitation. Such a term is written, in the general case (Madé *et al.*, 1994) :

$$\frac{d[M]}{dt} = \lambda_m . k_m . S_m . f(\text{pH}) \left( 1 - \frac{Q_m}{K_m} \right) \quad (5)$$

where  $[M]$  is the number of moles of mineral  $M$  in the considered REV,  $\lambda_m$  is the number of moles of the species in the mineral  $M$ ,  $k_m$  ( $\text{mol.m}^2.\text{s}^{-1}$ ) is an intrinsic temperature-dependent kinetic coefficient for  $M$ ,  $S_m$  ( $\text{m}^2.\text{m}^{-3}_{\text{mineral}}$ ) is the reactive surface area offered by the mineral  $M$ ,  $f(\text{pH})$  is a function of pH and  $\left( 1 - \frac{Q_m}{K_{eq,m}(T,P)} \right)$  is the departure from equilibrium at  $T$  and  $P$  for

the mineral  $M$ . In some instances a pseudo-Damköhler number can be calculated, but in the general case it is difficult to reach an unambiguous indication and it is preferable to achieve a complete calculation including kinetics. In the following sections are commented applications of the DIAPHORE code.

## First example : precipitation of clay minerals in subarkosic sandstones

Many petrographical and geochemical studies of subarkosic sandstone formations such as those of the Brent Group reservoirs (Middle Jurassic, North Sea) showed two important steps of diagenesis, with the formation of kaolinite followed by the formation of illite (*e.g.*, Bjørlykke *et al.*, 1992 ; Giles *et al.*, 1992 ; Glasmann, 1992 ; Harris, 1992).

The first step occurs at relatively moderate burial depth and temperature, with possible meteoric derived water flow induced by the tectonic structure, determined by the tilting of blocks in a rifting context. It was observed that at least a part of the feldspar and mica content is dissolved, whereas kaolinite and sometimes calcite precipitate. Such a configuration was simulated in a 1-dimensional, 10 km-long reservoir system, along with a temperature gradient from 25 to 75°C. The initial mineralogical

composition is typical of a sub-arkosic sandstone with, additionally to quartz, 8 % K-feldspar, 4 % albite, 4 % anorthite, 6 % Fe,Mg-muscovite (phengite), 1 % calcite and 30 % porosity. The reaction-transport problem was solved with a 11 elements geochemical system : Al, Si, K, Na, Ca, Mg, Fe, Cl, C, H and O. In a basic modeling test the water-flow rate was  $5 \text{ cm.yr}^{-1}$  and the water infiltrating the aquifer at  $25^{\circ}\text{C}$ , in crestal position of the tilted block, was meteoric derived. It was shown, using an approximative Peclet number, that diffusion can be neglected in this case. Simulations showed that anorthite dissolution is a short-term event (order of magnitude 100,000 years), that does not depend on the water movement. Correlative are the precipitation of kaolinite and calcite everywhere in the system. In contrast, the role of water movement is determinant in the dissolution of albite, with the correlative precipitation of additional kaolinite. A reaction front developed, that slowly migrated from the inlet to the outlet of the system. It was only on the long term (more than 10 Ma), or in presence of higher flow rates ( $50 \text{ cm.yr}^{-1}$ ), that K-feldspar and phengite dissolution could be observed, with massive kaolinite and calcite precipitation in the case of a meteoric regime. Complementary simulations are presented. The first one illustrates two opposite effects : a higher water salinity increased the efficiency of albite dissolution at the system inlet (low temperature), but promoted albite stability in the warmest part of the system. The competition between the two effects could be observed. Accordingly, the problem of albite stability in the considered range of temperatures seems difficult to constrain. A second simulation showed that a marine derived water is able : (a) to precipitate kaolinite from the plagioclase, in the same way as the fresh water ; (b) to precipitate some K-feldspar at the expense of phengite. On the long term, or in the presence of very active fluid flow, K-feldspar was stable but not kaolinite. In conclusion, the modeling approach made on this type of system showed the strong influence of the water composition and velocity on the diagenetic transformations and the importance to gain reliable experimental controls on these aspects.

The second step, characterized by illite precipitation, occurs at a relatively deep burial depth, seemingly *ca* 2,500 m in the Brent Group sandstones according to their burial history and to the K-Ar dating of illitic fractions (discussed in Giles, 1997). At such a depth, hydrodynamism is probably much restricted, or even the systems are closed. This situation was simulated using the same type of tilted block, either in a  $70\text{-}120^{\circ}\text{C}$  or in a  $100\text{-}150^{\circ}\text{C}$  temperature range. The initial mineralogical composition was chosen from the results of the step-1 simulations. When the system was closed two episodes of illite formation were successively computed, the first one *ca*  $95^{\circ}\text{C}$  and the second one *ca*  $125^{\circ}\text{C}$ . They correspond to the destabilization of the kaolinite / K-feldspar assemblage, followed by the destabilization of phengite. Complementary simulations are shown to investigate : (a) the role that can play a moderate water flow ( $10^{-3}$  to  $10^{-2} \text{ m.yr}^{-1}$ ) ; (b) the effect of the illite elemental composition (abundance of Fe and Mg in the crystallographic structure). In conclusion, the modeling work in this case showed the importance of choosing representative geochemical systems, and of getting accurate thermodynamic data for addressing correctly the precipitation of clay minerals.

The simulations are discussed by comparison with petrographical observations and geochemical data acquired from core samples of the Hild and the Dunbar fields. At Dunbar, a map could be drawn of the distribution of cements at the field scale, that was

interpreted as a control of the modeling results. As a general result, the geochemical modeling gives new evidence of km-scale transfers of mobile elements (such as Na or K).

### Second example : silicification

Open or closed-system simulations showed that most of the transformations involving dissolution of feldspars and micas, with correlative precipitation of, first, kaolinite, secondly, illite, are silica-prone, and should contribute to precipitate quartz. Nevertheless, the amounts of quartz overgrowth obtained in this way were generally lower than those observed from core samples (*e.g.*, at Dunbar, Hogg *et al*, 1992 ; and Brosse *et al*, 2000). One explanation is that pressure-solution, which was not accounted for in the mentioned modeling tests (DIAPHORE is not including compaction effects), is responsible for the observed additional quartz cement.

The quartz cementation problem illustrates the gap that can exist between experimental data available on the kinetic rates of quartz and the analytical data obtained from natural case studies. Kinetics of quartz dissolution were published by Rimstidt and Barnes (1980) and by Dove (1994). For instance, at  $100^{\circ}\text{C}$ , these authors found rates of  $10^{-7} \text{ mol.m}^{-2}.\text{s}^{-1}$  (Rimstidt) or  $10^{-6} \text{ mol.m}^{-2}.\text{s}^{-1}$  (Dove). In the usual modeling formalisms it is considered, using the "micro-reversibility" principle, that the kinetic rate for precipitation is the reverse of the dissolution rate. From a thorough examination of Jurassic sandstones from the Norwegian shelf Walderhaug (1994a) showed that silicification occurs above a temperature threshold of *ca*  $90^{\circ}\text{C}$ . Many formation waters in reservoirs of the same type are oversaturated with respect to quartz, up to temperatures *ca*  $120^{\circ}\text{C}$  (*e.g.*, Bazin *et al*, 1997). According to these data, quartz does not precipitate at equilibrium before the sandstone reaches a relatively high temperature. Such observations were used to derive "apparent" kinetics for quartz precipitation, *ca*  $10^{-16} \text{ mol.m}^{-2}.\text{s}^{-1}$  at  $100^{\circ}\text{C}$  (Walderhaug, 1994b ; Oelkers *et al*, 1996). Coming back to the calculation of a Damköhler number presented above, it can be shown that the hypothesis of local equilibrium is no more valid at the 1 km scale. In conclusion, the gap between the two ranges of values expresses badly understood phenomena (*e.g.*, quality of the reactive surfaces), which are not easy to constrain in a modeling approach.

### Third example : diffusive mineral transfers between shale and sandstone layers

Very documented petrographical and geochemical studies on sediments from the Gulf Coast basin, particularly from the Frio formation (*e.g.*, Moncure *et al*, 1984 ; Milliken *et al*, 1994 ; Land *et al*, 1997 ; Lynch, 1997), suggest that some mineral transfers occur during burial diagenesis between shale and sand lithologies. Water-rock interaction modeling was a way to investigate this problem. This was done using the DIAPHORE code for simulating purely diffusive mineral exchanges between a sand layer intercalated between two shale layers. The detailed compositions provided by Lynch (1997) were used : quartz, K-feldspar, plagioclase ( $\text{Ab}_{0.7}$ ), albite,  $\text{I-S}_{20\%(\text{Fe,Mg})}$ -illite (Na-rich), (Fe,Mg)-illite, kaolinite and chlorite, with distinct proportions of these mineral phases in the two lithologies. Accordingly, the elements considered for modeling this system were Al, Si, K, Na, Ca, Mg, Fe, Cl, C, H and O. The simulations showed relatively

important amounts of chemical transfers, relative to Ca, Mg, Na and K. Remarkable patterns develop at the boundaries between the two lithologies, that were already noted from petrographical data.

## Conclusions

The numerical modeling of mineral diagenesis is a domain of mathematical geology that made important progresses since 20 years, partly due to many excellent pioneer works, and partly to the increasing capacity of computers. More and more sophisticated processes are integrated into the available codes, and the results are increasingly realistic, provided the input parameters are carefully calibrated on experimental data. Once validated in places where analytical data are available from core samples, these results help to understand the phenomena, and have a predicting value. As for any modeling approach of complex natural processes, the acquisition of a sufficient amount of crucial data is the key point of the modeling success.

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