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# Pressure solution in sandstones: influence of clays and dependence on temperature and stress

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## Abstract

The enhancement of dissolution of quartz under the influence of clays has been recognized in sandstones for many years. It is well known that a grain of quartz in contact with a clay flake dissolves faster than when in contact with another grain of quartz. This phenomenon promotes silica transfer during the diagenesis of sandstones and is responsible of deformation and porosity variations. Here we make an attempt to explain the process of this rock deformation using a pressure solution mechanism.

The model of water film diffusion assumes that matter is dissolved inside the contact between two grains. The resulting solutes are transported to the pore fluid through diffusion along an adsorbed water film. Between two micas, this trapped film is thicker than between two grains of quartz. As a consequence diffusion is easier and the rate of pressure solution faster.

Experiments on pressure solution show that diffusion controls the mechanism at great depth whereas a model based on natural mica indentation indicates that kinetics is the limiting process through the precipitation rate of quartz at low depth, thus temperature is a crucial parameter. There should be a transition between thermally controlled rate and diffusion limited evolution.

**Keywords:** pressure solution; clay; sandstone; compaction; diagenesis

## 1. Introduction

The deformation of rocks by a dissolution-transport-precipitation mechanism, called ‘pressure-solution’ by most authors, has been observed for many years, particularly in sandstones and limestones (Heald, 1955; Weyl, 1959; and others). A simple model of the whole process (Fig. 1) can be described by the three-step mechanism. First, mineral dissolves inside the contact between two grains, then solutes

diffuse to the pore where, at last, precipitation occurs. Thus, the whole mechanism, modifying the grain size and the volume of the pores, is a factor in the deformation of rocks.

In many cases, particularly in sedimentary rocks that are rich in clays, pressure solution is a major process of deformation. When pressure solution is localized, zones of fast dissolution, where most of all the minerals dissolve except clays, appear in the bulk rock and form stylolites (Heald, 1955; Houseknecht, 1987; Carrio-Schaffhauser et al., 1990). Such observations indicate that the rate of pressure solu-

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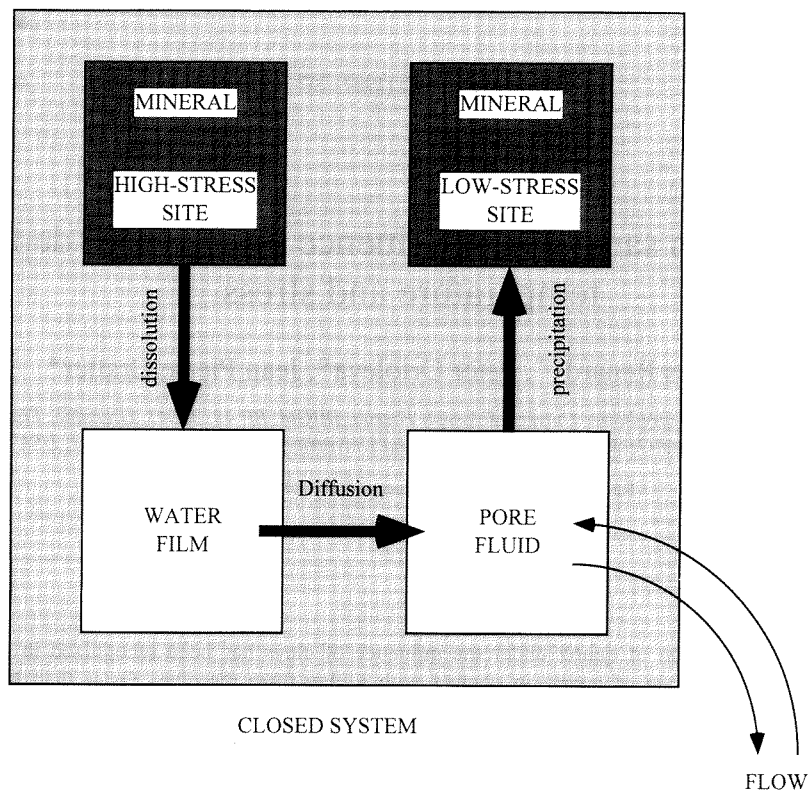


Fig. 1. Deformation by pressure solution through a 3-steps mechanism: dissolution inside the contact between two grains, diffusion along the water film, and precipitation in the pore. The rate of deformation is given by the slowest step. In a case of an open system, the pore fluid can flow and exchange solutes.

tion is higher in the presence of clays (Heald, 1955; Engelder and Marshak, 1985).

## 2. Observations of mica/quartz relationships

Many authors have observed that clays enhance dissolution of quartz (Heald, 1955; Weyl, 1959; Hickman and Evans, 1995). In sandstones, micas can penetrate quartz (Heald, 1955; Bjørkum, 1996) and the flat contact between the two minerals is the result of pressure solution. Some examples are given in Fig. 2, in which it is observed that quartz dissolves at the contact with mica, whose surface seems to be unaffected by the process (Fig. 2a). In the case of Fig. 2b, the mica flake is slightly bent and not broken. Bjørkum (1996) has calculated that the effective stress needed to obtain this curvature was less than 10 bars. This uniaxial 'fiber stress' along the mica is required for bending. In the following discussion,

we have chosen to study this case because the stress along the mica is well determined.

## 3. Derivation of a rate of deformation

Experiments on pressure solution using an indenter (Gratier, 1993a,b) or on aggregates (Rutter, 1976, 1983; Gratier and Guiguet, 1986; Cox and Paterson, 1991; Schutjens, 1991) show that the mechanism of pressure solution of quartz or salt can be limited by the diffusion of matter along the trapped water film between the indenter and the crystal. In such a case, the factor controlling the deformation is the stress which acts both on the driving force of reaction and the water film thickness.

After observations of sandstone thin-sections, Bjørkum (1996) and Oelkers et al. (1996) concluded that, at all temperatures (0–2000 m), the mechanism of compaction/deformation by mass transfer is

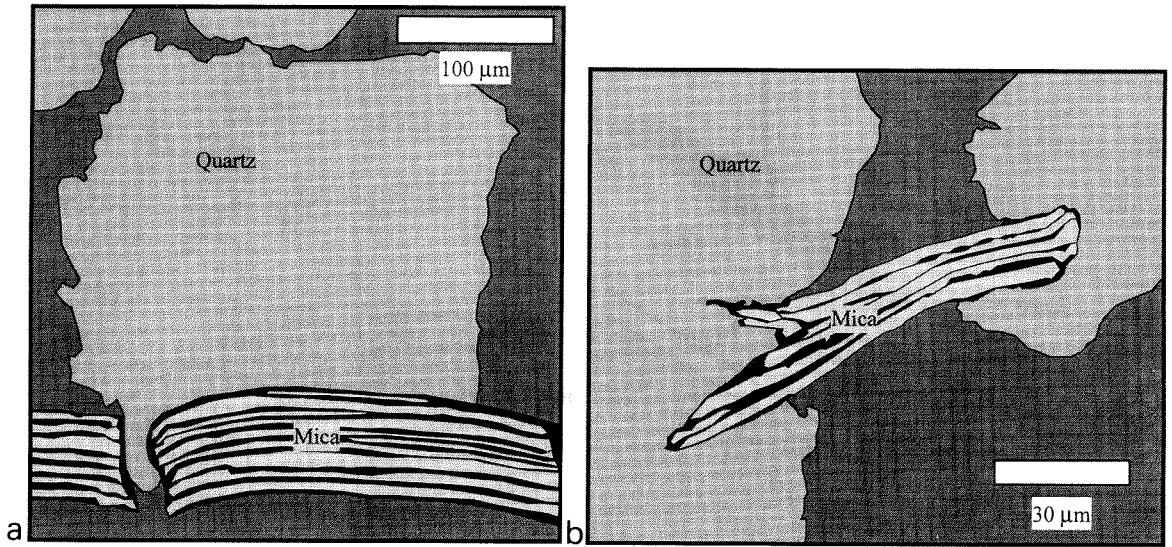


Fig. 2. Thin section of a sandstone. (a) Dissolution at the quartz–mica interface (after Heald, 1955). (b) Detrital mica indenting a grain of quartz. The mica is slightly bent and not fractured, which implies that the stress exerted on this mica flake cannot be greater than a few bars. Note that this feature does not result in quartz overgrowth around the mica (after Bjørkum, 1996).

temperature-controlled through the kinetics of quartz precipitation and that the effect of stress is not important. This comes from the very low kinetics of dissolution and precipitation of quartz at low temperature (Rimstidt and Barnes, 1980). Is it possible to find a mechanism that takes into account these two apparently contradictory observations?

The rate of shortening by solution under the mica flake is estimated using a steady-state approximation:

$$2(L_{\text{cont}} + l_{\text{cont}}) D_{\text{cont}} \Delta \frac{c_{\text{cont}} - c_{\text{pore}}}{l_{\text{cont}}/2} = \frac{A_{\text{cont}}}{V_{\text{qz}} G_{\text{cont}}} \quad (1)$$

where  $L_{\text{cont}}$  and  $l_{\text{cont}}$  (m) are the spatial dimensions of the flake normal to applied load,  $\Delta$  (m) is the thickness of the water film trapped between the mica and the quartz,  $D_{\text{cont}}$  ( $\text{m}^2/\text{s}$ ) the coefficient of diffusion inside the water film,  $c_{\text{cont}}$  and  $c_{\text{pore}}$  are the concentrations ( $\text{mole}/\text{m}^3$ ) of aqueous silica inside the contact and in the pore fluid, respectively,  $A_{\text{cont}} = L_{\text{cont}} l_{\text{cont}}$  is the surface of the actual contact under the flake,  $V_{\text{qz}}$  is the molar volume of quartz ( $\text{m}^3/\text{mole}$ ) and  $G_{\text{cont}}$  ( $\text{m}/\text{s}$ ) the rate at which the flake enters the grain of quartz (see Fig. 3). We make the assumption that dissolution in the contact is faster than diffusion so the water film is always saturated and its concentration of silica is equal to

the solubility under stress. If the activity coefficient of silica is taken to be 1, we have  $c_{\text{cont}} = K_{\text{cont}}$ .

The conservation of matter in a closed system gives a second equation that relates the dissolution under the contact to the precipitation on the free face of the pore.

$$A_{\text{cont}} G_{\text{cont}} + A_{\text{pore}} G_{\text{pore}} = 0 \quad (2)$$

Inside the pore,  $A_{\text{pore}}$  is the surface area of precipitation and  $G_{\text{pore}}$  ( $\text{m}/\text{s}$ ) the rate of precipitation.  $A_{\text{pore}}$  is calculated from the assumption that the surface of precipitation is the surface of the grain in contact with the pore fluid. A model of cubic packed truncated spheres (Dewers and Ortoleva, 1990) gives a good approximation for this surface area, once the grain size and the porosity are known.

Following Rimstidt and Barnes (1980), the rate of precipitation,  $G_{\text{pore}}$ , depends on the saturation of the fluid in contact with the mineral through:

$$G_{\text{pore}} = k_{\text{prec}} \overline{V}_{\text{qz}} \left( 1 - \frac{c_{\text{pore}}}{K_{\text{pore}}} \right) \quad (3)$$

where  $K_{\text{pore}}$  is the equilibrium constant inside the pore and the rate constant  $k_{\text{prec}}$  for precipitation ( $\text{mole m}^{-2} \text{s}^{-1}$ ) is given by Rimstidt and Barnes (1980) as a function of temperature and surface area of reaction.

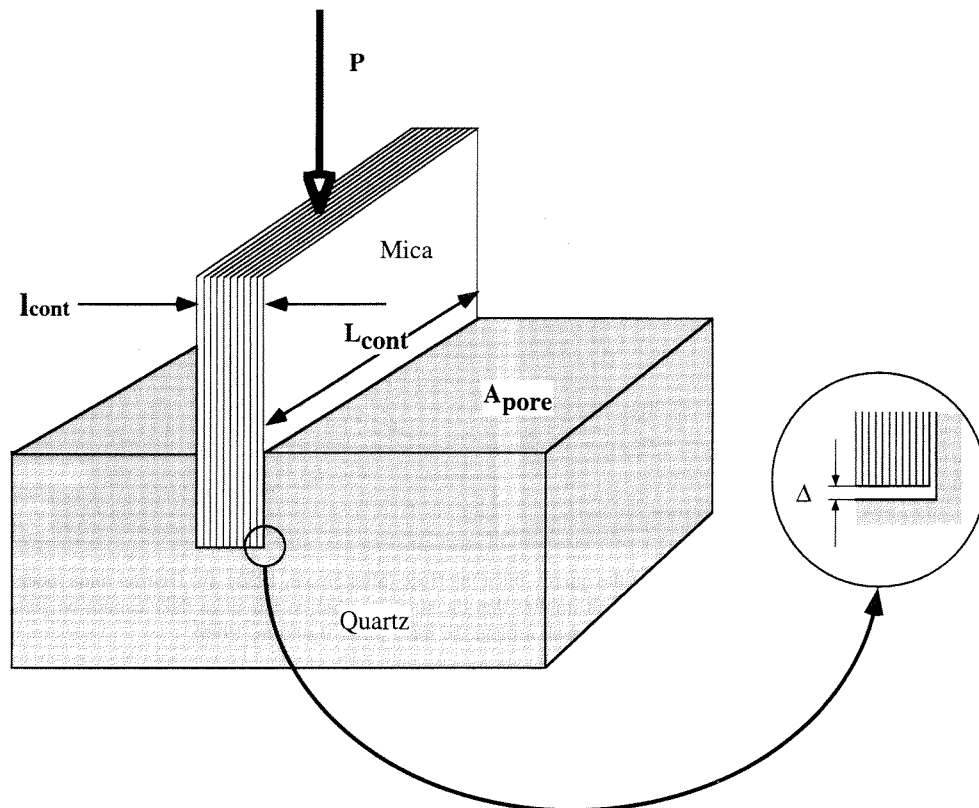


Fig. 3. Schematic representation of a mica indenting a grain of quartz. The variables are used in Eqs. 1–3.

The last three equations take into account the diffusion along the water film between the mica and the grain of quartz and the precipitation on the pore free face. Depending on the temperature, stress and chemistry of the pore fluid, one of these two steps will be slower than the other and will control the rate of deformation.

### 3.1. Existence and thickness of a water film

Experimental evidence shows that a water film can be trapped between minerals (Pashley and Kitchener, 1979; Pashley and Israelachvili, 1984; Horn et al., 1988, 1989). The thickness of this film has been measured or calculated for many minerals and varies from a few ångströms to several nanometers (Peschel and Adlfinger, 1971; Heidug, 1995). In Fig. 4, the thickness of the water film is given as a function of the disjoining pressure, i.e. the amount by which the fluid pressure acting on the fluid–solid interface

exceeds the hydrostatic pressure in the bulk fluid. The disjoining pressure is equivalent to an effective stress. On the mica curve, the oscillatory fluctuations of the water film thickness have been disregarded (Karaborni et al., 1996). The curves clearly indicate that the water film thickness is smaller between two sheets of silica than between two micas. In this study, we will assume that between a mica and a grain of quartz, the water film is like that between two micas.

In the case of clay indentation (in Fig. 2b), the low stress at the edge (less than 10 bar) indicates that the water film ranges from 5 to 20 nm in thickness, a factor which promotes diffusive silica egress along the water film. On the contrary, at depths between 2 and 5 km, the stress is more like 100–300 bar at quartz–clay interfaces. These values imply a 0.5 to 3 nm thick water film. In this case, pressure solution slows down because mobility along the water film decreases.

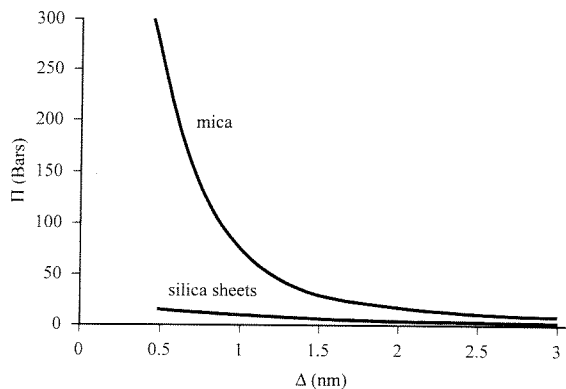


Fig. 4. Disjoining pressure  $\Pi$  for a water of thickness  $\Delta$  confined between parallel silica and mica surfaces (from Heidug, 1995). The disjoining pressure is the difference between the pressure acting on the solid and the fluid pressure; it is similar to an effective stress.

### 3.2. Diffusion along the water film

The diffusivity of a particle in a large volume of liquid is described by the Stokes–Einstein equation (Hunter, 1986):

$$D = \frac{kT}{6\pi\eta a} \quad (4)$$

where  $D$  is the diffusivity ( $\text{m}^2/\text{s}$ ),  $k$  the Boltzmann constant ( $1.38 \times 10^{-23} \text{ m}^3 \text{ Pa s}^{-1}$ ),  $a$  the size of the particle, and  $\eta$  the viscosity of the liquid. Horn et al. (1989) have measured the viscosity of a water film trapped between silica sheets and found a value in the order of magnitude of  $10^{-3} \text{ Pa s}$ . Taking for  $a$  the size of a molecule of water, the diffusivity inside the water film at  $25^\circ\text{C}$  is therefore  $7 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ , an order of magnitude less than diffusion in free water. Other authors have deduced a coefficient of diffusion from pressure solution experiments from which they calculate the product  $D_{\text{cont}} \Delta$  ( $\text{m}^3 \text{ s}^{-1}$ ). They found results from  $10^{-19}$  at  $350^\circ\text{C}$  (Gratier and Guiguet, 1986) to  $10^{-21} \text{ m}^3 \text{ s}^{-1}$  (Rutter, 1976). Assuming a water film thickness of  $5 \text{ \AA}$  in their conditions, the coefficient of diffusion is from  $2 \times 10^{-9}$  to  $2 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ ; these values are several orders of magnitude higher than diffusion in solids (Freer, 1981). For the present study, the water film is thick enough to allow easy transport, thus we have chosen the coefficient of diffusion to be  $10^{-10} \text{ m}^2 \text{ s}^{-1}$  at  $25^\circ\text{C}$  and assumed to follow an Arrhenius law with an activation energy of  $15 \text{ kJ mol}^{-1}$  (Nakashima, 1995).

### 3.3. Equilibrium constants

In the sites where minerals dissolve, the Gibbs free energy is higher than in the sites of precipitation. This increase of free energy is explained by a difference in the state of stress: inside the contact, the stress approaches lithostatic; on the free face of the pore, the stress is due to fluid pressure. Following Gibbs (1878), Kamb (1959), and Paterson (1973), the condition for equilibrium of a fluid of pressure  $P$  in contact with a one-component solid is that the chemical potential  $\mu_{\text{qz}}$  of the component of the solid in the fluid be given by:

$$\mu_{\text{qz}} = \mu_{\text{qz}}^*(T, 0) + P \bar{V}_{\text{qz}} \quad (5)$$

Here  $\mu_{\text{qz}}^*(T, 0)$  is a reference chemical potential at zero stress  $\bar{V}_{\text{qz}}$  is the molar volume of the solid in the stressed state (as a first approximation, we will consider that  $\bar{V}_{\text{qz}}$  is independent of stress).

The chemical reaction of dissolution or precipitation of quartz is taken to be:



and at equilibrium, if we assume that the activity coefficient of aqueous silica and the activities of solid quartz and water are 1, the equilibrium constant equals the solubility of silica and is given by:

$$RT \ln(K^{\text{eq}}) = \mu_{\text{qz}} \quad (6)$$

The last two equations give a relationship for the constant of equilibrium:

$$K_p^{\text{eq}} = K_0(T) \exp\left(\frac{P \bar{V}_{\text{qz}}}{RT}\right) \quad (7)$$

In this equation  $K_0(T)$  is a function of temperature only, as given by Rimstidt and Barnes (1980):

$$\log(K_0) = 1.881 - 0.002028 \times T - \frac{1560}{T} \quad (8)$$

Eq. 7 shows that the solubility of aqueous silica is greater when the stress on the crystal increases since the difference of solubility between the stressed crystal inside the contact and the less stressed pore surface is the driving force of pressure solution.

The driving force for pressure solution is the stress state that determines the supersaturation between the water film and the pore fluid. Therefore effective stress must be involved in the constitutive

law described by Eqs. 1–3 and 7. To simplify our model of mica flake, we choose a constant effective stress of 10 bar at all depths as assumed by Bjørkum (1996) to avoid mica breakage.

### 3.4. Kinetics of quartz precipitation

In Eq. 3  $k_{\text{prec}}$  is the rate constant for precipitation (mole  $\text{m}^{-2} \text{s}^{-1}$ ). Quartz kinetics are very complicated and depend on many factors such as salinity or pH (Dove, 1995) that can induce variations of several orders of magnitude in the rate of precipitation. For example, the presence of a small concentration of  $\text{Na}^+$  can increase the kinetics by an order of magnitude. In our model, we will consider a neutral solution of pure water and use the Rimstidt and Barnes (1980) temperature-dependent formula for  $k_{\text{prec}}$ .

## 4. Effects of stress on deformation

An increase of effective stress on the grains has two consequences. On one hand it decreases the water film thickness, and on the other hand, it increases the driving force for diffusion. The first factor has a negative effect on the pressure solution rate, the second a positive one. It appears that under geological conditions, the second effect can dominate, and permit pressure solution. We verify this by applying our model to an experiment done by Gratier (1993a) in which an indenter was loaded on the surface of a crystal of quartz — see Gratier (1993b) for a similar experiment with crystals of halite. During the experiment, the temperature was 350°C. The fluid, at a pressure of 400 bar, contained a molar solution of NaOH, needed to increase silica solubility, and the stress under the indenter was varied from 1000 to 3000 bar. The main difficulty in doing pressure-solution experiments is the slowness of the processes. Each experiment with an indenter lasts several months. To increase the rate of deformation high effective stress, high temperature, and presence of sodium hydroxyde are necessary (Gratier and Guiguet, 1986). Even if these conditions do not apply at depths less than 5 km, it is the only way to compare our model with experimental results, assuming that the mechanisms are the same in geological conditions and in experimental studies.

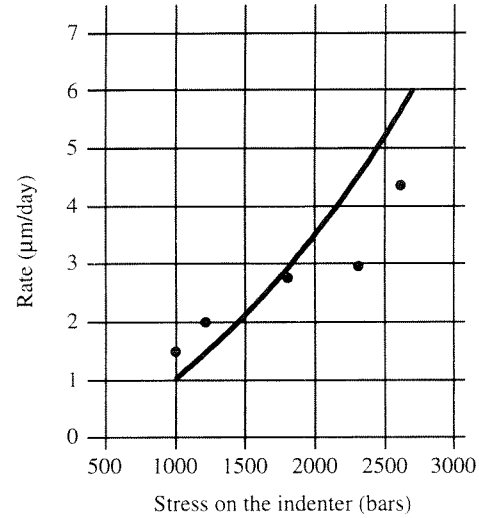


Fig. 5. The effect of stress on the rate of pressure solution. The solid circles are data from experiments by Gratier (1993a) for indentation on a crystal of synthetic quartz. The solid line is our model, see Eqs. 1–8.

In Fig. 5, our model is compared with the experimental results; it is seen that the increasing rate of the indenter inside the quartz with stress is in agreement with the experiment.

## 5. The limiting step of deformation

Using Eqs. 1–3, we may solve for the rate of pressure solution  $G_{\text{cont}}$ . One obtains:

$$G_{\text{cont}} = -\frac{D_{\alpha} A_{\text{pore}} k_{\text{prec}}}{A_{\text{cont}} (D_{\alpha} + A_{\text{pore}} k_{\text{prec}})} (K_{\text{cont}} - K_{\text{pore}}) \quad (9)$$

with

$$D_{\alpha} = 4D_{\text{cont}} \Delta \bar{V}_{\text{qz}} \frac{L_{\text{cont}} + l_{\text{cont}}}{l_{\text{cont}}}$$

The rate  $G_{\text{cont}}$  is a function of the diffusion effect through  $D_{\alpha}$  and the kinetics of precipitation with  $A_{\text{pore}} k_{\text{prec}}$ . The values of these two parameters determine which process limits pressure solution. If:

$$D_{\alpha} \gg A_{\text{pore}} K_{\text{prec}} \quad \text{and}$$

$$G_{\text{cont}} = -\frac{A_{\text{pore}} K_{\text{prec}}}{A_{\text{cont}}} (K_{\text{cont}} - K_{\text{pore}}) \quad (10)$$

the diffusion is relatively fast, the limiting process will be the precipitation of quartz on the free face

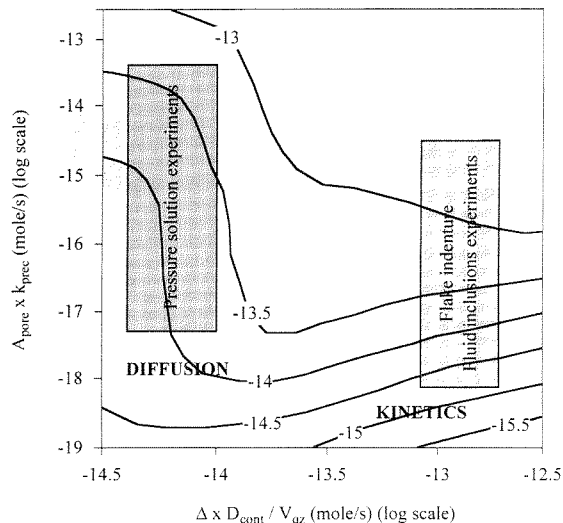


Fig. 6. Transition between the control by precipitation rate kinetics and the diffusion effect on the rate  $G_{\text{cont}}$  of pressure solution (curves). Here,  $\Delta D_{\text{cont}}/V_{\text{qz}}$  represents the 'diffusion effect' and  $A_{\text{pore}}k_{\text{prec}}$  the 'precipitation rate effect'. Both variables have the same units (mol/s). At high coefficient of diffusion or water film thickness,  $G_{\text{cont}}$  (m/s) depends on the surface area of precipitation multiplied by the rate of precipitation. In this case, the deformation is precipitation kinetics-limited (right hand side of the figure). On the contrary, for a low coefficient of diffusion,  $G_{\text{cont}}$  is independent of the precipitation in the pore. In this case, pressure solution rate is limited by diffusion. This reflects the fact that, on the left of the figure,  $G_{\text{cont}}$  is not affected, for example, by the free face contact area. Pressure solution experiments (Gratier and Guiguet, 1986; Cox and Paterson, 1991; Gratier, 1993b) are limited by diffusion, whereas mica indenture (Oelkers et al., 1996) or fluid inclusions deformation (Gratier and Jenatton, 1984) are limited by kinetics. We have represented these two fields in grey.

inside the pore. On the contrary, if:

$$D_{\alpha} \ll A_{\text{pore}}K_{\text{prec}} \quad \text{and}$$

$$G_{\text{cont}} = -\frac{D_{\alpha}}{A_{\text{cont}}}(K_{\text{cont}} - K_{\text{pore}}) \quad (11)$$

diffusion will slow down and limit the rate of pressure solution. Some  $G_{\text{cont}}$  values are calculated in Fig. 6 for different values of the coefficient of diffusion and the surface area of precipitation. If  $\Delta D_{\text{cont}}$  is high enough, the rate of pressure solution is affected by the surface area of precipitation, and the mechanism is limited by the kinetics of precipitation. On the other hand, if  $\Delta D_{\text{cont}}$  is low, the rate of pressure solution is independent of the surface area of

precipitation and the limiting factor is diffusion. For an intermediate value of  $\Delta D_{\text{cont}}$ , both diffusion and kinetics of precipitation affect the pressure solution rate.

As the activation energy for kinetics of precipitation or dissolution are very different (45 and 15 kJ/mole, respectively), kinetics are the limiting process at low depths, and, as a correlation, temperature is a crucial parameter (as found by Bjørkum, 1996).

Kinetics are also the limiting process for a small effective stress because the water film is thick enough and allows the solutes to diffuse easily. On the contrary, at high effective stress, the diffusion becomes the slowest process that limits the whole mechanism of deformation even if the driving force needed for the dissolution reaction is increased. Finally for depths between 0 and 2 km, the precipitation rate is so slow that regardless of the stress, the limiting factor will be the kinetics of quartz dissolution/precipitation.

## 6. Transition between mechanical and chemical compaction

Bjørkum (1996) has observed that this flake indentation configuration (Fig. 2b) occurs at depths greater than 2500 m (no observations were made at shallower depths). We believe that at lesser depths, the rate of pressure solution is not high enough to allow indentation. In other words, if the rate of mechanical compaction is higher than the rate of pressure solution and the increase of stress on the mica indenter is too fast, mica breaks (Bjørkum, 1996).

Different mechanisms allow compaction in the first kilometers of the upper crust: grain rearrangement, elastic compaction, or cataclastic deformation. To estimate the transition between mechanical and pressure solution compaction, we will consider that compaction is isotropic and assume a fixed overall rate of compaction of  $10^{-17}$  m/s. If the rate of flake indentation is slower than the fixed rate, the mica will fracture. Fig. 7 represents the rate of mica indenting quartz with depth, as predicted by our model (Eq. 1 to Eq. 3). These results are in agreement with calculations of Oelkers et al. (1996). Conditions are as those in Fig. 2b with  $l_{\text{cont}} = 5 \mu\text{m}$  and  $L_{\text{cont}} = 15 \mu\text{m}$ . The pressure solution rate,  $G_{\text{cont}}$ ,

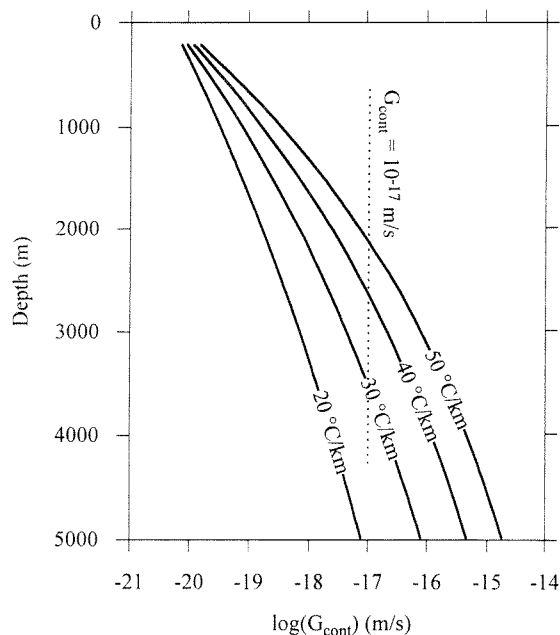


Fig. 7. Temperature effect on pressure solution. The data represent the case of Fig. 2b. Particularly, the effective stress on the flake is 10 bar; this stress does not vary with depth, because if it increases, the flake breaks. Here it is seen that the higher the temperature gradient, the faster the rate of indenting. The fixed rate of compaction of  $10^{-17} \text{ m s}^{-1}$  is represented as a dashed line.

becomes faster than the fixed rate of decrease of pore diameter at around 2500 m for a temperature gradient of  $40^\circ\text{C/km}$ .

The following burial scenario seems to emerge from observations and our model. At depths less than the transition depth (approximately 2500 m), mechanical compaction occurs and flakes accommodate loss of pore volume through breakage, and perhaps rotations. With further burial, below the indentation–breakage transition zone, the increase in kinetics and solubility allows indenter to keep pace with the rate of pore space loss, avoiding breakage. Thus breakage can be observed at all depths; indenter is observed only below the indentation–breakage transition zone (Fig. 8).

## 7. Conclusions

The difference of solubility inside the contact between two grains and in the pore fluid is assumed

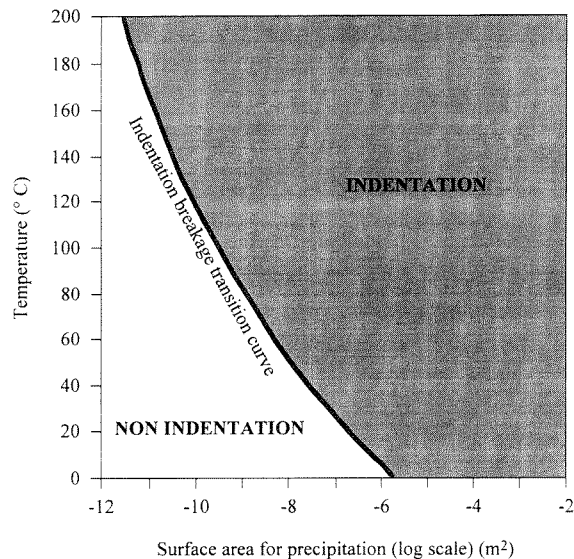


Fig. 8. Criteria for mica indentation as a function of temperature and free surface area. Above the limit curve, mica indentation is possible, below the curve, indentation is not possible. We have used Eqs. 1–3 to calculate the rate of indenting and assumed a critical rate of compaction to be  $10^{-17} \text{ m s}^{-1}$ . At  $25^\circ\text{C}$ , the coefficient of diffusion was chosen to be  $10^{-10} \text{ m}^2 \text{ s}^{-1}$ , the water film thickness 13 nm, and the rate constant for precipitation  $3.8 \times 10^{-10} \text{ mole m}^{-2} \text{ s}^{-1}$ .

to create a gradient of diffusion, allowing solutes to precipitate in the pore. This overall process consists of several steps, the slowest of which yields the rate of deformation.

We explain the contradictory observations that the process can be limited by diffusion or by kinetics, depending on stress and temperature. At great depths, temperature is high and the water film thickness small; the process is limited by diffusion. At low depths, kinetics are the limiting parameters.

In laboratory pressure solution experiments, the effective stress is high and the water film very thin. Therefore, the limiting step is diffusion, especially at high temperature since, in this case, the kinetics of solid/fluid reaction are high.

On the other hand, geological observational evidence suggests that when the effective stress is lower, diffusion becomes faster and the driving force for solute transport lower, therefore the limiting step can be the rate of precipitation on the pore free face. In clay-rich sandstones, especially at low temperature, the kinetics of precipitation of quartz can limit the



rate of deformation. Thus a flake indentation configuration can be obtained. We have shown that clays promote relatively fast mineral dissolution through water film diffusion because a thick water film can be preserved near their surface.

There is a transition domain between mechanical compaction and pressure solution by mica flake indentation which is both dependent on the temperature and surface area for precipitation.

Our model takes into account facts that seem to be in contradiction and show that both clay indentation and clay enhancement of compaction are seen to be natural implications of a water film diffusion model of pressure solution.

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