

PRESSURE SOLUTION AS A MECHANISM FOR CRACK SEALING AROUND FAULTS

Natural and Experimental Evidence

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INTRODUCTION

Several models pertaining to earthquake cycles imply intermittent fluid flow through fault zones (Sibson, 1977; Lachenbruch, 1980; Byerlee, 1990; Rice, 1992; Sleep & Blanpied, 1992). Fluid pressure in a fault zone is expected to change from a high value before an earthquake (leading to a progressive weakening of the fault) to a low value after the event (due to dilatancy associated with the rupture). To account for the low frictional heating and lack of strength of active faults, as inferred from low heat flow measurements (Hickman, 1991), earthquakes are postulated to occur when fluid pressures are close to lithostatic levels. Evidence for high fluid pressures has been found in tectonically active areas. As an example, in the Alps, a comparison between fluid density in horizontal veins (from fluid inclusion studies), temperature (from K/Na ratios), and the thickness of the cover, showed that fluid pressure in those veins was very near lithostatic values at depths ranging from 6 to 10 km (Poty et al., 1974; Bernard et al., 1977). Based on borehole records from gas and oil wells, Hubert & Rubey (1959) and Berry (1973) showed many examples of pore pressures largely above hydrostatic pressures, sometimes even approaching lithostatic pressures at depths ranging from 0.3 to 4 km.

In seismic zones, earthquakes are expected to change the permeability of the country rock and may temporarily reduce fluid pressures to hydrostatic values (Sibson, 1992; Chester et al. 1993; Boullier & Robert, 1992). During the interseismic period, the progressive increase of fluid pressure from near hydrostatic to near lithostatic levels is therefore a crucial parameter which may play an important role in the mechanisms leading to earthquakes (Sibson, 1977; Lachenbruch, 1980; Byerlee, 1990; Rice, 1992; Sleep & Blanpied, 1992). To achieve such high fluid pressures in faults zones, different geodynamic mechanisms have been proposed, including flow of deep fluids at high pressure (Rice, 1992) and compaction within fault gouges (Sleep & Blanpied, 1992). In these models, the very rapid sealing of cracks is postulated to occur in the country rock after the occurrence of an earthquake. The subsequent increase in the fluid pressure is presumed to be only dependent on the rates of these geodynamic processes. This assumption of rapid sealing is based upon experimental results of crack healing (Smith & Evans, 1984; Brantley et al., 1990). However, self-healing may not necessarily be the dominant mechanism associated with natural crack sealing. If the rate of change of permeability in the country rock is not fast relative to the geodynamic rates, then changes in the permeability may be the key factor in controlling the rate of increase in fluid pressure during the interseismic period.

The aim of our study was, therefore, to consider the different mechanisms and the kinetics of crack sealing near active faults. Our study used two approaches: natural observations were made to identify the mechanisms of crack sealing, and secondly, pressure solution experiments allowed

us to investigate the kinetics of the sealing process. The main result of this study is the finding that natural sealing is usually not achieved by a self-healing process. Pressure solution, which is based on the transfer of material from solution cleavages to cracks, appears to be a much more important mechanism for crack sealing. Based on our experimental results, the kinetics of pressure solution are estimated to be rather slow, similar to the recurrence time of earthquakes. The kinetics of crack sealing by pressure solution may thus be important in controlling the increase in fluid pressure around faults, as well as the mechanisms associated with critical failure.

CRACK SEALING MECHANISMS

Mechanical compression can lead to crack closure (Brace, 1972); however, as cracks often open perpendicular to the direction of least effective stress, their closure implies that the effective stress directions have changed either by a decrease in the fluid pressure, or by a change in the state of stress; however, variations in stress directions will not be considered here. The mechanical closure of microcracks may be indicated by the presence of continuous (non-sealed) microcracks. Another mechanism, *plastic flow*, can also result in the reduction of pore volume within a matrix. However, this flow mechanism should be important only at higher temperatures and pressures, such as below the seismic/aseismic transition zone (Kirby, 1983).

Chemical processes can also lead to crack sealing, where different mechanisms may be operative, depending on the aperture of the cracks (Wilkins et al., 1985). For the case of low aperture cracks (0.3 - 3 μm ; Brantley et al., 1990), *self-crack healing* is driven by the lowering of the crack surface free energy (Smith & Evans, 1984). This mechanism may be evidenced by trails of fluid inclusions along the healed crack surfaces (Fig. 1a). On the other hand, cracks may also eventually become self-sealed without any traces of fluid inclusions (B. Evans et al., this volume). Large aperture cracks (10 to 200 μm) generally show evidence for the input of material, as is commonly found in naturally-occurring crack-seal structures (Ramsay, 1980; Cox & Etheridge, 1983; Gratier & Gamond, 1990; Boullier & Robert, 1992). Various mechanisms, which are not always easily distinguishable, can control this process. Processes which can lead to the transfer and deposition of material in a crack are discussed in more detail below.

A change in *pressure* generally has only a small effect on mineral solubility. An exception to this is the case of fluid boiling (Hedenquist et al., 1992), which may be induced by a pressure drop associated with earthquakes (Sibson, 1987). This mechanism for crack sealing is associated with mass transfer by infiltration. When considering the irregular nature of paths associated with mass transfer, only the narrowest parts would be sealed after fluid infiltration and deposition had occurred (Fig. 1b). Since boiling would be associated with rapid fluid displacement, asperities along the fault might induce local drops in the fluid pressure and promote localized deposition in pressure shadow zones. Asymmetric dissolution precipitation near asperities (Fig. 1b') may, therefore, be evidence for this mechanism and indicate the sense of fluid displacement. This type of mechanism has also been evidenced in experimental studies (Lefauchaux, 1988).

Temperature changes can lead to the sealing of large fractures based on a change in mineral solubility. However, this effect is not the same for all minerals. As an example, quartz and calcite display regular and inverse solubility versus temperature dependencies, respectively (Fyfe et al., 1978). For the case of a normal solubility-temperature dependence, a progressive decrease in temperature leads to widespread deposition (Cathles, 1977). The sealing of cracks with this mechanism is also based on the infiltration of fluids into the matrix. However, complete sealing is probably not possible in that after the sealing of the narrowest paths, some isolated void spaces still remain open (Fig. 1c). *Fluid composition and pH* changes can also lead to either deposition (fluid oversaturation) or dissolution (fluid undersaturation) in open veins (Brimhall & Crerar,

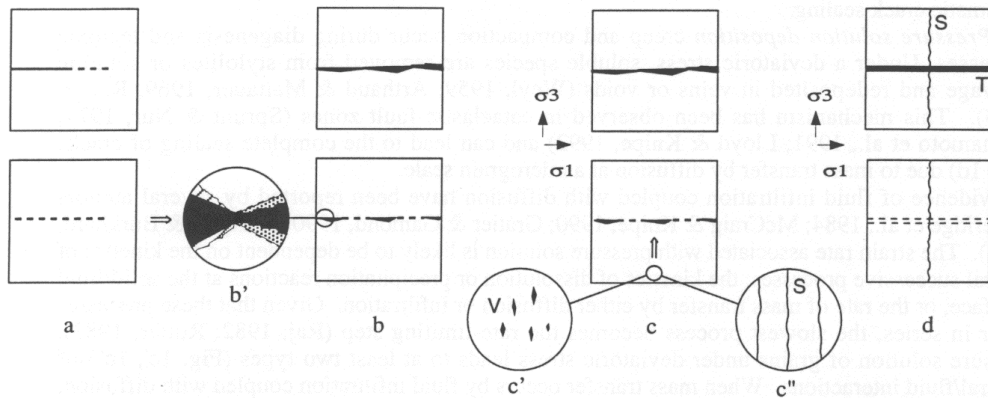


Figure 1: Microstructures expected with various mechanisms of crack sealing, geometry before (top) and after (bottom) sealing: solid is white, fluid is black. (a) Trails of fluid inclusions along the healed crack surfaces with self-healing (no external input of material); (b) and (c) Input of material associated with fluid infiltration, only the narrowest part of the path was sealed; (b') with rapid fluid displacement (double arrow) dissolution occurs on surface subjected to high stress (hatched) and precipitation occurs on surface with low stress (shaded) around the same asperity. With fluid infiltration (double arrow) the kinetics of sealing may be reaction rate controlled (c') with free-face dissolution around voids in matrix (V), or diffusion rate controlled (c'') with water-film diffusion along solution cleavage surfaces (S). (d) Complete sealing with mass transfer by diffusion from solution cleavage (S) to microcrack (T); two trails of fluid inclusion mark the initial limits of the crack.

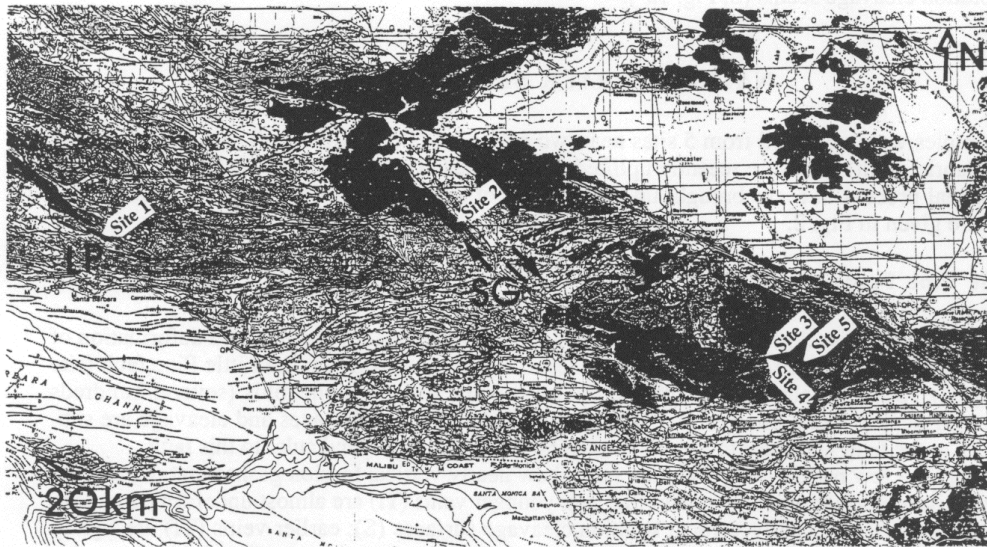


Figure 2: Geological map of the Transverse Range Province (from Jennings, 1977) and location of the sampling sites: site 1 - Little Pine fault (LP) and sites 2,3,4, & 5 - San Gabriel fault (SG).

1987; Wintsh et al, this volume). Special conditions would therefore be required to produce systematic crack sealing.

Pressure solution-deposition creep and compaction occur during diagenesis and tectonic processes. Under a deviatoric stress, soluble species are removed from stylolites or solution cleavage and redeposited in veins or voids (Weyl, 1959; Arthaud & Mattauer, 1969; Rutter, 1983). This mechanism has been observed in cataclastic fault zones (Sprunt & Nur, 1977; Shimamoto et al., 1991; Lloyd & Knipe, 1992) and can lead to the complete sealing of cracks (Fig. 1d) due to mass transfer by diffusion at a micrograin scale.

Evidence of fluid infiltration coupled with diffusion have been reported by several authors (Etheridge et al., 1984; McCraig & Knipe, 1990; Gratier & Gamond, 1990; Marquer & Burkhard, 1992). The strain rate associated with pressure solution is likely to be dependent on the kinetics of several successive processes: the kinetics of dissolution or precipitation reactions at the solid/fluid interface, or the rate of mass transfer by either diffusion or infiltration. Given that these processes occur in series, the slowest process becomes the rate-limiting step (Raj, 1982; Rutter, 1983). Pressure solution of grains under deviatoric stress leads to at least two types (Fig. 1c', 1c'') of mineral/fluid interactions. When mass transfer occurs by fluid infiltration coupled with diffusion, the kinetics of the sealing process is strongly dependent on the mechanism of the fluid/mineral interactions. When dissolution occurs on the free faces of grains, diffusion coefficients are high (for example: 10^{-9} m²/s for silica at 350° (Ildefonse 1980)) and the limiting step becomes the rate of detachment reactions at the solid/fluid interface (Gratier & Jenatton, 1984; Tada & Siever, 1986). In this case the kinetics of sealing may be relatively fast. Evidence for free-face dissolution consists of pores, voids, and etch pits in the matrix (V, Fig. 1c'). The other possibility is that dissolution occurs within a trapped fluid film between grain contacts. Since mass transfer by diffusion is *much slower* than in a free fluid (for example, the diffusion coefficient for silica is 10^{-13} m²/s for water-film diffusion at 350°C; Gratier and Guiguet, 1986), the limiting step is not the rate of reaction at the solid/fluid interface, but rather the rate of mass transfer by water-film diffusion (Rutter, 1983; Gratier & Guiguet, 1986; Spiers & Schutjens, 1990). In this case the kinetics of sealing is always slow (see discussion). Evidence for this mechanism is the presence of solution cleavage seams (S, Fig. 1c'').

NATURAL CRACK SEAL FEATURES

Samples were collected from 5 sites near two major faults in California (Little Pine fault and San Gabriel fault) that were recently uplifted and exposed. This permitted the observation of the crack seal process in rocks brought up from depths of several kilometers. The locations of the sites are given in Fig. 2.

Little Pine Fault

Examples of dissolution features associated with the progressive opening and deformation of tension gashes have been found in Upper Miocene Monterey shales near the Little Pine fault (site 1, Fig. 2). Solution cleavage parallel to the axial plane of large tight folds (several hundreds meters in size) is found kilometers away from the major fault. Folds axes and cleavage are sub-parallel to the fault and attest to intense shortening more or less perpendicular to this fault.

Samples a few meters away from the fault show nearly vertical tension gashes that register the progressive deformation of the rocks (Fig. 3a): younger veins (Tl) are almost undeformed and are perpendicular to the nearly vertical solution cleavage surfaces (S); earlier veins (Tf) are tightly folded and locally dissolved near large zones of solution cleavage; intermediate fold geometries may be found as well, indicating a progressive shortening during the formation of the veins. This

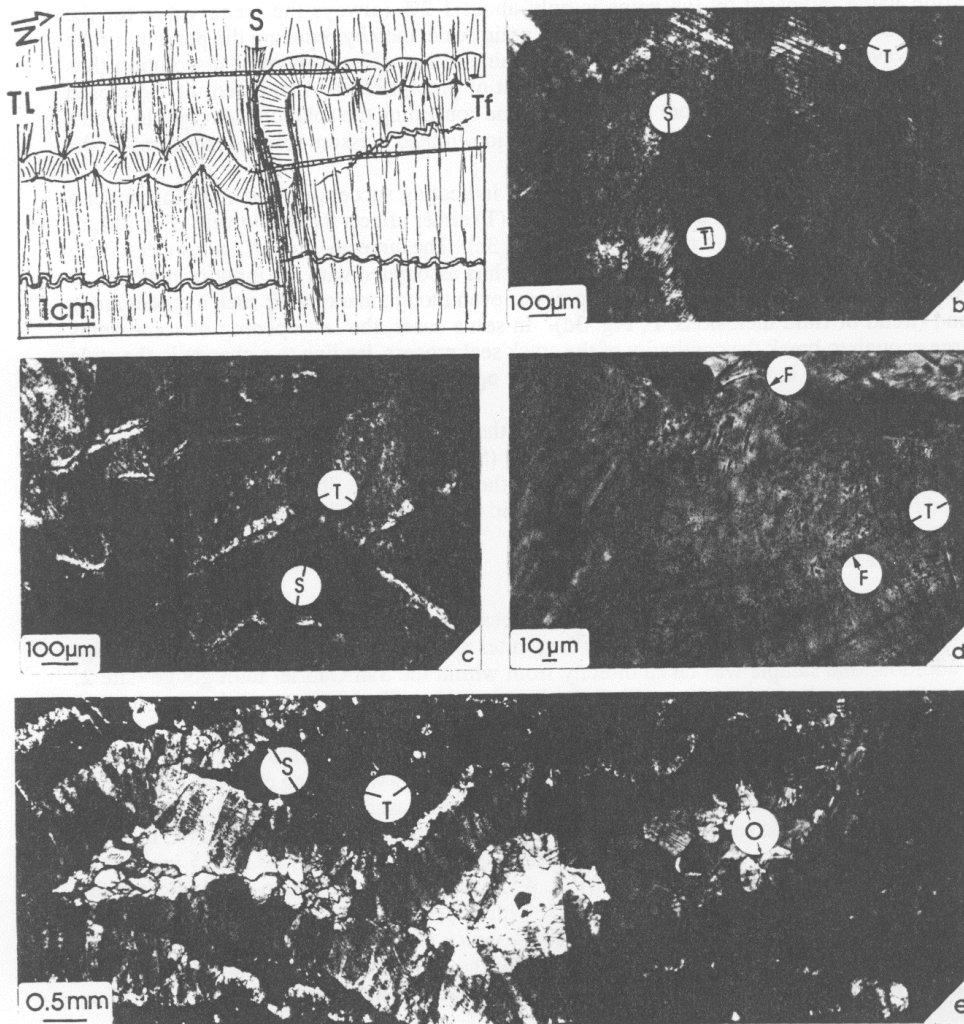


Figure 3: Solution cleavage (S) and crack sealing (T) (with input of material), near the Little Pine fault (site 1, Fig. 2). (a) Sketch of an outcrop, the younger veins (Tl) are almost undeformed and perpendicular to the solution cleavage (S), earlier veins (Tf) are tightly folded and locally dissolved. (b) & (c) Crack seal mechanism in calcite veins, each crack event either affects the whole vein (b) or is localized to just the outer limits of some grains (c). (d) Trail of fluid inclusions (F) which may indicate local self-crack healing. (e) In some cases, the fast opening of some veins was large enough to break the continuity of the crack sealing process, leading to a change in the nature of deposition (O).

shortening perpendicular to the cleavage may be estimated by the unfolding of the later veins. The shortening reaches a minimum mean value of 30% for the outcrops (ten-hundred meters). The elongation value indicated by the veins is only about 1-2% (always the mean value for the outcrop). Precipitation might have occurred on a grain scale, thereby reducing the porosity of the rocks. However, no evidence of this is found in thin section. The amount of dissolution thus appears to have been much greater than the amount of precipitation. The extent of the closed system may have encompassed more than just the zone containing the solution cleavage. In such a case, mass transfer occurred by infiltration and diffusion-limited pressure solution, resulting in the removal of material from the shales.

Some veins are sealed by synaxial calcite fibers, indicating successive crack sealing under a nearly constant state of stress. Each crack seal event (T) either affects the whole vein (Fig. 3b), or is localized just at the outer limit of some grains (Fig. 3c). The large aperture of each successively sealed crack (from 10 to 100 μm , Fig. 3b, c, e), which is associated with the input of material, is incompatible exclusively with a self-healing process, even though markers for such a process may be found (trend of fluid inclusions, F, Fig. 3d). In some cases, the last opening of some veins was large enough to break the continuity of the crack seal process, leading to a change in the style of deposition. This is indicative of large permanent openings, which may have been associated with a high degree of fluid infiltration (O, Fig. 3e).

In conclusion, these observations suggest that the sealing of cracks, due to the input of material, is associated with pressure solution seams (S), both around faults (solution cleavage, Fig. 3a) and within veins (lateral dissolution of the fibers, Fig. 3b,c,e). Mass transfer is postulated to have occurred by fluid infiltration coupled with diffusion-limited pressure solution.

San Gabriel Fault

Samples were collected from 3 sites several meters to hundreds of meters away from the central gouge zone of the San Gabriel fault (in granites and metamorphic rocks; sites 3,4,5, Fig. 2). In addition, one sample was taken directly from within the San Gabriel fault gouge (site 2, Fig. 2). Sketches of thin sections (Fig. 4) give the main displacement and the principal deformation features. Three of the thin sections were cut in an horizontal plane (sites 2,3,5), perpendicular to several meter-long strike slip faults with displacements on the order of a centimeter. One sample was cut in a vertical plane normal to an E-W trending cleavage (site 4).

The fault displacement was accommodated by a typical cataclastic process (Fig. 5a). Secondary fractures near the narrowest cataclastic zone were developed either as Riedel fractures (R, Fig. 4 & 5a, site 2 in the main gouge) or as conjugate fractures (C & C', Fig. 4, sites 3 & 5 in country rock). Away from the cataclastic zones (Fig. 4, 5), the crack orientation (T) indicates the direction of shortening and ranges from 20° to 45° with respect to the faults. This variation may be related to a different state of stress between the country rock and the main gouge zone, but it may also have been associated with a local reorientation of the stress state within bridges connecting segments of faults, or to the rotation/distortion of some blocks. Several dissolution and sealing processes can be distinguished in the samples.

Away from the cataclastic zones, the sealed cracks (T) are associated with and are perpendicular to solution cleavage (S), Fig. 5. This solution cleavage can be seen at the scale of several grains (stylolites with partial dissolution of some minerals, Fig. 5c) or at the scale of a single grain boundary (offset of dissolved markers along a stylolitic surface, Fig. 5d,e). Quartz and feldspar both show evidence (see Fig. 5) for dissolution (solution cleavage, S) and precipitation (crack sealing, T). The aperture of the cracks (or at least the thickness of the filling material) is easy to estimate when the sealing mineral is different from the host mineral (Fig. 5f,g,h,i). Crack apertures range from 10 to 100 μm . Apertures are more difficult to estimate when the sealing and the host mineral are not easily differentiated. An example of transgranular cracks (T, Fig. 5i)

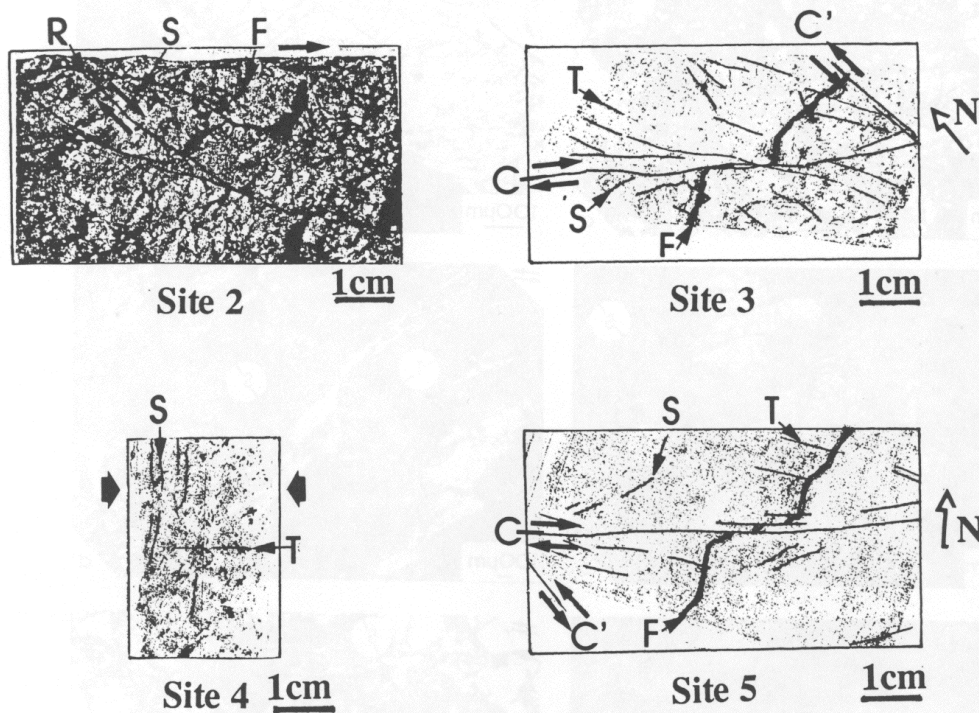
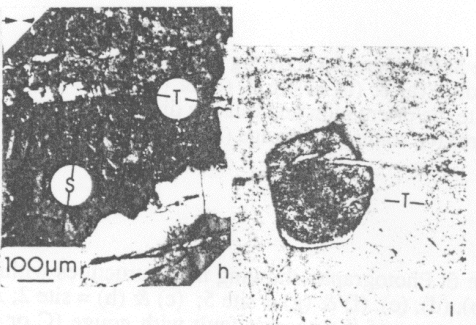
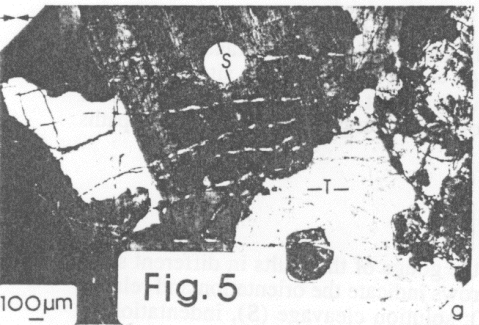
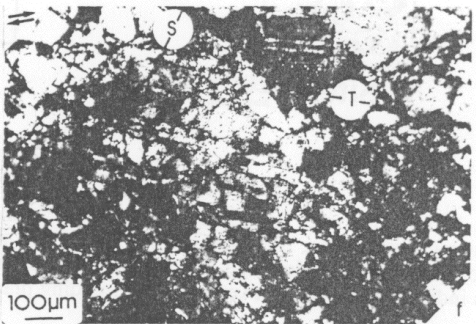
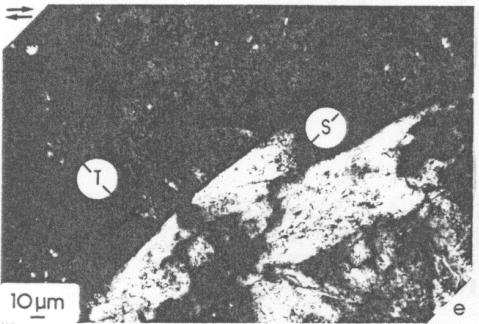
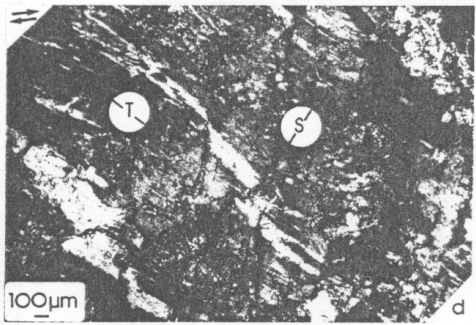
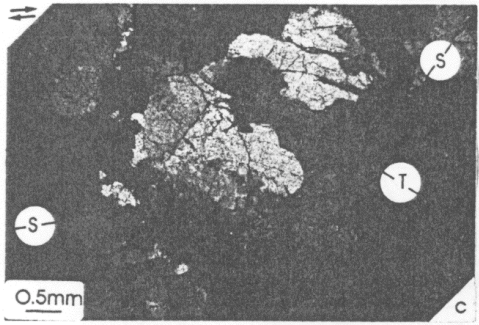
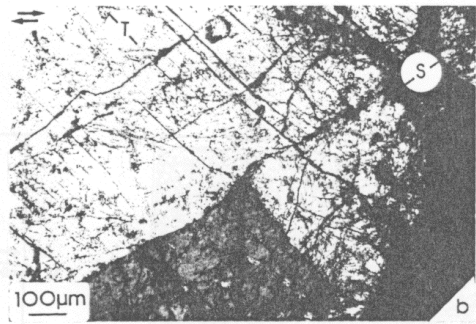
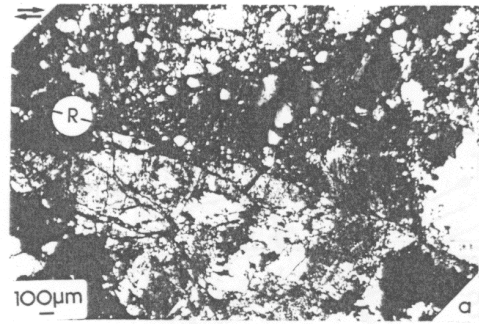


Figure 4: Thin sections from samples collected from sites 2,3,4, & 5 (Fig. 2) in granites and metamorphic rocks near the San Gabriel fault. Fault with gouge (C and C'), Riedel fracture (R), foliation (F), solution cleavage (S), crack (T). Sections in horizontal planes for sites 2,3, & 5; arrows indicate North, and in vertical E-W trending plane for site 4.

Figure 5: Photographs showing microstructures located at different sites some distance away from the faults: (a) & (b) = site 2, (c) & (d) = site 5, (e) & (f) = site 3, (g) & (h) = site 4. Arrows indicate the orientation and relative displacement of the nearest fault with gouge (C or C'); Riedel fracture (R), solution cleavage (S), crack (T) and growth of micas along the main infiltration paths (M).

Figure 6: Photographs showing microstructures within the gouge of the faults in different sites: (a), (b), (d), (e), (f) & (g) = site 5; (c) & (h) = site 2; Arrows indicate the orientation and relative displacement of the nearest fault with gouge (C or C'); solution cleavage (S), indentation by dissolution (I) and zone of local plasticity (P).



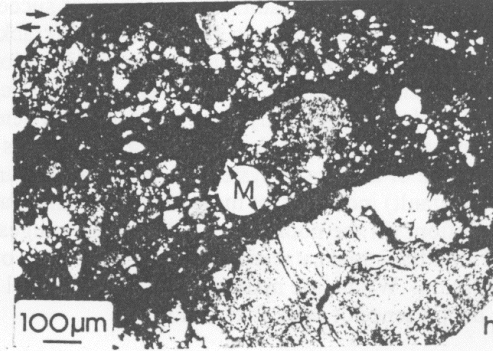
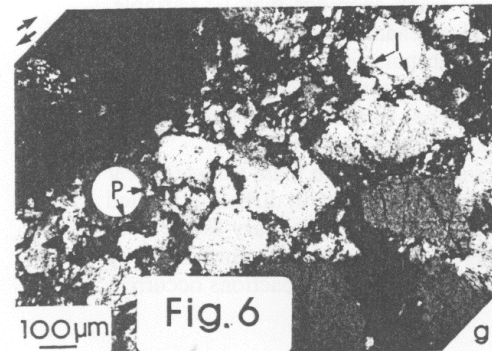
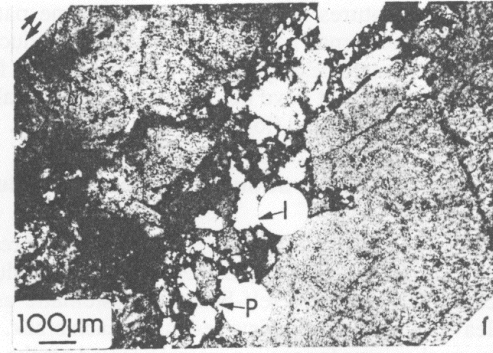
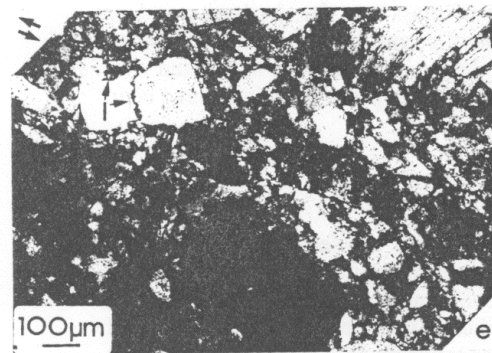
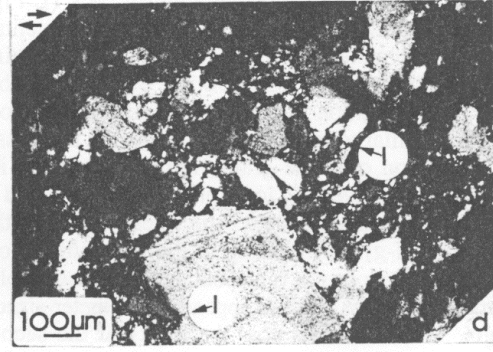
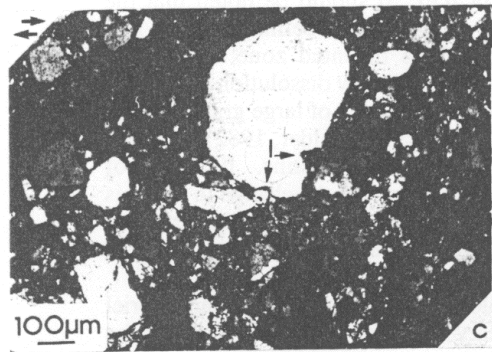
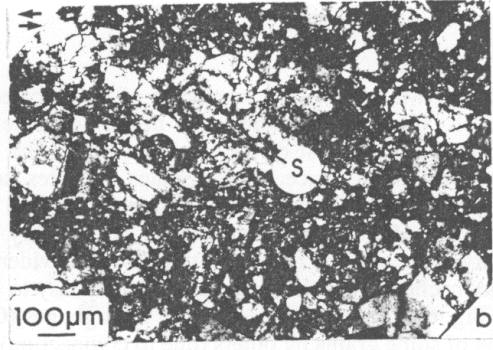
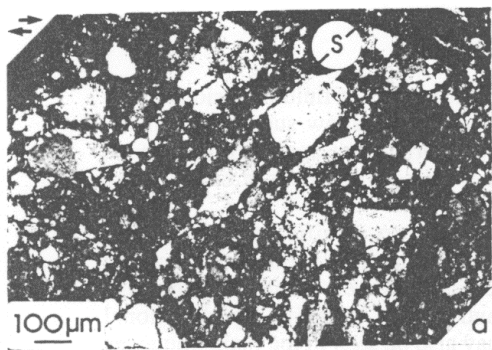


Fig. 6

shows that the two limits of the cracks are often indicated by two trails of fluid inclusions in the host mineral. Contrary to some veins in the Miocene shale (which show successive micro-opening and sealing), most of the fractures within the San Gabriel fault appear to have been opened and sealed only once. This perhaps indicates successive heterogeneous states of stress associated with possibly large earthquakes on this fault.

The sealing of interconnected cracks is probably sufficient to significantly reduce the permeability of the country rock. But there is even evidence for compaction within the cataclastic zones. The mechanism of this compaction is not easy to identify, however. Solution cleavage is found (S, Fig. 6a,b) within two conjugate fault gouges (C and C', Fig. 4 site 5). Equidirectional indentation of some grains by others (due to compaction) may also be seen in the main gouge, as well as in secondary gouges in the country rock (I, Fig. 6c,d,e). It should be noted that the mechanical indentation of grains has been experimentally reproduced (Deelman, 1976); this produces crushed material which remains in the vicinity of the indented zone. The field observations, in our case, do not show any evidence for this. Indentation by dissolution is then the most plausible explanation for these structures. In addition, the indentation of large grains (I, Fig. 6c) looks very similar to indented fossils created by pressure solution (Engelder, 1982). Finally, the interpenetration of some grains implies a change in shape of these grains. Evidence of undulating extinction revealing local plasticity is observed (P, Fig. 6f,g), but the change in shape is not always associated with such structures. Most of this change is associated with pressure solution (I, Fig. 6f-g). Partitioning of the mechanisms of deformation has thus been observed, where part of the compaction is attributable to pressure solution.

Some special features seem to appear in decimeter-sized blocks collected within the San Gabriel fault gouge. Mica minerals coat the limits of the secondary fault gouges (M, Fig. 6h). The growth of these minerals is also observed in pressure shadow zones around large quartz grains (Fig. 6h). These features indicate that along some paths of mass transfer, hydrothermal or metamorphic reactions may have contributed to the reduction of permeability (see also Wintsh et al, this volume). Nonetheless, these reactions are limited for the most part to areas associated with large fracture systems that have not been completely sealed. These types of reactions have been related to large fluxes of fluid flow (Chester et al., 1993).

Conclusions from observations of naturally sealed cracks

Fluid inclusions indicate that a fluid phase was present during the sealing of the cracks. Observations show that most of the cracks were sealed due to the precipitation of input material; these cracks could not have self-healed. Systematic associations between crack sealing and solution cleavage, and the complete sealing of each crack, indicates that pressure solution is one of the primary mechanisms for sealing interconnected cracks near active faults.

In close proximity to the faults, the location of successive crack openings may be variable. Successive openings can occur in the same vein (crack-seal mechanism; see Ramsay, 1980), where this leads to mineral fibers parallel to the direction of constant displacement (part of the veins near the Little Pine fault). Alternatively, they can occur as different cross-cutting veins, each one being opened and sealed only once in a zone with heterogeneous (seismic) displacement (part of the veins near the Little Pine fault and most of the veins near the San Gabriel fault).

The large size of the closed system and the large distances associated with mass transfer (near the Little Pine fault) indicate that mass transfer by infiltration was coupled with diffusion-limited pressure solution. In the country rock, the small gouges associated with secondary faults were compacted after the initial cataclastic event. At least part of this compaction was accommodated by pressure solution. Within the main gouges, hydrothermal and/or metamorphic reactions occurred, indicating the presence of large fluxes of fluid.

EXPERIMENTAL DEFORMATION AND SEALING PROCESSES

Compaction and deformation in flow-through experiments

Methods: Deformation experiments using halite aggregates (8.8% initial porosity) were performed in a flow-through system at room temperature (Chen et al., 1992). Saturated NaCl solutions, maintained at a constant in-flow fluid pressure, were used as the infiltrating fluid. In addition, one run used ethanol as the infiltrating solution. A constant axial stress ($\sigma_1 = 2$ to 6 MPa) and constant confining pressure ($\sigma_3 = 1$ MPa) were also imposed on the system (Fig. 7a). During the experiments, several parameters were monitored: displacement of the piston (Fig. 7b), mass of the fluid at the output (Fig. 7c), and in-situ porosity (using an acoustic wave velocity scanner; Fig. 7d). After the experiments, the porosity and the X-ray tomographic density were measured (Fig. 8a-8d); these were supplemented by optical microscope and SEM observations (Fig. 9a,b).

Results: The overall effect of the flow-through experiments under applied stress was to decrease the permeability; our results are similar to those obtained in other flow-through experiments on quartz aggregates (Elias & Hajash, 1992; Blanpied et al., 1992) and during the initial stages of experiments on jointed marble (Marone et al., 1988). In our experiments, changes in permeability were strongly dependent on the deviatoric stress level. When the deviatoric stress was zero ($\sigma_1 = \sigma_3$), finger-shaped dissolution zones occurred, which during the first day, lead to a dramatic increase in the sample permeability (Fig. 7c). With the deviatoric stress ($\sigma_1 - \sigma_3$) ranging from 1 to 5 MPa ($\sigma_3 = 1$ MPa), the permeability initially increased with the appearance of finger-shaped dissolution zones, but then subsequently and rapidly decreased after the first few days (Fig. 7c,d). Deformation was associated with the changes in the permeability. In all of the experiments, the measured strain rates always approached a stable limiting value as a function of time (see Fig. 7b). In the presence of ethanol, the recorded deformation was an order of magnitude less than observed for similar experiments using saturated NaCl solutions (Fig. 7b), and the permeability remains almost stable with only a very slow decrease (Fig. 7c).

During the first few days of the experiments using NaCl saturated solutions, the extent of dissolution was indicated by a decrease in the acoustic wave velocity (Fig. 7d); this was subsequently confirmed by SEM observations of free-face dissolution (Fig. 9a). The initial stages of the experimental runs involved the rapid dissolution of some minor minerals (carnallite, sylvite) which were not in equilibrium with the original NaCl saturated solutions. In addition to this, dissolution reactions were driven by the enhanced solubility of surface fines created by the crushing of the grains (surface curvature-solubility effect) and by the increased elastic and plastic surface strain energy associated with the imposed stress (free-face pressure solution, Fig. 9a).

After the first few days, the permeability always decreased over a period of several months of NaCl solution infiltration, up to the point where the samples became impermeable to further fluid flow (Fig. 7c,d). The impermeability was induced by the progressive sealing of the paths of fluid flow. This can be seen in the X-ray tomographic density cross sections of the samples after different time durations of fluid flow (Fig. 8a-d). In these figures, low-permeability zones are white colored areas that laterally spread out over time (Fig. 8a = initial, 8b = 4 days, 8c & d = 2 months). Just before the complete sealing of the sample, infiltration was localized along single finger-shaped paths of transport. Traces of these localized transport pathways (observed as zones of high porosity) remained on some cross sections (Fig. 8d) even after circulation had stopped. The remaining porosity (non-connected) may decrease over time, due to the action of slow mass transfer processes, such as diffusion. SEM observations of the samples (post-experimental) also show that the open paths of fluid flow (Fig. 9a) were side-by-side with zones of pressure solution

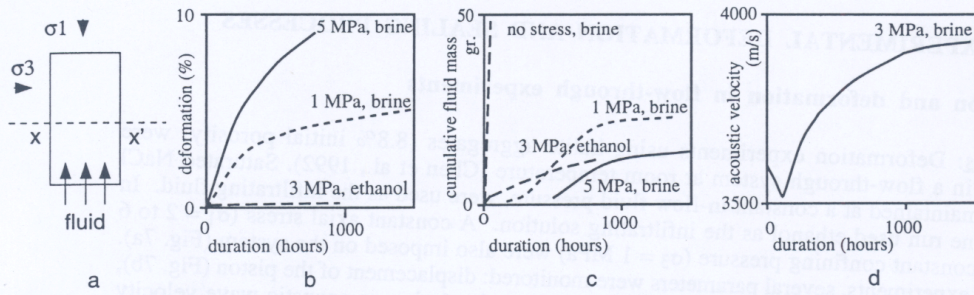


Figure 7: Experimental deformation of halite in a flow-through system: (a) sketch of the experimental system with stress and fluid flow directions (arrows). (b) deformation (c) cumulative mass fluid collected at output (d) acoustic wave velocity versus time. Stress values are the deviatoric stress ($\sigma_1 - \sigma_3$). Brine is the NaCl saturated solution.

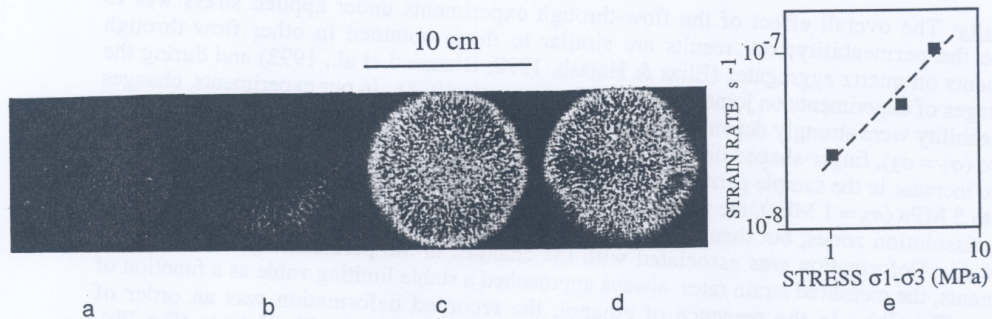


Figure 8: X-ray tomographic density image of the sample (white = low porosity, black = high porosity), location of the cross sections (a), (b) & (c) in Fig. 7a (XX'): (a) initial (b) after 4 days (c) & (d) after 2 months of fluid flow (e) Strain-rate versus stress relation for compacted aggregate (deformation with constant strain rate when most of the porosity is destroyed, see Fig. 7b).

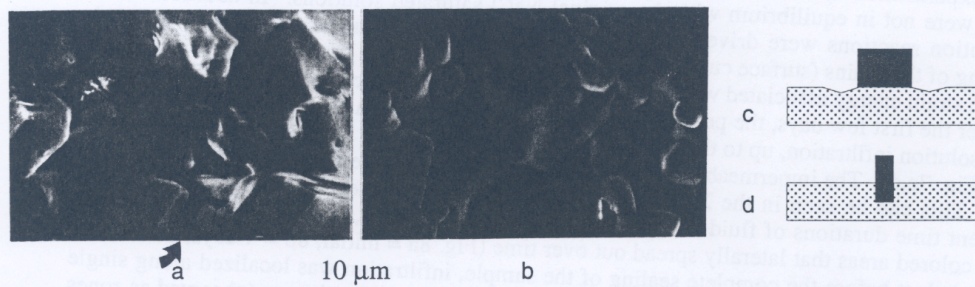


Figure 9: Scanning electron microscope (SEM) observations after fluid flow: (a) fluid transport path enhanced by free-face dissolution (arrow) (b) compaction of the grains by pressure solution (with associated water-film diffusion). Indenter experiments on halite: (c) free-face pressure solution, or (d) water-film diffusion pressure solution; the mechanism depended in part on the initial saturation state of the solution and on the diameter of the indenter.

compaction (Fig. 9b). Observations show that dissolution occurred on all the faces under stress (whatever their orientation versus stress).

Discussion: The thermodynamic driving forces associated with compaction and deformation by pressure solution are a function of solution undersaturation, strain energy, and normal stress. The order of these factors is closely related to their chronological importance in the experiments, as is discussed below.

1 - Solution undersaturation: The initial contact between the NaCl-saturated brine and the halite salt core produced the rapid dissolution of minor secondary phases, such as sylvite (KCl) and carnallite ($\text{KMgCl}_3 \cdot 6\text{H}_2\text{O}$). The driving force for the dissolution reactions of these minerals is related to the degree of solution undersaturation. Thus, the Gibbs free energy (ΔG_{diss}) driving the dissolution reaction (i.e. $\text{AB} \rightarrow \text{A}^+ + \text{B}^-$) for a given salt phase can be expressed as

$$\Delta G_{\text{diss}} = RT \ln \frac{a_{\text{A}^+} a_{\text{B}^-}}{K} \quad (1)$$

where $a_{\text{A}^+} a_{\text{B}^-}$ is the activity product of aqueous species A^+ and B^- at any given time, and K is the equilibrium constant for the dissolution reaction. The reaction is driven in the forward direction until the activity product equals the equilibrium constant. Based on the chemical analyses of the output solutions, the concentrations of K^+ and Mg^{2+} in solution exceeded those of Na^+ (by up to a factor of 4), indicating that the initial stages of dissolution favored the dissolution of sylvite and carnallite. This is due to the fact that the initial solutions were undersaturated with respect to these phases.

2 - Strain energy: The second phase of dissolution and deformation, which occurs on a time scale of days to months, is related to the plastic and elastic deformation of surfaces *surrounding* grain-to-grain contacts. The associated dissolution occurs on grain surfaces in contact with a free liquid. The strain energy associated with plastic and elastic deformation (Reuschlé et al., 1988) serves to increase the Gibbs free energy for the dissolution reaction in the following manner:

$$\Delta G_{\text{diss}} = (\Delta G_{\text{diss}}^0 - \Delta E) + RT \ln K' \quad (2)$$

where ΔG_{diss}^0 is the Gibbs free energy for the reaction with all components in the standard state, ΔE is the strain energy associated with deformation, and K' is the solubility product of the strained phase. The Gibbs free energy of reaction, based on K' , becomes

$$\Delta G_{\text{diss}} = RT \ln \frac{a_{\text{A}^+} a_{\text{B}^-}}{K'} \quad (3)$$

Thus, the above reaction will proceed in the forward direction so long as the activity product $a_{\text{A}^+} a_{\text{B}^-}$ is less than the solubility product (K') of the strained phase. It is important to note that since $K' > K$, dissolution (which is equivalent to free-face pressure solution) can occur in saturated solutions only with respect to those grain areas that have been subjected to strain.

3 - Normal stress: Finally, dissolution and deformation by pressure solution reactions *between* grain contacts occurs very slowly, on the order of months to years. The effect of a normal stress on a solid serves to increase the driving force for dissolution by a quantity equal to the product of the normal stress on the solid (σ_n) and the molar volume (V_α) (see Robin, 1978). Therefore, the Gibbs free energy driving pressure solution (i.e. dissolution *between* grain-to-grain contacts) can be expressed as

$$\Delta G_{\text{diss}} = (\Delta G_{\text{diss}}^0 - \sigma_n V_\alpha) + RT \ln K'' \quad (4)$$

where K'' is the solubility product associated with the normal stress σ_n . The above relationship predicts an increase in the kinetics of dissolution (i.e. strain rate) at grain-to-grain contacts with an increase in the normal stress. However, as discussed earlier, the limiting reaction mechanism may not be a function of the rate of detachment of species at the surface/fluid interface, but rather may be limited by the rate of diffusion of product species in the trapped fluid film between the grain contacts (water-film diffusion). The value of the diffusion coefficient estimated from the aggregate deformation (see below, comments of Fig. 8e) may be compared with other standard values (see discussion). Precipitation of products occurs on surfaces in contact with a free liquid, or on those faces subjected to the minimum compressive stress.

The observed equidirectional dissolution of mineral grains, which seems to be a characteristic of the compaction process, has also been observed with other experiments (Spiers & Schutjens, 1990; Cox & Paterson, 1991), as well as on some natural structures (Tigert & Al Shaieb, 1990). This type of dissolution occurs as long as the fluid pressure remains less than σ_3 . This is due to the fact that the chemical potential of faces perpendicular to both σ_1 and σ_3 is higher than the chemical potential of free faces (faces submitted to fluid pressure). The higher the chemical potential, the greater is the driving force for dissolution; this is due to the fact that the gradient in chemical potential drives pressure solution.

When the connectivity between the pores is destroyed, the fluid pressure in isolated voids increases to a value approaching σ_3 . Mass transfer of material takes place between faces perpendicular to σ_1 (zones of dissolution) and faces perpendicular to σ_3 (zones of precipitation). This progressively leads to a change in grain shape (i.e. deformation- see sketch of this evolution in Fig. 10a). With levels of finite compaction/deformation less than 9%, these changes in grain shape were not easily observed. Nonetheless, when considering the derived stress versus strain rate relation (Fig. 8e) obtained just before the complete sealing of the sample (last part of the strain versus time curve, with constant strain rate in Fig. 7b), this relation is linear and characteristic of pressure solution creep. At this latter stage, even if some isolated finger-shaped transport zones remained open (Fig. 8d), the majority of the sample had already undergone a significant degree of compaction (mean final porosity = 2.7%).

Finally, it is important to recall that no significant compaction was obtained on halite aggregates in the presence of ethanol. The relatively insoluble nature of halite in ethanol precluded its dissolution by pressure solution, either by free-face or by water-film diffusion. This is a confirmation that the compaction and deformation recorded in our experiments with NaCl saturated solutions was due to pressure solution, and did not occur via plastic deformation.

Driving force and kinetics of mass transfer: dissolution under indenters

An indenter technique was used to perform pressure solution experiments on halite (Gratier, 1993) and quartz. The experiments permitted the differentiation of two different mechanisms of pressure solution-deposition, depending on the conditions of deformation. *Free-face pressure solution* (Fig. 9c) occurred rapidly (time scale of days to weeks) and was driven by elastic and plastic strain energy (Tada & Siever, 1986), as indicated by dissolution around the indenters (i.e. conical-shaped grooves, with internal diameter equal to the indenter diameter). This effect was only observed when the initial solution was slightly undersaturated. It was independent of the diameter of the indenters and it was probably reaction rate limited. Dissolution at the indenter/mineral interface, associated with *water-film diffusion* (Fig. 9d), occurred slowly (over months, to one year) and was driven by the difference in normal stress, as indicated by dissolution under the indenters (i.e., cylindrical-shaped holes with diameters equal to the indenter diameter). The displacement rate was inversely proportional to the diameter of the indenter and was probably water-film diffusion rate limited. In this case, a linear relation was observed between the displacement rate and the deviatoric stress values (Gratier 1993); this result was analogous to the deformation relation observed for the halite aggregates (Fig. 8e).

The conclusion that can be drawn from such experiments is that the saturation state of the solution may change the relative importance of the various driving forces of mineral/water mass transfer. Since large and intermittent fluid flow can occur during rock deformation (Sibson, 1977; Etheridge et al., 1984; Kerrich, 1986), the rate-limiting mechanism associated with pressure solution may change from free-face reactions (when the solution is slightly undersaturated) to water-film diffusion (a process which continuously occurs, but at much slower rate).

Conclusions from the experimental approach

Fig. 10a summarizes the proposed interpretation of how pressure solution mediated dissolution of halite aggregates occurs. In general, these conclusions are also valid for the experiments carried out using the indenter method.

1 - Free-face fluid/solid reactions (often reaction rate limited) usually occur rapidly, on the order of days to weeks. These reactions are driven by solution undersaturation with respect to minor secondary phases, by surface energy (dissolution of crushed grains), and by elastic and plastic strain energy. These reactions always lead to the dissolution of grains, and depending on the degree of solution supersaturation, will also lead to precipitation reactions. Nonetheless, only special conditions generally lead to systematic crack sealing at this initial step in the deformation process.

2 - Compaction by pressure solution (water-film diffusion-limited) occurs slowly (months to years), with dissolution occurring at grain-to-grain contacts (equidirectional dissolution) and precipitation occurring on grain surfaces in contact with a free liquid. This process occurs as long as the fluid pressure remains lower than the minimum compressive stress.

3 - Deformation by pressure solution (also water-film diffusion-limited) is developed with dissolution occurring at grain-to-grain contacts perpendicular to the maximum compressive stress and precipitation occurring at surfaces submitted to the minimum compressive stress. This process occurs also very slowly (months, see discussion below) after the pore pressure reaches the minimum compressive stress (due to pore sealing).

DISCUSSION

Successive crack seal mechanisms in the interseismic period

When integrating both natural and experimental observations, the following possible successive crack sealing processes may be expected to occur in active faults during interseismic periods (see sketch, Fig. 10b). Initially, an earthquake will serve to increase the overall permeability and reduce fluid pressures to levels approaching hydrostatic values within the fault zone.

The first stages of fluid/rock interactions can be characterized by rapid kinetics which are dependent on the reactions taking place on free faces (Fig. 10b-1). The nature of the crack aperture also plays an important role. Small aperture cracks can be self-healed, where the driving force for the reaction is the minimization of crack surface free energy (Smith & Evans, 1984; Brantley et al., 1990). Fluid/mineral interactions in larger aperture cracks can be driven by a variety of processes: rapid dissolution of extremely small grains created in cataclastic zones (Elias & Hajash, 1992), dissolution of surfaces with elevated elastic and plastic strain energy (Tada & Siever, 1986) dissolution of mineral phases not in equilibrium with the infiltrating solution (Chester et al., 1993). Systematic sealing by free-face reactions is only expected under special conditions, such as: cracks with a small aperture, the presence of supersaturated fluids, hydrothermal or metamorphic reactions which produce new minerals (Wintsh et al, this volume). The kinetics of free-face reactions are generally reaction rate limited in the case of low solubility minerals (silicates, oxides), which have high activation energies (50-90 kJ/mole/K: Lasaga, 1984;

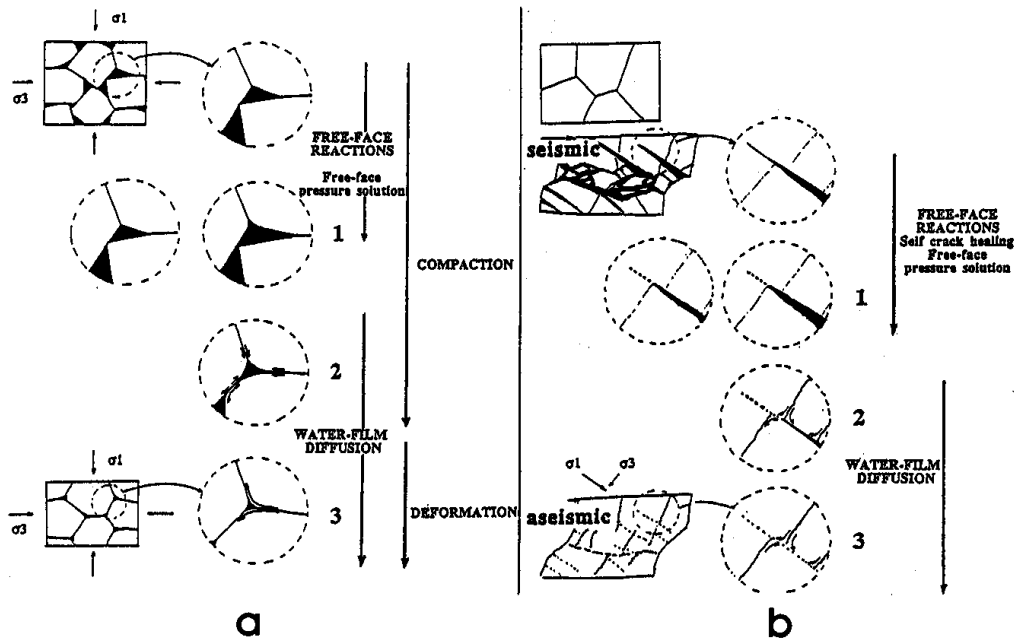


Figure 10: Possible evolution of the mechanisms of dissolution and precipitation (a) in aggregate and (b) in fractured rocks, solid is white, fluid is black. At the beginning of the compaction (a-1) or after an earthquake (b-1) free-face reactions occur rapidly due to effect of increased surface, plastic, and elastic energies (days to months). But only special conditions can lead to the sealing of the sample with such a mechanism. When pressure solution is driven by differences in normal stress along grain boundaries (a2-3) or around the blocks (b2-3), the rate of sealing will be determined by water-film diffusion with very slow kinetics (years to thousands of years).

Smith & Evans, 1984; Gratier & Jenatton, 1984; Gratz et al., 1990). The dissolution of high solubility minerals, however, may be limited by diffusion at the (free) fluid/mineral interface.

Pressure solution creep (Fig. 10b, 2 -> 3) is the process responsible for the progressive and subsequent complete sealing of cracks and fissures associated with fault zones. The gouges are compacted in part by pressure solution (see Fig. 10a-2 with related discussion). Dissolution of material at grain-to-grain contacts normal to the maximum effective stress, followed by water-film diffusion, eventually leads to the precipitation of dissolved material, where precipitation is initially favored on the free faces of grains (sealing and compaction), then on faces normal to the minimum compressive stress (deformation). Such diffusive mass transfer generally occurs in small closed systems (100 μm to mm scale, Fig. 1d). However, large-scale infiltration coupled with diffusion-limited pressure solution occurs on larger scales (Fig. 1c"), which is the reason why larger closed systems are amenable to sealing, as well. The progressive sealing of a system implies that fluid pressures should increase from hydrostatic to lithostatic levels with time.

Kinetics of crack sealing by pressure solution

The estimation of the kinetics of crack sealing is possible using certain theoretical relations derived for pressure solution. A simplified geometry for crack sealing is given in Fig. 10b (step 2 -> 3). Dissolution occurs on surfaces normal to the maximum compressive stress, whereas precipitation occurs on the free faces parallel to this compressive stress (crack limits). A method for estimating the incremental displacement rate ($\dot{\gamma}$) for the dissolution of a stressed face is based on the following expression (Rutter, 1976):

$$\dot{\gamma} = \alpha D w c \Delta\sigma v / R T d^2 \quad (5)$$

where D is the water-film diffusion coefficient for the rate limiting species, w is the width of the transport path, c is the solubility, v is the molar volume of the soluble species, $\Delta\sigma$ is the deviatoric stress (i.e. the difference in stress between the zone of dissolution and the zone of precipitation), R is the universal gas constant, T is the temperature (K), d is the diameter of the dissolution zone, α is a numerical constant of integration dependent on the geometry of the block (32 for a disk-shaped dissolution surface, Rutter, 1976).

The major unknown parameters for pressure solution are D and w . These values may be estimated from the quartz and halite experiments. Using Eqn. 5, Dw may be estimated from the indenter experiments on halite. A mean value of $4.4 \times 10^{-19} \text{ m}^3/\text{s}$ was determined. From the flow-through halite aggregate experiments, a value of $6.6 \times 10^{-20} \text{ m}^3/\text{s}$ was obtained. These values may be compared with those obtained by Hickman (1989): $Dw = 10^{-19} \text{ m}^3/\text{s}$ for the dissolution/deformation of halite at the same range of stress. The deformation of quartz aggregates (at 350°C in a 1.0 N NaOH/H₂O solution) yielded perhaps more reliable values for crustal deformation. The values obtained ranged from 2.5×10^{-19} to $2.5 \times 10^{-20} \text{ m}^3/\text{s}$ (Gratier & Guiguet, 1986). Experiments using indenter techniques (unpublished data on quartz) gave values of about $10^{-19} \text{ m}^3/\text{s}$.

Dw values ranging from 10^{-19} to $10^{-20} \text{ m}^3/\text{s}$ seem to be a rough approximation for quartz at depths corresponding to temperatures of 350°C . The other parameters necessary for applying Eqn. 5 to natural deformation may be taken from the literature. The solubility of quartz in solution at 350°C can be derived from $\log K = -1.89$ (Rimstidt and Barnes, 1980). Based on the geometry of naturally deformed rocks around faults, the dimensions associated with the dissolution zone may be estimated. It is commonly observed that in natural systems mass transfer is not limited to the scale of single grains, but rather is constrained to a multigrain scale. This scale corresponds to multigranular blocks that are limited in size by solution cleavage (at the base) and cracks (sides)

(Gratier, 1987; Gratier & Gamond, 1990). A commonly observed crack width is on the order of 10 μm (see Fig. 5). The mean diameter of the zone of dissolution (d) around each crack can be estimated to range from 100-500 μm . Using an assumed deviatoric stress value of 50 MPa, the time needed for complete crack sealing ranges from 5.6 to 56 years (with $d = 100 \mu\text{m}$), and 140 to 1400 years (for $d = 500 \mu\text{m}$). Of course, a more accurate estimation depends on knowledge of both the exact geometry of the dissolution and deposition zones and on the use of more accurate values for the diffusion coefficient (D) and the width of the water-film (w).

In conclusion, the kinetics of crack sealing by pressure solution can be quite variable. The rate of crack sealing may be quite rapid if dissolution occurs on free faces (Fig. 1c', reaction rate-controlled). However, when kinetically-limited by diffusion through a trapped water-film, the rate of diffusion may constrain the rate of increase of fluid pressure within faults. In such a case (Fig. 1c", 1d), pressure solution is not a rapid process relative to other geodynamic processes which can lead to elevated fluid pressures. The estimated values for pressure solution crack sealing are of the same order of magnitude as the recurrence time of earthquakes.

Strain softening and hardening associated with pressure solution

Since pressure solution on polymineralic aggregates can induce chemical differentiation, this process is associated with strain softening and hardening. The strength evolution of rocks can be considered both in terms of time and space.

Spatial evolution. Most soluble minerals (quartz, feldspar, calcite) are mechanically stronger than insoluble micas and clays (minerals which are typically alteration products). The selective removal of soluble minerals decreases the strength of rocks and leads to a progressive localization of dissolution along cleavage seams (Cosgrove, 1976; Robin, 1979). On the contrary, precipitation of minerals increases the strength of rocks and leads to zones of mineralization that are stable with respect to further dissolution. This mineralogical differentiation occurs in active faults between the gouge zone (zone of dissolution with decreased quartz and feldspar contents) and the country rock (zone of crack sealing with increased quartz and feldspar contents).

Time evolution: The dependence on time is a function of the geometry of the features associated with pressure solution. When pressure occurs with parallel solution cleavage normal to planar veins (Fig. 3, 4, 5, 6a-b) the geometry is stable and the banding (layer with dissolution / layer with precipitation) evolves with time up to an equilibrium state which depends on the conditions of deformation (Gratier, 1987). On the contrary, when dissolution/precipitation occurs at the scale of a grain (Fig. 6c-g), the time evolution depends on the geometry and on the type of pressure solution mechanism. For the case of a monomineralic aggregate under a constant mean deviatoric stress, free-face dissolution reduces the overall grain size (Dewers & Ortoleva, 1990). The decrease in the grain-to-grain surface area (Fig. 9a) increases the local stress imposed on the areas of contact and leads to an accelerated dissolution rate; this can be considered to lead to strain softening. Counteracting this effect is the process of progressive grain-to-grain indentation. The associated increase of grain-to-grain surface decreases the local stress on the contacts and leads to progressive strain hardening of the aggregate (Fig. 6 and 9b).

CONCLUSIONS

Experiments and observations of natural structures show that various types of solid/fluid reactions can occur during interseismic periods. The kinetics of these reactions depends on the nature of the limiting process. Solid/fluid reactions may be divided into two categories:

Reactions which occur between a solid under stress and a free fluid phase. These types of reactions include self-crack healing, dissolution of crushed grains, free-face pressure solution (driven by strain energy), metasomatic reactions. Most of these processes are reaction rate limited, where the slow step in the process is related to detachment or precipitation kinetics. Of course, highly soluble minerals may dissolve in a diffusionally-limited manner. Time scales for these free-face reactions are on the order of days to months. External conditions associated with deformation and mineral/fluid interactions (temperature, pressure, fluid and solid compositions, fluid flow rates) determine whether dissolution occurs by itself, or whether it is subsequently followed by localized precipitation.

Observations show that in active faults most of the cracks are sealed due to the input of material and have not self-healed. Hydrothermal and metamorphic reactions have been observed to occur within gouge zones, but they do not seem to be associated with the sealing of transport paths. Only special conditions can therefore produce systematic and rapid crack sealing with free-face reactions.

Reactions which occur within trapped fluid phases between two solids under stress. These reactions include compaction and crack sealing (driven by the difference between the normal stress at grain-to-grain contacts and the surfaces of grains in contact with free fluids present in interconnected voids and cracks). The dissolution process is postulated to be limited by the rate of diffusion in the trapped water-film. These rates are typically much slower, with crack sealing occurring on the order of years to thousands of years. In this case mass transfer can be either water-film diffusion (Fig. 1d), or fluid infiltration coupled with water-film diffusion (Fig. 1c").

The systematic association between crack sealing and solution cleavage indicates that pressure solution-deposition reactions are the primary mechanism for sealing interconnected cracks near active faults. This type of crack sealing, even though it occurs on a micrograin scale, can completely seal a fracture system at a much larger scale, due mainly to widespread fluid circulation and infiltration. The compaction of gouges after the original cataclastic event can in part be attributed to pressure solution.

Some consequences of pressure solution crack sealing. If pressure solution is rate limited by water-film diffusion, then the kinetics of crack sealing are slow enough to be an important process in controlling the increase in fluid pressure around faults during interseismic periods. It also has an important bearing on the associated mechanisms of critical failure. The time evolution of pressure solution, however, not only depends on the operative mechanisms, but also on geometrical constraints which determine the scale over which the process is effective (solution cleavage scale vs. single grain dissolution).

Deformation associated with pressure solution very often induces the chemical and mechanical differentiation of rocks. Gouge zones where preferential dissolution occurs generally lose mechanical strength in comparison to zones (country rock) where associated precipitation reactions take place. The progressive decrease in fluid flow through faults occurs in parallel with the progressive decrease of the post-seismic deformation rate due to pressure solution, perhaps to a near constant creeping rate (with mass transfer from cleavage to vein) when the fault fracture system has been completely sealed. This fits in well with the overall deformational behavior of the upper crust, where brittle behavior (cataclastic faults, seismic events, fast strain rates) alternates with ductile behavior (solution cleavage, aseismic creep, slow strain rates).

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