Experimental constraints on the diagenetic self-sealing capacity of faults in high porosity rocks

B.T. Ngwenya a,*, S.C. Elphick a, I.G. Main a, G.B. Shimmield a,b

a Department of Geology and Geophysics, University of Edinburgh, West Mains Road, Edinburgh EH9 3JW, UK
b Dunstaffnage Marine Laboratory, P.O. Box 3, OBAN, Argyll PA34 4AD, UK

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Abstract

A thorough understanding of fault seal processes is important in many practical and geological applications, which depend on subsurface flow of fluids. While the mechanisms involved in fault sealing are well known, the microscale processes involved and their relative contribution to sealing remain debatable. In particular, the extent to which diagenetic processes overprint cataclastic fault sealing has not been resolved, mainly due to the long time scales required to measure these effects. Here, we report results from a novel suite of room temperature experiments that combined continuous analysis of dissolved silica using on-line high performance liquid chromatography, with low strain rate creep loading on sandstone cores. This technique allowed changes in silica concentration during different phases of deformation to be resolved, and revealed a 7-fold increase in overall silica concentration immediately after dynamic faulting by localised cataclasis. Calculations based on these results show that the mass of dissolved silica from the resultant fault gouge increased by up to two orders of magnitude relative to that from the intact rock over the same time scale. This increase represents the first stage of the inherent diagenetic sealing capacity of the fault, presumably through localised diffusive mass transfer. Post-test microstructural studies suggest that the magnitude of diagenetic self-sealing depends on lithological and mechanical attributes of the host rock, which control fault gouge microstructure. Our experiments suggest that diagenetic processes may account for permeability reduction of up to two orders of magnitude, comparable to reductions due to cataclasis alone. Together, these two processes account for the 5–6 orders of magnitude reduction of permeability observed in natural faults and deformation bands. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Faults play an important role in the movement and channelling of fluids in the subsurface. Thus predicting their hydraulic behaviour has become a major pursuit in recent years [1–3], mainly because of its importance in practical (e.g. hydrocarbon exploitation; groundwater flow; contaminant transport; and geothermal energy production) as well as geological (e.g. crack-seal veining, earthquake recurrence cycles) applications. However, our understanding of the major factors that control the hydraulic properties of
faults remains incomplete. For example, field observations of exposed fault rocks suggest that the hydraulic characteristics of a given fault can vary strongly in space and time, so that the same fault may have acted as both a conduit and a seal during deformation [4,5]. A complete understanding of fault hydraulic properties therefore requires consideration of the full deformation cycle, in relation to the local mechanical and chemical processes, with strong feedback between the two. This requires careful integration of many types of data, including geometry of individual faults, the statistical properties of fault populations, experimental and numerical modelling studies of faulting processes and sealing mechanisms [6].

Despite some recent success in this endeavour, many issues remain to be resolved with regard to fault seal prediction. For example, with the exception of direct measurement from hydraulic pump tests, information about the actual petrophysical properties of fault rocks is often only available from direct studies of faulting processes and sealing mechanisms, as revealed by microstructural observations of fault rocks in the field or laboratory. Based on mainly field observations, several workers have proposed a generic classification of the mechanisms by which faults can seal [7–9]. Passive mechanisms create fault seals in a purely physical sense, by bringing rocks with different permeabilities against each other. They include simple ‘juxtaposition’ seals and clay smears. However, many fault sealing mechanisms are active, and result from a combination of physical and chemical processes occurring within the fault. Purely physical processes include porosity collapse due to cataclasis, grain disaggregation, mixing and translation, while purely chemical processes include localised pressure solution, and diagenetic cementation from regional advection of fluids [6,9].

So far, significant progress has been made in understanding the mechanics of clay smear seals, leading to development of fairly robust predictive algorithms that have been calibrated against known hydrocarbon column data [10]. In contrast, our understanding of the role of other sealing mechanisms is incomplete, and no comparable quantitative algorithms exist at present for predicting their effects on fluid flow. For example, while localised cataclastic seals have been known for a long time [11,12], the extent to which they reduce permeability has only recently been quantified, both experimentally and from outcrop studies. Outcrop measurements on natural cataclastic faults in high porosity sandstones show permeability decreases of between five and six orders of magnitude relative to the host rock [1,2]. However, laboratory experiments with a liquid permeant consistently show that cataclasis in porous sandstones only reduces fault permeability by up to three orders of magnitude relative to the host matrix [13,14]. To explain the difference, Fulljames et al. [13] suggested that the extra sealing may be due to local or regional diagenetic processes operating over longer time scales. In this paper, we examine this question of a local diagenetic source quantitatively using a novel suite of laboratory experiments, and by mass balance calculations based on the experimental results.

To date, relatively few experiments have attempted explicitly to quantify the magnitude of fault sealing due to local diagenetic processes in reservoir sandstones, largely because it has proven difficult to measure at the relevant pressures (≤ 700 bars) and temperatures (≤ 120°C). Experiments relevant to seismogenic (greenschist metamorphic) conditions only show permeability decreases of about one order of magnitude [15]. While this may partly reflect the short duration of the experiments (up to 5 days), such studies have sometimes been used as evidence that chemical processes may be only of secondary importance in fault sealing at lower temperatures. The problem of quantifying fault sealing due to chemical processes operating under diagenetic conditions therefore remains unresolved, particularly as direct measurement of permeability reduction by 2–3 orders of magnitude would require unrealistically long experiments. We set out to test if we could estimate the magnitude of potential diagenetic sealing on a local scale from a suite of laboratory experiments in which fault gouge dissolution is measured relative to that of the matrix rock with the same mineralogical composition.
The use of dissolution experiments to study aspects of the precipitation process is not new; a similar approach was used to calculate precipitation rates for quartz from solubility data by Rimstidt and Barnes [16].

We first describe the experimental methodology and the results, and then show using mass balance calculations that these are consistent with a two orders of magnitude potential reduction in permeability following the cataclastic failure of porous rock samples at low strain rates.

2. Experimental methods

2.1. Design considerations

Our objective was to determine changes in mineral dissolution associated with deformation in general and cataclastic faulting in particular, and to use these observations to place constraints on diagenetic self-sealing of faults. Since cataclasis generates fine grains (fault gouge), we expect the faulting process to be associated with an increase in solute concentration in the fluid. This may occur as a result of increased dissolution rate expected for finer grains [17], or due to reduction in the precipitation rate associated with reduced total surface area per grain [18], due to the improved packing properties of fault gouge.

Two problems must be overcome before attempting to quantify changes in mineral dissolution due to faulting. Firstly, the time span of conventional laboratory deformation experiments is too short relative to mineral dissolution rates at low temperatures to allow the effects of initial loading to be resolved from those due to subsequent faulting. Secondly, frequent sampling of the pore fluids is required, especially during or after faulting, but this is limited by the volume of fluid needed for analysis while maintaining the low flow rates necessary to observe changes in mineral dissolution. The novelty in our approach lies in combining high resolution sampling using on-line high performance liquid chromatography (HPLC), which only requires µl analytical volumes [19], with slow strain rate creep experiments. This combination allows the correlation of mechanical and geochemical attributes during rock deformation, and also successfully resolves faulting from loading effects.

In order to determine changes associated with cataclastic faulting, we measured the extent of mineral dissolution for intact samples and then compared this with dissolution after fracture. Central to our interpretation of the diagenetic sealing is the assumption that in a subsurface environment, an increase in dissolution due to faulting will represent 'supersaturation' with respect to grains in the intact rock. We show below that this assumption has a sound theoretical basis when our experimental data are extrapolated to natural conditions.

2.2. Theoretical background

The experiments were based on the scenario that a core sample of given length ($L$, cm) and cross-sectional area ($A$, cm$^2$) is loaded to a constant stress (Fig. 1) while a fluid of known composition is pumped through it at a constant flow rate, $Q$ (cm$^{-3}$ s$^{-1}$). Assuming, for simplicity, that the rock is monomineralic and neglecting aqueous phase reactions, which are relatively fast [20], we can write a one-dimensional material balance equation for the evolution of dissolved solutes at a given time ($t$, s) and at a given point ($x$, cm).
along the sample length thus [21]:

$$\frac{\partial C}{\partial t} = -v \frac{\partial C}{\partial x} + \frac{\alpha R^+}{\phi} - \frac{\alpha R^-}{\phi}$$

(1)

In this equation, $C$ is the concentration (mol l$^{-1}$) of the dissolved species in the advective fluid at position $x$ and time $t$, $\phi$ is the porosity of the sample (dimensionless), $\alpha$ is the stoichiometric constant of the dissolution–precipitation reaction (dimensionless), $v$ is the linear flow velocity along the sample (cm s$^{-1}$), while $R^+$ and $R^-$ are the dissolution and precipitation rates in the sample, respectively (mol cm$^{-3}$ s$^{-1}$). For simple oxides such as quartz, the precipitation rate varies linearly with concentration of dissolved silica in solution [16], so that Eq. 1 can be re-written thus [22]:

$$\frac{\partial C}{\partial t} = -v \frac{\partial C}{\partial x} + \frac{\alpha R^+}{\phi} - \frac{\alpha R^- C}{\phi}$$

(2)

where $R^\pm = R^-/C$ is the modified precipitation rate (l cm$^{-3}$ s$^{-1}$). Explicit in Eq. 2 is the fact that the concentration of the dissolved solute will vary along the length of the column. However, for times long enough for reactions to be sufficiently resolved from initial loading effects, the concentration in the fluid will reach steady-state at all points along the core, including at the outlet ($x = L$). Thus departures from this steady-state condition at fracture can be exploited to assess the effects of faulting on dissolution.

The full solution to this inhomogeneous equation under these conditions is given in the EPSL online Background Dataset¹, based on the analysis by Franklin et al. [22]. We repeat this analysis in a form that corrects two typographic errors in their derivation. A major assumption in this derivation is that, at long distances away from the inlet of the core, and at sufficiently low flow rates, the fluid composition will be at or close to equilibrium with the dissolving mineral, i.e. $C$ in the last term of Eq. 2 equals the equilibrium concentration, $C_e$. Since distilled water was injected into the core, the boundary condition at the inlet end is $C(x = 0) = 0$, whence

$$C^w(x) = C_e \left[ 1 - \exp \left( -\frac{\alpha R^- x}{\phi V} \right) \right]$$

(3)

where $C^w$ is now the steady-state concentration. We analysed at the outlet of the core sample in our experiments, whence $x = L$ in Eq. 3. The derivation assumes that the porosity of the sample remains effectively constant during the lifetime of these experiments. This is justified since the mass dissolved is very low, although it can be accurately measured (see later).

Eq. 3 allows us to predict steady-state concentrations in our experiments at a given temperature and pressure if the precipitation rate $R^-$ is known. Since the rate varies along the length of the core sample, it cannot be calculated simply from exit concentrations. However, for a given mineral, $R^-$ is related to sample characteristics by the equation [22]:

$$R^- = k_\rho A_s (1 - \phi)$$

(4)

where $k_\rho$ is the apparent precipitation rate constant (l cm$^{-2}$ s$^{-1}$), $\rho$ is the density of the dissolving or precipitating mineral (g cm$^{-3}$), and $A_s$ is surface area per unit mass (cm$^2$ g$^{-1}$), as measured from gas adsorption (BET) studies [23]. This allows us to use experimentally determined values of $k_\rho$ from the general literature. Because we did not measure surface areas, $A_s$ was estimated on a geometric basis [23]. However, it has recently been shown that geometric surface areas calculated this way for quartz are systematically about seven times lower than real reactive surface areas as measured by BET methods [24]. This so-called roughness factor was therefore accounted for in our calculations. By varying the flow rate (i.e. $v$) in Eq. 3, it is possible to show whether our measured solution concentrations follow the predicted values, and to validate the assumption of fault-induced supersaturation under natural flow rates.

2.3. Materials and methods

Two Permian aeolian sandstones (Clashach and

¹ http://www.elsevier.nl/locate/epsl; mirrorsite: http://www.elsevier.com/locate/epsl
Locharbriggs, UK) were used as test samples. These have almost identical detrital mineralogy of quartz and K-feldspar, but differing diagenetic and petrophysical attributes (Table 1). Clashach is notable for its quartz overgrowths, producing a well-cemented sandstone with porosities of 12–18% and permeabilities ranging from 200 mD to 1 D [19]. Haematite and illite coatings dominate the diagenetic assemblage in the Locharbriggs sandstone, and these have produced a weakly cemented sandstone with porosities of 22–26% and permeabilities ranging from 10 mD to 1 D. These differences also allowed us to assess the effects of host rock attributes on diagenetic fault sealing.

The experiments were conducted in a triaxial flow rig \( (\sigma_1 > \sigma_2 = \sigma_3) \) at room temperature and involved creep-to-failure, followed by frictional sliding on the new fault. Cores measuring between 75 and 80 mm long and 38 mm diameter were loaded to differential stresses ranging from 85 to 95% of their short-term failure strength, with a radial confining pressure of 6.9 MPa. The differential stress was maintained constant until the samples failed by accelerating creep. Axial strain was measured continuously using an externally mounted linear variable differential transducer, while helium-purged distilled water flowed through the cores at 0.1 ml min\(^{-1}\). For the range of porosity and cross-sectional area in our samples, this volumetric flow rate translates into linear flow velocities \( (u) \) of about 3.0 cm h\(^{-1}\). These velocities were too high for solutions to reach equilibrium with the minerals over the lengths of samples used, but were dictated by the need to deliver an accurate and constant flow rate.

At regular intervals, 100 \( \mu \)l samples of the exit pore fluid were extracted and loaded onto an online Water’s IC-PAK\(^{\text{a}} \) anion column for analysis of dissolved silica using 2.5 mM lithium hydroxide eluent flowing at 1.2 ml min\(^{-1}\), and a Water’s 431 conductivity detector. A three-standard calibration was run prior to analysis and the eluent, which degrades with time, was changed every 4–5 h. The method detection limit was 80 ppb of \( \text{H}_2\text{SiO}_4 \) and analytical errors were below 2%.

At the end of the tests, the samples were removed from the pressure cell while still encased in the confining rubber sleeve. After drying to constant weight at 60°C, the samples were impregnated under vacuum with an extra low-viscosity epoxy resin so that the gouge and other fine particles would not be displaced during impregnation. Thin sections made perpendicular to the shear fractures were used to study microstructural and fault gouge characteristics under backscattered scanning electron microscopy (SEM).

In order to quantify microcrack distributions, photomicrographs were taken across and near the fractures. These photographs were scanned into a software package called OPTIMAS\(^{\text{a}} \) for measurement and counting of microcracks, after calibration of the field of view. Depending on the magnification of the photomicrograph, the smallest microcracks resolvable by this technique were about 0.3 \( \mu \)m long.

### 3. Results

#### 3.1. Mechanical and chemical data

Measurements of axial strain and pore fluid

<table>
<thead>
<tr>
<th>Sandstone</th>
<th>Figure</th>
<th>Flow rate ( (\text{cm}^3 \text{ min}^{-1}) )</th>
<th>Length ( (\text{mm}) )</th>
<th>Porosity ( (%) )</th>
<th>Grain diameter ( (\text{mm}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clashach</td>
<td>(2a)</td>
<td>0.1</td>
<td>81</td>
<td>16</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>(2c)</td>
<td>0.1</td>
<td>77</td>
<td>14</td>
<td>0.4</td>
</tr>
<tr>
<td>Locharbriggs</td>
<td>(2b)</td>
<td>0.1</td>
<td>76</td>
<td>24</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>(2d)</td>
<td>0.1</td>
<td>75</td>
<td>22</td>
<td>0.2</td>
</tr>
</tbody>
</table>

The apparent precipitation rate constant \( (k_p) \) was calculated from the apparent dissolution rate constant \( (k_\varepsilon, \text{ mol cm}^{-2} \text{ s}^{-1}) \) from Berger et al. [40] and quartz solubility \( (\text{mol l}^{-1}) \) from Langmuir [27] based on the principle of detailed balancing [41] yielding \( k_p \) with units of 1 cm\(^{-2}\) s\(^{-1}\).
dissolved silica (H₄SiO₄) for the two sandstones are shown in Fig. 2. Before failure, strain was found to follow the three classic stages of creep: transient, steady-state and accelerating creep. Transient creep was found to obey a logarithmic law \[25\], and strain eventually attained steady-state rates of about \(10^{-9}\) s⁻¹. At failure, there was up to 1 mm axial shortening of the sample, followed by another phase of decelerating creep. These trends were repeated following frictional sliding.

We note that the concentration of dissolved silica in the exit fluid is correlated with the different stages of creep for the Clashach sandstone (Fig. 2a). After an initial rise due to loading, the concentration drops exponentially during the transient creep phase, attains a steady-state value during steady-state creep and increases steadily during the accelerating creep phase. Comparison with the Locharbriggs sandstone (Fig. 2b) run at the same stress magnitude (90% of failure strength) shows a higher concentration due to loading and no increase in dissolved silica during tertiary creep. However, most of the features of Fig. 2a are reproduced. The higher concentration due to loading for Locharbriggs samples may be due to the smaller grain size relative to Clashach samples (Table 1 and Fig. 3), which favours pressure solution. For the Clashach sandstone, the exit fluid reaches a steady-state concentration of about 1 ppm; rising to about 1.4 ppm during accelerating creep (Fig. 2a), while the steady-state concentration for the Locharbriggs sandstone is about 0.7 ppm (Fig. 2b).

Fig. 2. Graphs showing time-dependent strain and the concentration of dissolved silica (H₄SiO₄) in the exit fluid. The four graphs are shown on the same y-axis scale and the higher concentration of silica at fracture for Clashach relative to Locharbriggs samples is evident. F = faulting and S = sliding.
The development of the macroscopic fault is marked by a dramatic increase in silica concentration in the exit fluid. This behaviour is observed for both sandstones, but the Clashach samples showed a higher increase in concentration than the Locharbriggs samples. This increase was transient, and concentrations dropped to the pre-fracture levels within 5 days. In this respect, our observations are similar to those of Dewers and Hajash [18] for crushed powders, but also differ in two ways: (i) we produced a localised fracture whose thickness could be measured for use in calculations of total flux from the new surface area; (ii) there was continuous flow so that mineral precipitation was unlikely to occur, as would be the case in their semi-flow experiments. These differences allowed us to arrive at good estimates of the contribution to dissolved silica by the fracture and hence an estimate of diagenetic sealing of the fault.

Further experiments conducted at different stresses show that these geochemical changes are reproducible and hence characteristic of the creep-to-failure process (Fig. 2c,d). The Clashach sample deformed at 92% of failure strength (Fig. 2c) fails earlier than that at 90%, and the steady-state concentration appears not to have been attained before failure occurred. A rather interesting observation is that after failure, the exit tubing was blocked (tested by reversing flow), resulting in a progressive increase in pore pressure within the sample. This induced frictional sliding along the fault and the resulting continuous ‘grinding’ is manifest in a continuous increase in dissolved silica in the exit fluid. Meantime, the second Locharbriggs sample (Fig. 2d) loaded to 85% of failure strength lasted longer, reaches lower steady-state concentrations, and shows a much smaller silica signal at failure. These differences in failure times are consistent with the stress dependence of failure times found in other studies [25].

Frictional sliding also manifests in an increase in silica concentration, but to levels lower than those due to fracture. However, the increased levels due to sliding appear to be sustainable over longer periods relative to the dynamic failure (Fig. 2a), consistent with a more efficient grinding process. Fig. 2 also highlights the advantages of combining creep tests with on-line chemical analysis. We can resolve the 3–4 h transient increase in silica concentration, associated with the initial phase of loading. In a comparative conventional deformation test, using a strain rate of $10^{-5} \text{s}^{-1}$, fracture occurred within 1–2 h of initial loading.

Fig. 3. Backscattered electron photomicrographs comparing microstructural distributions and fault gouge characteristics for samples with on-line data shown in Fig. 2a,b. The Clashach sample (a) shows pervasive microcracks in the sample while the fault gouge consists wholly of comminuted grains (shown on the north-east corner of the shear fracture). In contrast, the microcracks are restricted to a thin fringe bordering the shear fault in the Locharbriggs sample and some intact grains have been incorporated into the fault gouge (b). Differences in microcrack density lead to different amounts of fresh surface area in the two rock types and may explain the difference in silica concentrations associated with failure.
and the silica concentration at fracture could not be resolved from that due to initial loading.

3.2. Microstructural observations

Fig. 3 shows backscattered electron photomicrographs of the fractured samples taken around areas in the vicinity of micro-faults created. The samples failed by a single fracture oriented at about 40° to the maximum principal stress, although small subsidiary axial fractures were also present. The two photographs were taken at the same scale and the slightly larger grain size for the Clashach samples (Fig. 3a) is evident, while the Locharbriggs samples have a higher porosity, with less obvious grain–grain contacts (Fig. 3b). The Clashach sample shows pervasive intra-granular and trans-granular microcracking and the fault gouge consists wholly of comminuted grains (Fig. 3a). This suggests that the formation and linkage of microcracks was the dominant deformation mechanism that led to the failure of this sandstone. Microcracking increases surface area to volume ratio in the whole sample, but particularly in the fracture, by producing grains much finer than the bulk grains. In contrast, microcracks in the Locharbriggs sandstone were restricted to within 0.5 mm of the fault and the fault gouge still contained whole grains of the matrix material (Fig. 3b). These observations are suggestive of deformation by compaction and grain translation, producing gouge with a lower surface area to volume ratio.

These qualitative observations of deformation mechanisms were quantified by analysis of microcrack distributions around the micro-faults. We counted the number, and measured the total length of microcracks in eight separate photographs taken on a transect across each micro-fault (Fig. 4). Firstly, both microcrack density and total crack length are higher at all points in the Clashach relative to the Locharbriggs samples. Secondly, both parameters decrease exponentially away from the micro-fault, and take the general form \( N = a e^{-bx} \), where \( N \) is the number or density of microcracks per unit area, \( x \) is the distance (mm) away from the micro-fault and \( a \) and \( b \) are constants. This is consistent with previous observations of microcrack distributions in samples of deformed granites as well around fault zones generally [26].

4. Discussion

We have shown that the fracture process leads to a significant increase in the concentration of dissolved silica in distilled water flowing through the core, and that concentrations are nearly back to the bulk, steady-state levels approximately 120 h after fracture (Fig. 2a). Since differential pressures across the samples were not measured, it is
not possible to quantify permeability changes due to deformation and/or dissolution. However, in one case during the test shown in Fig. 2a, the pump used had a pressure transducer to indicate backpressure downstream of the fluid reservoir. A record of this pressure is shown in Fig. 5a, from which we observe a 6-fold increase in back-pressure immediately at failure. This pressure increase is a qualitative indication of a decrease in permeability associated with cataclastic faulting. Although the pressure drops to about 0.1 MPa as the fault gouge dissolves the small changes in dissolved silica and the erratic subsequent changes in backpressure suggest that these permeability changes are mostly of mechanical origin. Nevertheless, we can use observed changes in dissolution to estimate the potential for diagenetic self-sealing.

4.1. Diagenetic sealing potential

Our main objective was to investigate whether the fracture process contributes significantly to the amount of diagenetic sealing seen in natural faults, where solutions are likely to be at equilibrium with bulk rock minerals. We can get a semi-quantitative idea of the diagenetic sealing potential of faults if we use the steady-state concentration before failure (Fig. 2) to represent an equivalent ‘equilibrium’ concentration with respect to the bulk rock grains. Since the flow rate was maintained constant throughout our experiments, the measured increase in dissolution after fracture can be thought of as representing the degree of supersaturation with respect to the bulk rock grains. In nature, the excess silica will precipitate on large grains through Ostwald ripening, leading to fault seal. Calculated steady-state silica values for bulk grains at 25°C are about 0.8 ppm, close to our experimental measured values of 1 ppm. Note that at both grain sizes, equilibrium concentrations are obtained at the outlet even at much higher flow rates under diagenetic (100°C) conditions.
Eqs. 3 and 4 and the Clashach data (Fig. 2a). The calculation assumes a monomineralic rock, which can be justified for Clashach samples as they contain ≥ 90% quartz [19]. Under the room temperature conditions of our experiments, Eq. 3 predicts an outlet \([H_4SiO_4]\) of 0.80 ± 0.1 ppm. True equilibrium concentrations require flow rates of \(≤ 0.001\) ml \(\text{min}^{-1}\), i.e. three orders of magnitude less than we were able to achieve with available pumps. The calculated silica concentrations agree reasonably well with our measured values of 1 ppm, particularly as our calculations did not account for the presence of K-feldspar and effects of non-hydrostatic loading on grain contacts. Outlet silica concentrations at 100°C were also calculated based on quartz solubility and enthalpy data from Langmuir [27], and are also shown in Fig. 5b. Under these conditions, which are more representative of diagenetic environments in the top few kilometres of the earth, exit fluids reach equilibrium with quartz even at flow rates as high as 1 ml \(\text{min}^{-1}\). The calculated values are similar to flow rate-independent concentrations measured by Franklin et al. [22] using these flow rates at 100°C. The discussion above emphasises the fact that in some natural fault settings, flow rates may be sufficiently low, or temperatures are sufficiently high, for fluids to be at equilibrium with the bulk rock minerals. Any amount of fault-related dissolution such as that observed in our experiments and those of Dewers and Hajash [18] will therefore produce fluids supersaturated with respect to these bulk minerals, potentially leading to fault seal by mineral precipitation.

Mechanistically, fault-induced supersaturation may be evaluated using the Freundlich–Ostwald equation, where the smallest (most reactive and most abundant) particles determine solubility, so that the new equilibrium solubility is given by [28]:

\[
C = C_0 \exp \left( \frac{2\sigma V_m B}{3rRT} \right)
\]  

(5)

Here, \(C\) is now the solubility (mol \(\text{l}^{-1}\)) of finer grains produced due to faulting, with diameter \(r\) (cm), \(V_m\) is the molar volume of the dissolving mineral (\(\text{cm}^{-3}\) \(\text{mol}^{-1}\)), \(\sigma\) is its surface free energy (\(J \text{ cm}^{-2}\)), \(B\) is a geometric constant (dimensionless), \(R\) is the gas constant (\(J \text{ mol}^{-1} \text{ K}^{-1}\)), \(T\) (K) is the temperature and other terms are as defined before. As pointed out by Parks [23], this condition is true only if the kinetics of precipitation are slow, as has recently been demonstrated to be the case [18]. Calculations using Eq. 5 indicate that significant increases in dissolution occur only at grain radii of less than 0.1 \(\mu\)m [28,29]. The smallest grain size that we observed by SEM was of 0.25 \(\mu\)m radius. However smaller, amorphous grains have been reported in fault gouges [30], particularly at high strains. In addition, surface energy depends more on radius of curvature rather than grain size [28]. Our gouge was examined after several days of dissolution, so it is reasonable to assume that much lower grain sizes would have been present immediately after fracture. In any case, their high dissolution rates dictate that they may not even be preserved in the short term in an aqueous environment. Included in Fig. 5 are corresponding curves for 0.1 \(\mu\)m grains at both 25°C and 100°C. It is noticeable that even at this grain size, supersaturation with respect to bulk grains exists in any fault zone under diagenetic flow rate and temperature conditions, even without having to adjust the calculation for reaction rate dependence on grain size [17]. Thus, our experiments mimic what should happen during faulting in nature, and should allow us to arrive indirectly at order of magnitude estimates of diagenetic self-sealing.

We estimated the additional mass of dissolved silica due to fracture by integrating the \([H_4SiO_4]–\text{time} \) curve over the 120 h duration when \([H_4SiO_4] \) is above steady-state values and multiplying this by the flow rate. The curve exhibits a linear relationship with time up to peak concentration, followed by a power-law decay post-peak:

\[
M = Q \int_{219}^{219} kCdt + dt + \int_{337}^{0.25} C_0 t^{-n}dt
\]  

(6)

where \(M\) is the mass of \(H_4SiO_4\) (mg), \(Q\) is the volumetric flow rate (1 \(\text{min}^{-1}\)), \(k\) is the slope of the linear component, \(C_0\) is a constant and \(n\) is the exponent. The total mass of additional
H$_4$SiO$_4$ calculated this way is about 2 mg. By comparison, the total amount of H$_4$SiO$_4$ that would be released from dissolution of the bulk core over the same duration is about 0.7 mg. While this difference is small, the additional silica over this time frame must be interpreted in terms of its source volume. The 0.7 mg came from the whole sample while the additional 2 mg came from the fault zone only. By normalising the masses calculated to their respective source volumes (9.1$\times$10$^{-2}$ dm$^3$ for the whole sample and 2.7$\times$10$^{-3}$ dm$^3$ for the fault, calculated from the fracture thickness and inclination), we get values of 7.7 mg dm$^{-3}$ for the bulk sample and 748 mg dm$^{-3}$ for the fracture. In this case, faulting increases total dissolved silica per unit volume of rock by about two orders of magnitude. For the case where minerals were initially in equilibrium with the fluid, the excess silica represents a potential source for the diagenetic cementation of the fault even at room temperature, albeit at times much longer than our experiments. Our experiments have also shown that continuous shear along the fault produces more fine particles, leading to continuous increases in fluid silica concentrations (Fig. 2c). Such long-term ductile compaction is thought to be an important mechanism for generating excess pore pressures in seismogenic zones [31]. It would therefore seem plausible that a significant component of sealing measured in natural cataclastic faults at reservoir temperatures [13] is due to this diagenetic self-sealing.

4.2. Diagenetic sealing mechanisms and geologic applications

It is not possible from our experimental data to identify mechanisms that operate in nature during diagenetic self-sealing of faults. However, the increased silica concentration associated with fracturing and frictional sliding can be attributed to two processes that have previously been suggested as diagenetic fault seal mechanisms. Ostwald ripening is the process where small grains in a multimodal population dissolve in the fluid and the dissolved solute re-precipitates on larger grains [23,32]. In contrast, pressure solution results from the presence of a chemical potential gradient between grain-to-grain contacts under normal stress and the ambient pore fluid, where the pressure is hydrostatic [33,34]. In such a system, grains dissolve at the contacts and the dissolved material diffuses into the pore spaces where precipitation occurs. Both Ostwald ripening and pressure solution are enhanced by reduced grain sizes and both result in a reduction of porosity and permeability during granular compaction. It has been suggested that in nature, the two processes are complementary [35]. Our experimental data suggest that Ostwald ripening may be favoured in the early stages of fault formation while pressure solution operates over longer time scales as the fault zone undergoes shear-enhanced compaction [34], equivalent to the faulting and sliding episodes, respectively, in our experiments.

A striking feature of Fig. 2 is the difference in solubility at faulting and frictional sliding between the two sandstones. It is probable that the difference in silica concentration between the two tests is consistent with the differing microscopic deformation mechanisms described in Section 3.2, and suggests that lithological and mechanical attributes of the host rock have a first order control on self-sealing deformation [36]. These attributes control fault gouge microstructure and include grain size, porosity and degree of diagenetic cementation. For example, growth faults, which form when sands have high potential for compaction and grain translation, are likely to show less diagenetic self-sealing. The grain size control seen in the fault gouge is also evident in the bulk rock dissolution, since the finer Locharbriggs sandstone shows higher silica concentration during the initial loading phase, consistent with pressure solution.

Finally, our experiments may be used to resolve some of the outstanding issues regarding sources of quartz cement for fault sealing in reservoir sandstones. Among the chemical seals, quartz is one of the most important causes of porosity and permeability reduction in faults. However, the source of the quartz is debatable, and the local versus external source arguments for explaining quartz diagenesis in reservoir sandstones have recently been applied to the fault seal problem [9]. In reservoir sandstones, opinion is divided almost
equally between those who favour local sources of silica via pressure solution and stylolitisation [37], and those that favour external sources. The latter include compaction of mudrocks, density-driven convection [38] and fault-focused fluid flow [39]. Our experiments suggest that most of the quartz sealing can be explained without necessarily resorting to external sources, although such sources are important for other types of cements.

5. Summary

We have demonstrated that the dynamic formation of fault gouge leads to transient increases in dissolution of minerals in the gouge relative to that in the host rock by up to two orders of magnitude. This increase provides an upper bound for the inherent diagenetic sealing capacity of the fault, since the dissolved material can precipitate within the fault and reduce its permeability. However, the overall magnitude of this self-sealing depends on a range of lithological and mechanical attributes of the host rock, which control fault gouge microstructure. Overall, our experiments suggest that diagenetic processes may account for up to two orders of magnitude of the total sealing observed in natural faults.

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