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**Importance of Water-Clay Interactions for Fault Slip in Clay-Rich Rocks**

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**Abstract** Clay-rich rocks are integral to subduction zone dynamics and of practical importance, for example, as barriers in nuclear waste and CO\(_2\) repositories. While the effects of swelling strain on the self-sealing capabilities of these rocks are relatively well-established, the implications of polar fluids interacting with charged clay particles on the frictional behavior, and the role of swelling stress in initiating slip in critically stressed faults, remain ambiguous. To address these uncertainties, we conducted triaxial friction experiments using saw-cut samples, with the upper half composed of Opalinus claystone (OPA) and the lower half of Berea sandstone (BER). The frictional strength of the non-wetted OPA-BER interface was estimated based on experiments at confining pressures of 4–25 MPa and constant axial loading rate (0.1 mm/min). Fluid injection friction experiments were performed using decape (non-polar fluid) or deionized water (polar fluid) at 10 and 25 MPa confining pressures and constant piston displacement control. Macroscopic mechanical data were complemented by distributed strain sensing on the sample surface. Compared to decape, the frictional strength of the OPA-BER interface tended to decrease when injecting water, which is attributed to phyllosilicate lubrication and the transition of the OPA from a solid rock to an incohesive mud near the saw-cut surface. When injecting water, slip was initiated during initial hydration of the OPA-BER interface, although the apparent stress state was below the yield stress. To explain this behavior, we propose that the swelling stress is a crucial factor that should be integrated into the effective stress model.

**Plain Language Summary** Clay-rich rocks are important in subduction zones and for practical applications like nuclear waste containment and CO\(_2\) storage. A unique property of clay is the ability to swell by incorporation of water. If swelling deformation is constrained, swelling stress can develop. While the enhanced swelling ability of faults due to swelling is well-known, the effect of water-clay interactions on friction and the contribution of swelling stress to fault reactivation remain unclear. We studied these effects with laboratory experiments where Opalinus claystone (OPA) was sheared against Berea sandstone. First, using experiments without fluids, we established that our set-up is representative for claystones. In further experiments, we injected a non-polar fluid that does not interact with clay. We compared these with experiments where we injected water, which is polar and therefore interacts with the charged clay. The frictional strength tended to decrease when injecting water, likely due to water acting as a lubricant and transforming OPA into a slurry. Surprisingly, movement along the interface started although the applied stress was below the threshold. We explain this by the contribution of swelling stress to the overall stress state. These findings improve our understanding of clay-rich rock behavior in nature and practical applications.

### 1. Introduction

Rocks such as claystones, argillites, or clay-shales commonly contain more than 2/3 clay-sized particles and are rich in phyllosilicates (e.g., Folk, 1968; Potter et al., 2005). Clay-rich rocks are abundant in the shallow part of subduction zone megathrusts and in the sedimentary cover of the downgoing plate (Chester et al., 2013; Underwood, 2007). Their frictional properties have been investigated in a wide range of studies showing that they are generally weak and velocity strengthening, that is, not prone to seismic rupture nucleation (e.g., Aretusini et al., 2021; Crawford et al., 2008; Faulkner et al., 2011; Ikari et al., 2007, 2009; Kohli & Zoback, 2013; Orellana et al., 2018a, 2018b, 2019; Rabinowitz et al., 2018; Saffer & Marone, 2003; Tembe et al., 2010). Beyond their tectonic significance, clay-rich rock formations have also attracted interest for critical practical applications.
to their high sealing potential, they are considered, for example, as suitable cap-rocks in CO$_2$ storage facilities (e.g., Busch et al., 2008; Song & Zhang, 2013; Zappone et al., 2021) and as natural barriers in nuclear waste deposits (e.g., Bossart et al., 2017; Delage et al., 2010), where sealing is additionally increased by engineered barriers of compacted clay minerals (e.g., Madsen, 1998; Sellin & Leupin, 2014).

The high sealing potential of clays and clay-rich rocks is a consequence of the low permeability and the naturally occurring closure of cracks (self-sealing), where the key factor for the latter is the swelling ability of clay minerals (Bock et al., 2010; Wenning, Madonna, Kurotori, et al., 2021). Typically, two mechanisms are thought to cause swelling: (a) innercrystalline swelling (also called intracrystalline or interlayer swelling), and (b) osmotic swelling (also called intercrystalline swelling) (Madsen & Müller-Vonmoos, 1989). Innercrystalline swelling is based on dipolar molecules (e.g., water) wetting the interlayer cations of swellable clay minerals (e.g., smectite), which leads to an increase of the interlayer distance (e.g., Komine & Ogata, 2003; Madsen & Müller-Vonmoos, 1989). Osmotic swelling, on the other hand, is based on an increased cation concentration close to the negatively charged external surfaces of clay minerals, also referred to as electrical double layer (e.g., Delville & Laszlo, 1990; Madsen & Müller-Vonmoos, 1989; Revil, 2012; Wersin et al., 2004; Yven et al., 2007). A superposition of electrical double layers of adjacent clay particles leads to a concentration gradient that drives water to enter the interspace between the clay particles, which in turn increases the distance between them. In densely packed clay aggregates, osmotic swelling acts similarly to intercrystalline swelling, but between adjacent clay minerals instead of between layers of a single clay mineral (Horseman et al., 1996; C. L. Zhang et al., 2010). Osmotic swelling occurs in all clay minerals, that is, also in “non-swellable” clay minerals such as illite or kaolinite. Under confined conditions, where the expansion of clays is inhibited, swelling stress develop (Madsen & Müller-Vonmoos, 1989; Wagner, 2013). Besides clay mineralogy, the amount of swelling and the extent of swelling stress build-up depends on the pore water chemistry (e.g., Abdullah et al., 1999; Giot et al., 2019; & Müller-Vonmoos, 1989; Wagner, 2013). The interaction between clay minerals and water can further lead to the division of clay particles, where innercrystalline space becomes intercrystalline (Laird, 2006; Saiyouri et al., 2004).

Opalinus claystone (OPA), the clay-rich rock studied here, is the envisaged host rock for a nuclear waste repository in Switzerland (Bossart et al., 2017) and is considered a potential cap-rock for CO$_2$ storage (Zappone et al., 2021). Hence, the hydro-mechanical properties of the OPA have been widely investigated. This includes, among others, studies concerning mechanical creep (Naumann et al., 2007; Schulze, 2011), compressive strength (Minardi et al., 2021; Schuster et al., 2021; Winhausen et al., 2022), dry and wet frictional properties (Bigaroni et al., 2023; Orellana et al., 2018a, 2019; Schuster et al., 2023), swelling strain, stress, and its anisotropy (Bossart & Thury, 2008; Minardi et al., 2016; Thury & Bossart, 1999; C. L. Zhang et al., 2010), and the closure of cracks by self-sealing processes (Bock et al., 2010; Fang et al., 2017; Voltołini & Ajo-Franklin, 2020; Wenning, Madonna, Kurotori, et al., 2021; C. L. Zhang, 2011). A stress concept for claystones such as OPA has been established, showing that an externally applied load is carried by the bound water of clay particles, that is, by the immobile water of the electrical double layers, and that the OPA may be considered as a colloidal system (Horseman et al., 1996; Rodwell et al., 1999; C. L. Zhang, 2017, 2018; C. L. Zhang et al., 2010).

While the permeability is enhanced by fault slip as shown by fault reactivation experiments (Cappa et al., 2022; Guglielmi et al., 2015) and microstructural analysis (Akker et al., 2023), swelling strain is responsible for the closure of cracks, and thus keeps the permeability of clay-rich rock formations low (Giot et al., 2019; Wenning, Madonna, Kurotori, et al., 2021). However, the contribution of swelling stress to slip initiation of critically-stressed faults remains elusive and the effect of polar fluids interacting with electrostatically charged clay particles on the frictional properties of clay-rich rocks is unclear. While comprehensive investigations are lacking, previous studies hypothesized that the expulsion of water from the interlayers of smectite during shearing may locally increase the pore pressure (C. Morrow et al., 1992), or that reactivation of faults may be assisted by the build-up of swelling stresses (M. Zhang et al., 2018). On the other hand, Moore and Lockner (2004) concluded, based on friction experiments with different sheet silicate powders, that innercrystalline swelling of smectite has no significant effect on its frictional properties.

In this study, we assess the effect of water-clay interactions on the frictional strength of clay-rich rocks by investigating OPA-Berea sandstone (BER) interfaces in triaxial saw-cut experiments. In a first test series, we examined the frictional properties of the non-wetted interface, which showed that the frictional strength is dominated by the weak OPA and thus the experimental set-up is representative for clay-rich rocks, although a bimaterial interface was used. In second and third test series, fluid injection friction tests were performed using a
non-polar fluid (decane) and a polar fluid (deionized water), respectively. This enabled us to distinguish mechanical effects (normal stress reduction by fluid pressure) from water-clay interactions that depend on the polar character of water (e.g., swelling or disaggregation of the claystone) (Wenning, Madonna, Kurotori, et al., 2021).

2. Methods

2.1. Experimental Set-Up

Triaxial friction tests were performed using oblique saw-cut cylindrical samples with a diameter $D \sim 50$ mm, a total length $L \sim 100$ mm, and an angle $\theta$ of $55^\circ$ between a plane normal to the long axis and the saw-cut plane (Figure 1a; actual sample dimensions in Table 1). The top half consisted of a clay-rich rock (OPA) and the bottom half of a permeable sandstone (BER). An angle $\theta$ of $55^\circ$ was chosen to provide a slip plane ideally oriented for reactivation, assuming that the OPA dominates the frictional strength and has a friction coefficient of 0.36, that is, a friction angle of $20^\circ$ (Marschall & Giger, 2016). The bedding-parallel foliation of the OPA was oriented parallel to the long axis of the sample and the strike of the inclined saw-cut plane. The OPA was obtained from borehole D5 drilled at the Mont Terri Underground Laboratory, Switzerland (Zappone et al., 2021). The Mont Terri Underground Laboratory currently has an overburden of ~250 m while the maximum overburden reached ~1,350 m during the Cretaceous (Mazurek et al., 2006), that is, the OPA samples used are over-consolidated under in-situ conditions of the Mont Terri Underground Laboratory. The OPA contains 10–12 wt.% quartz, around 1 wt.% feldspar, 8–24 wt.% calcite, and 58–72 wt.% phyllosilicates, which consist of 25–37 wt.% dioctahedral phyllosilicates (illite and muscovite), 16–24 wt.% kaolinite, 8–12 wt.% interstratified illite and smectite, and 4–9 wt.% chlorite (Wenning, Madonna, Zappone, et al., 2021). Despite the relatively low portion of smectite (a swellable clay mineral), OPA is well known for its ability to swell (e.g., Thury & Bossart, 1999; Wenning, Madonna, Kurotori, et al., 2021; C. L. Zhang et al., 2010), which is thought to be predominantly due to osmotic swelling (C. L. Zhang et al., 2010). The natural water content of OPA samples such as those used in this study ranges from 6.0 to 8.5 wt.% (Amann et al., 2011; Pearson et al., 2003; Wild et al., 2017). BER was purchased commercially and the supplier specifies a permeability of $2 \times 10^{-13} - 5 \times 10^{-13}$ m². The high permeability allows fluids injected from the bottom of the BER to arrive relatively quickly (within a few tens of seconds) and uniformly along the sandstone-claystone interface. Further material parameters are presented in Table S1 in Supporting Information S1 (Bossart & Thury, 2008; H. Wang, 2000; Wenning, Madonna, Zappone, et al., 2021).
Table 1
Applied Confining Pressure(s) \( P_c \), Sample Numbers, and Sample Dimensions (Diameter D and Length L) for Each Experiment

<table>
<thead>
<tr>
<th>Test</th>
<th>( P_c ) (MPa)</th>
<th>Top sample half</th>
<th>Bottom sample half</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sample Nr.</td>
<td>( D ) (mm)</td>
<td>Sample Nr.</td>
</tr>
<tr>
<td>Test 1: Friction experiments without fluid injection</td>
<td>10</td>
<td>D5_S13_4_1</td>
<td>50.0 ± 0.4</td>
</tr>
<tr>
<td>Exp. 1.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Exp. 1.2</td>
<td>10, 15, 20, 25</td>
<td>D5_S13_4_2</td>
<td>49.8 ± 0.2</td>
</tr>
<tr>
<td>Exp. 1.3</td>
<td>10, 15, 20, 25</td>
<td>D5_S31_1_1</td>
<td>50.0 ± 0.2</td>
</tr>
<tr>
<td>Exp. 1.4</td>
<td>4, 6, 8, 10, 12, 8, 6</td>
<td>D5_S24_4_1</td>
<td>49.8 ± 0.2</td>
</tr>
<tr>
<td>Test 2: Decane injection friction experiments</td>
<td>10</td>
<td>D5_S31_3_1</td>
<td>50.0 ± 0.2</td>
</tr>
<tr>
<td>Exp. 2.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Exp. 2.2</td>
<td>10, 25</td>
<td>D5_S31_3_2</td>
<td>49.7 ± 0.2</td>
</tr>
<tr>
<td>Exp. 2.3</td>
<td>10, 25</td>
<td>D5_S31_4_1</td>
<td>49.6 ± 0.3</td>
</tr>
<tr>
<td>Test 3: H₂O injection friction experiments</td>
<td>10</td>
<td>D5_S31_4_2</td>
<td>49.7 ± 0.3</td>
</tr>
<tr>
<td>Exp. 3.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Exp. 3.2</td>
<td>10</td>
<td>D5_S31_1_2</td>
<td>49.7 ± 0.4</td>
</tr>
<tr>
<td>Exp. 3.3</td>
<td>10, 25</td>
<td>D5_S24_1_1</td>
<td>49.8 ± 0.1</td>
</tr>
</tbody>
</table>

*Opalinus claystone (extracted from borehole D5, Mont Terri Underground Laboratory, Switzerland). Berea sandstone (commercially acquired). Assembled length.*

Cores of OPA and BER were extracted from larger pieces of sample material by a two-inch drill. To achieve the desired geometry, the samples were secured during the cutting of the top, bottom, and oblique saw-cuts using custom-designed fixtures. No additional surface preparation was performed. In the case of the OPA, the drilling and sawing machines were operated dry in order to avoid water-clay interactions. After each preparation step, the OPA samples were vacuum sealed in foil to prevent humidity loss. Before starting the friction tests, the OPA samples were placed into a humidity chamber with a relative humidity of 90% and a temperature of 23°C for at least 21 hr. The determination of the mass during this period shows either an exponential asymptotic increase, a relatively constant value, or, for one sample, an exponential asymptotic decrease (Figure S1 in Supporting Information S1). In all cases, the change in mass of the sample before removal from the humidity chamber was minor. This suggests that the innercrystalline swelling of the swellable clay minerals was in equilibrium with 90% relative humidity. Wild et al. (2017) measured no significant changes in water content when similarly treated samples were exposed to a relative humidity of 93%, suggesting that the water content of OPA in our experiments was close to the natural conditions. When mounting the sample in the triaxial cell, both halves were enclosed with a nitrile jacket to seal the sample assembly from the confining oil. A metal mesh (aperture size 0.085 × 0.085 mm) was placed on the top and bottom of the sample assembly to prevent rock particles from entering the hydraulic system of the triaxial deformation test apparatus.

2.2. Triaxial Friction Tests

Three categories of triaxial friction test series were performed at room temperature, hereafter referred to as Test 1–3,

- No fluid injection (Test 1): Four experiments without fluid injection (Exp. 1.1–1.4) were conducted to estimate the frictional slip envelope of the non-wetted OPA-BER interface. Note that although no fluid was injected during experiments of this test series, these are not dry friction experiments, as the natural water content of the OPA was preserved as far as possible during sample preparation and during the experiment. Slip was initiated by increasing axial displacement with a constant rate of 0.1 mm/min (causing increasing axial stress \( \sigma_a \)) under constant confining pressure \( P_c \). Multi-stage triaxial testing was performed (Kim & Ko, 1979) for experiments of Test 1 involving confining pressures of 4–25 MPa (Table 1). The estimation of the non-wetted frictional slip envelope was then used in Test 2 and 3 to define the initial stress conditions before fluid was injected.
• Non-polar fluid (Test 2): Three friction experiments (Exp. 2.1–2.3) were conducted by injection of a non-polar fluid (decane) at constant confining pressure ($P_c = 10$ and 25 MPa) and fixed far-field axial position, that is, constant piston displacement control (Ye & Ghasemi, 2018). The axial position was set so that the initial differential stress $\sigma_D$ corresponded to $\sim 70\%$ of the peak strength according to the non-wetted frictional slip envelope (i.e., $\sigma_D \sim 11.1$ and 19.6 MPa for $P_c = 10$ and 25 MPa, respectively). The fluid pressure $P_f$ was increased step-wise applying the following general protocol: At $P_c = 10$ MPa, the injection of decane was initiated with a fluid pressure of 0.1 MPa, which was held for 15 min. Subsequently, the fluid pressure was increased to 0.5 MPa and then incrementally to 8 MPa with 0.5 MPa steps, holding each fluid pressure for 5 min. At $P_c = 25$ MPa, the fluid pressure was increased in a similar incremental manner from 0.1 MPa up to 18 MPa, with steps of 0.4–1 MPa and holding times of 1–5 min. Deviations from this general protocol, that is, longer holding times, occurred due to technical issues. At a certain fluid pressure, slip along the saw cut was initiated causing a decrease of the axial stress $\sigma_1$, thus returning the sample assemblage to stable conditions. The non-polar character of the injected decane in Test 2 allows the assumption that the fluid does not interact with the electrostatically charged clay particles.

• Polar fluid (Test 3): Three friction experiments (Exp. 3.1–3.3) were performed by injection of a polar fluid (deionized water) applying the same general protocol as in Test 2. The polar character of the injected water caused interactions of the fluid with the clay minerals (e.g., Madsen & Müller-Vonmoos, 1989).

The effect of water-clay interactions on the stress state and the initiation of slip was estimated by comparing the stress- and strain data of the two injection friction tests (Test 2 and 3).

### 2.3. Test Apparatus and Processing of Macroscopic Mechanical Data

The triaxial friction experiments were performed at the Rock Physics and Mechanics Laboratory at ETH Zurich using the triaxial deformation test apparatus LabQuake (Figure S2 in Supporting Information S1). Confining pressure was applied inside a high-pressure triaxial cell using oil as confining medium. Far-field axial displacement $\Delta x_{FP}$ was measured by the piston position using a potentiometric transducer. Additionally, axial displacement $\Delta x_{LVDT}$ was measured using an axial linear variable differential transformer (LVDT). The LVDT was mounted inside the high-pressure triaxial cell, measuring axial deformation $\sim 40$ mm away from the sample assemblage, between $\sim 72$ mm above and below the sample (Figure 1b).

For the fluid injection friction tests, two external Teledyne ISCO 260D syringe pumps were used, one to inject decane (Test 2) and one to inject deionized water (Test 3). Before injecting fluids, the air was removed from the pore space of the BER using a vacuum pump. Fluids were injected through the lower platen into the BER while the pore-fluid outlet valve remained closed (Figure 1a). Due to the sealing capacity of OPA, the pore-fluid outlet through the upper platen remained air filled at approximately ambient conditions throughout the experiment. This was confirmed by (a) opening the pore-pressure outlet at the end of fluid injection experiments, which did not cause any fluid discharge, and (b) no macroscopic indications of fluid bypass or flow through the OPA on the post-mortem samples. Both mechanical data from LabQuake and hydraulic data from the ISCO pumps were recorded with a sampling rate of 1 Hz.

The normal stress $\sigma_N$ and shear stress $\tau$ on the slip surface were calculated by

$$\sigma_N = \frac{(\sigma_1 + \sigma_3) + (\sigma_1 - \sigma_3) \cos(2\theta)}{2},$$

and

$$\tau = \frac{(\sigma_1 - \sigma_3) \sin(2\theta)}{2},$$

respectively, where $\sigma_1$ is the axial stress, $\sigma_3$ the confining pressure ($P_c = \sigma_2 = \sigma_3$), $\sigma_1 - \sigma_3$ the differential stress $\sigma_D$, and $\theta$ the angle between the normal of the slip surface and the long axis of the piston-sample assembly (Figure 1a).

For fluid injection friction experiments (Test 2 and 3), the effective normal stress $\sigma'_N$ was calculated by

$$\sigma'_N = \sigma_N - P_f,$$
Figure 2. Corrected differential stress \( \sigma_{ij} \) (black) and estimated real slip distance \( s_{\text{app}} \) (brown) as a function of apparent slip distance \( s_{\text{app}} \) of Exp. 1.4 performed at different confining pressures \( P_c \) (black numbers below \( \sigma_{ij} \) curve). Thin black line shows non-smoothed ("raw") \( \sigma_{ij} \) data, bold black line shows smoothed \( \sigma_{ij} \) data (moving average, window length of 10 data points). Pink dots indicate peak differential stress \( \sigma_{ij}^{\text{max}} \), violet arrows indicate steady state range \( \sigma_{ij}'' \). Brown crosses indicate the estimated time of slip onset (based on moving slope with a window length of 15 data points and a threshold of 20 MPa/mm) defining the point after which the estimated real slip distance \( s_{ij} \) starts to count until the end of the according confining pressure stage.

where \( P_f \) is the fluid pressure (i.e., the injection pressure applied by the ISCO pump). Equation 3 is based on the effective stress law

\[
\sigma'_{ij} = \sigma_{ij} - \alpha \delta_{ij} P_f,
\]

where \( \sigma_{ij} \) is the stress tensor, \( \sigma'_{ij} \) is the effective stress tensor, \( \delta_{ij} \) is the Kronecker delta, and \( \alpha \) is the Biot's coefficient (for simplicity, we consider \( \alpha = 1 \), e.g., Paterson & Wong, 2005; Scholz, 2019). Axial displacement based on the piston position (\( \Delta x_{\text{pp}} \)) and based on an LVDT (\( \Delta x_{\text{LVDT}} \)) were corrected by subtracting the elastic shortening of the piston column and the platens, respectively. This can be expressed by

\[
\Delta x'_{m} = \Delta x_{m} - (F_{\text{axial}}/k_{m}) \quad \text{with} \quad m = PP \text{ or LVDT},
\]

where \( \Delta x'_{m} \) is the corrected axial displacement, \( \Delta x_{m} \) is the recorded axial displacement, \( F_{\text{axial}} \) is the applied axial load, and \( k_{m} \) is the stiffness correction factor. The latter depends on the confining pressure \( P_c \) and was calibrated for confining pressures of 5–160 MPa (Figure S3 in Supporting Information S1).

The apparent slip distance along the saw cut \( s_{\text{app}} \) was not measured directly but can be approximated based on trigonometric considerations by

\[
s_{\text{app}} = \frac{\Delta x'_{pp} \sin(\theta)}{\sin(\theta)}
\]

However, rigid body motion is assumed in Equation 6, that is, \( s_{\text{app}} \) also takes into account axial displacement not related to slip along the saw cut (e.g., elastic deformation of the rock matrix). To estimate the real slip distance, the time of slip onset was estimated based on a moving slope of the \( s_{\text{app}} \) versus \( \sigma_{ij} \) curve with a window length of 15
data points and a threshold of 20 MPa/mm (Figure 2). The first data point below the threshold defined the point in time from which the estimated real slip distance \( s_{\text{r}} \) started to count.

For experiments without fluid injection (Test 1), the axial stress \( \sigma_1 \) was corrected for decreasing contact area between the two sample halves with evolving slip (Scott et al., 1994) and for the jacket resistance (see Text S1 and Figure S4 in Supporting Information S1). Both decreasing contact area and jacket resistance correction depend on the amount of slip along the saw cut \( (s_{\text{r}}) \), which was several millimeters for the experiments without fluid injection (Test 1), while it was about an order of magnitude smaller for the fluid injection experiments (Test 2 and 3). These corrections would be below the noise level of the axial stress data for the fluid injection experiments and were therefore not applied.

2.4. Distributed Strain Sensing

Distributed strain sensing (DSS) was performed using fiber optics (FO) strain sensors described for laboratory applications by Salazar Vásquez et al. (2022). Polyimide-coated fibers with a cross-sectional diameter of 155 μm and a LUNA ODiSI 6104 interrogator were used. The latter was operated with two channels (i.e., two FO cables), a spatial resolution of 1.3 mm, and a sampling rate of 20 Hz. The LUNA ODiSI 6104 is an optical backscatter reflectometer that is based on Rayleigh backscattering occurring due to imperfections in the FO cable. By measuring changes in the backscattered light spectrum, the frequency shift \( \Delta f \) can be related to variation in strain \( (\Delta \varepsilon = \varepsilon - \varepsilon_0) \) and temperature \( (\Delta T = T - T_0) \) according to

\[
\Delta f = C_s \cdot \Delta \varepsilon + C_T \cdot \Delta T,
\]

where \( C_s \) and \( C_T \) are the strain and temperature coefficients, respectively. Assuming that the temperature remained stable and any thermal increase due to mechanical work was negligible, \( \Delta T \) was set to 0, which simplifies Equation 7 to

\[
\Delta \varepsilon = \frac{\Delta f}{C_s}.
\]

The strain coefficient of the polyimide fiber was calibrated to \( C_s = 0.15 \times 10^6 \) GHz before the experiments (Salazar Vásquez et al., 2022). From the time-strain \( (\Delta \varepsilon) \) data, the strain rate \( \dot{\varepsilon} \) was determined based on a moving slope with a window length of 2.5 s (50 time steps). If necessary, the time synchronization between DSS data and macroscopic mechanical data was adjusted manually based on a point in time with a distinct signal in both data sets, such as the beginning of axial loading.

The polyimide FO cables were glued to the sample surface (Figures 1b–1d) to measure surface strain during a decane injection friction experiment (Exp. 2.3) and an H₂O injection friction experiment (Exp. 3.2). One cable per sample half was used, each covering two vertical parts and a saw-cut-parallel part (Figure 1d). Along these regions of interest (ROIs), the cables were stretched by hand while gluing in order to provide that the strain of the sample is transferred to the cable when the sample is under compression (i.e., applied confining pressure and axial loading). Curves between the vertical and the saw-cut-parallel parts are for changing the direction only and strain transformation from the sample to the cable is not provided in these regions. Therefore, data from these curves are not analyzed. Note that the saw-cut-parallel cables are generally influenced by both axial and radial strain, with the axial contribution vanishing at the lowermost part of the cable (i.e., at the heel of the sample). Definition of the distance for the ROIs is shown in Figure 1d, where the beginning and the tip of the black arrows correspond to zero and the maximum distance, respectively. The first and last points of the ROIs were determined prior to the experiments by heating these points with a pen-tipped soldering iron, with extension due to thermal expansion indicating the positions along the cable for the heated points.

3. Results

3.1. Friction Test Without Fluid Injection

We conducted four multi-stage triaxial friction experiments without fluid injection (Exp. 1.1–1.4) under confining pressures ranging from 4 to 25 MPa (Table 1). Figure 2 shows the evolution of the differential stress \( (\sigma_D) \) as a function of apparent slip distance \( (s_{\text{app}}) \) for Exp. 1.4, which was performed under confining pressure stages of 4–
12 MPa (see also Table 1). During the first confining pressure stage \( (P_c = 4 \text{ MPa}) \), the continuous piston displacement caused a quasi-linear increase in differential stress, which then progressed to a peak differential stress \( (\sigma_{D0}^{pk}) \) before gradually decreasing to a steady state differential stress \( (\sigma_{D0}^{ss}) \). At higher confining pressure stages \( (P_c = 6–12 \text{ MPa}) \), there was an absence of peak differential stress, and the steady state regime consisted of a minor increase in differential stress (i.e., strain hardening). When reducing to lower confining pressure stages (e.g., from 12 to 8 MPa), peak differential stresses were observed again. However, the transition to the steady state regime took place over a smaller slip distance compared to the initial confining pressure stage (Figure 2). The experiment involved the confining pressure stages of 4, 6, and 8 MPa twice each. When these confining pressures were applied for the second time, that is, at a higher slip distance, higher steady state differential stresses were obtained compared to the first time. All experiments without fluid injection (Exp. 1.1–1.4) were characterized by the disappearance of a peak differential stress after the first confining pressure stage (at higher confining pressures), the reappearance of less pronounced peak differential stresses when going back to lower confining pressures, the occurrence of strain hardening in the steady state regime, and higher steady state differential stresses with increased slip distance but at equal confining pressure (Figure 2 and Figure S5 in Supporting Information S1). Exp. 1.1–1.3, which had an initial confining pressure of 10 MPa, exhibited a more pronounced decay from peak differential stress to steady state during the initial confining pressure stage, compared to Exp. 1.4 with an initial confining pressure of 4 MPa \( (\sigma_{D0}^{pk} - \sigma_{D0}^{ss} \sim 0.5 \text{ MPa}) \) to 1.6 MPa for \( P_c = 10 \text{ MPa} \) and \( \sigma_{D0}^{pk} - \sigma_{D0}^{ss} \sim 0.2 \text{ MPa} \) for \( P_c = 4 \text{ MPa} \). The most pronounced strain hardening in the steady state regime of about \( 1.9 \text{ MPa/mm} \) was observed in Exp. 1.3 at \( P_c = 25 \text{ MPa} \).

### 3.2. Fluid Injection Friction Tests

#### 3.2.1. Macroscopic Mechanical Data

Test 2 consists of three decane injection friction experiments, with Exp. 2.1 conducted under a confining pressure of 10 MPa, and Exp. 2.2 and 2.3 conducted under confining pressures of 10 and 25 MPa. The experiments can be separated into four phases, described hereafter for Exp. 2.3 (Figures 3a–3d) and in more details in Text S2 in Supporting Information S1.

During phase (I), the piston was displaced at a constant rate of \( 0.1 \text{ mm/min} \) until a differential stress of \( 11.1 \text{ MPa} \) \( (P_c = 10 \text{ MPa}) \) and \( 19.6 \text{ MPa} \) \( (P_c = 25 \text{ MPa}) \) was reached (Figures 3a and 3c; schematically represented in Figure 3e). The piston was then held at a constant position for phases (II) to (IV) by keeping the uncorrected far-field axial displacement, that is, \( \Delta x_{far} \), constant. During the initial loading stage (phase I), the differential stress increased quasi-linearly with increasing axial displacement. In phase (II), a waiting period was employed, during which the differential stress relaxed to a nearly constant state of \( \sim 9 \) and \( \sim 17 \text{ MPa} \) at confining pressures of 10 and 25 MPa, respectively. In phase (III), the fluid pressure \( P_f \) was increased step-wise up to 8 and 18 MPa for \( P_c = 10 \) and 25 MPa, respectively. It should be noted that the \( P_c = 25 \text{ MPa} \) part of the experiment followed the \( P_c = 10 \text{ MPa} \) part, and therefore loading (phase I) and relaxation (phase II) took place under a fluid pressure of 0.1 MPa. Based on the changes in differential stress and axial LVDT displacement in response to the fluid pressure changes, phase (III) can be further separated into two parts (Figures 3b and 3d, schematically represented in Figure 3f). Phase (III-a) includes lower fluid pressure steps, where the differential stress tended to remain constant or slightly decreased and the axial LVDT showed extension after imposing a higher fluid pressure. Phase (III-b) encompasses higher fluid pressure steps, in each of which the differential stress decreased and the axial LVDT indicated compression. The magnitude of the differential stress drop and the compressive axial displacement increased with higher fluid pressures. It has to be noted that the transition from phase (III-a) to (III-b) is not sharp, in particular at \( P_c = 25 \text{ MPa} \) (Figure 3d). In phase (IV), the fluid pressure was decreased from the highest value to 0.5 MPa, resulting in a differential stress decrease and an increase in axial LVDT (Figures 3a and 3c). The behavior during phases (I) to (IV) described above was consistently observed across all decane injection friction experiments (Figure 3 and Figure S6 in Supporting Information S1).

Three \( \text{H}_2\text{O} \) injection friction experiments were performed, with Exp. 3.1 and 3.2 under a confining pressure of 10 MPa and Exp. 3.3 under confining pressures of 10 and 25 MPa. The general test protocol was identical to the decane injection friction test, and the experiments can be separated into the same four phases (Figure 4). The results are described in the following based on Exp. 3.2 at \( P_c = 10 \text{ MPa} \) (Figures 4a and 4b) and Exp. 3.3 at
Figure 3. Macroscopic mechanical data of a decane injection friction experiment (Exp. 2.3) at confining pressures of 10 MPa (a, b) and 25 MPa (c, d). (a, c) Differential stress $\sigma_D$ (smoothed with moving average, window length of five data points), fluid pressure $P_{Dec}$ and axial displacement based on axial linear variable differential transformer (LVDT) $\Delta x_{LVDT}$ as a function of time. $P_{Dec}$ and $\Delta x_{LVDT}$ data are shown for phase (III) and (IV) only, that is, stages where fluid pressure changes were actually applied to the sample. Black circles indicate differential stress at stages of the experiment that define the frictional slip envelope ($\sigma_f^m$). (b, d) Differential stress $\sigma_D$ as a function of $\Delta x_{LVDT}$ for phase (III). Color code corresponds to fluid pressure $P_{Dec}$. Black circles correspond to the first data point after $P_{Dec}$ was increased, black numbers indicate the actual $P_{Dec}$ in MPa for points mentioned in Text S2 in Supporting Information S1. Dotted black lines indicate the slope defined by the piston relaxation factor PRF (Equation 9). Different behavior during phase (III) are indicated with (III-a) and (III-b), see text for further information. (e) Sketch illustrating phase (I), that is, axial
$P_c = 25$ MPa (Figures 4c and 4d), with a focus on differences with respect to the decane injection friction experiments. A more detailed description is provided in Text S3 in Supporting Information S1.

In Exp. 3.2 ($P_c = 10$ MPa), loading (phase I) was followed by a waiting period of 110 min (phase II) during which the differential stress decreased to a nearly constant value of $\sim$8 MPa (Figure 4a). The extended waiting period was due to technical issues that were resolved during this time. During phase (III), H$_2$O injection was performed applying the same protocol as for the decane injection experiments, except for partly different holding times. Phase (III) for H$_2$O injection friction experiments with initial fluid contact ($P_c = 10$ MPa) can be separated into three parts (Figure 4b, schematically represented in Figure 3f). Phase (III-init) encompasses the initial fluid injection with $P_{f,initial} = 0.1$ MPa. In comparison to the initial decane injection, where no significant change in differential stress and axial LVDT was observed, the initial injection of H$_2$O led to a differential stress decrease of $\sim$1 MPa within $\sim$2.5 min and an axial LVDT increase of $\sim$5 µm. In phases (III-a), (III-b), and (IV), the response to changes in fluid pressure was similar to the decane injection experiments (compare Figures 3a and 4a, and Figures 3b and 4b). Similar behavior during phases (I) to (IV) were observed across all H$_2$O injection friction experiments at $P_c = 10$ MPa (Figures 4a and 4b, and Figure S7 in Supporting Information S1), with the exception of Exp. 3.3 during phase (III) (Figures S7c and S7d in Supporting Information S1): The differential stress decreased notably during phase (III-a), the transition from phase (III-a) to (III-b) occurred at lower fluid pressures, and the differential stress decrease during phase (III-b) was associated with less axial displacement.

In Exp. 3.3, the H$_2$O injection friction experiment was additionally conducted at $P_c = 25$ MPa (Figures 4c and 4d). After loading (phase I), a waiting period was applied (phase II), during which the differential stress decreased to 16.7 MPa. In phase (III), the fluid pressure was incrementally increased using the same protocol as described for the decane injection experiments, except for partly different holding times. At $P_c = 25$ MPa, phase (III-a) was not observed. Instead, throughout phase (III), fluid pressure steps led to a decrease in differential stress and an increase in axial LVDT (Figure 4d), corresponding to phase (III-b) in the other experiments. With higher fluid pressures, the magnitude of differential stress drop and compressive axial displacement increased, reaching $\sim$0.8 MPa and $\sim$3 µm, respectively, when increasing the fluid pressure from 17.5 to 18 MPa.

For both decane and H$_2$O injection experiments (Tests 2 and 3, respectively), the piston position, that is, $\Delta x_{pp}$, was kept constant during phase (III) (constant piston displacement control, e.g., Ye & Ghassemi, 2018). Assuming the elastic properties of the sample assemblage remained constant during phase (III), the corrected axial LVDT $\Delta x_{LVDT}^{corr}$ theoretically quantifies the amount of relaxation or compression of the piston assembly resulting from a differential stress decrease or increase, respectively (Figure S8 in Supporting Information S1). To estimate the expected differential stress decrease related to a unit of piston relaxation measured by the axial LVDT, we calculated the piston relaxation factor $PRF$, that is, the negative piston stiffness as a function of axial stress, according to

$$PRF = -\frac{k_{pp}}{A_c}, \quad (9)$$

where $k_{pp}$ is the stiffness correction factor used to correct the axial position (Figure S3a in Supporting Information S1) and $A_c$ is the circular area of the cylindrical sample. The slope defined by the piston relaxation factor was plotted with different intercepts in Figures 3b, 3d, 4b, and 4d, and Figures S6b, S6d, S6f, S7b, and S7d in Supporting Information S1. The axial displacement versus differential stress curve at phase (III-b) tends to be subparallel to the $PRF$ slope, particularly at high fluid pressures. This indicates that the displacement measured with the axial LVDT during phase (III-b) is governed by the stiffness of the piston assembly and is not a direct measure of the amount of axial deformation that took place in the sample assemblage. For experiments where the experimental data deviate from the $PRF$-defined slope, particularly Exp. 3.3 at $P_c = 10$ MPa (Figure S7d in Supporting Information S1), we hypothesize that this is due to measurement issues with the axial LVDT, due to a change in the bulk elastic modulus of the sample assemblage (e.g., a decreasing bulk Young’s modulus would require a lower differential stress at constant axial deformation), or due to competing deformation processes that

loading by constant axial position displacement. Represents both, decane and H$_2$O injection friction tests. (f) Sketch showing changes in axial LVDT and differential stress data for phase (III). Phase (III-a) and (III-b) represent both, decane, and H$_2$O injection friction tests. Phase (III-init) represents H$_2$O injection friction test only (see Figure 4).
Figure 4. Macroscopic mechanical data of H₂O injection friction experiments at confining pressures of 10 MPa (a, b, Exp. 3.2) and 25 MPa (c, d, Exp. 3.3), see Figures 3e and 3f for schematic representation of the data. (a, c) Differential stress $\sigma_d$ (smoothed with moving average, window length of five data points), fluid pressure $P_f^{\text{HDO}}$, and axial displacement based on axial linear variable differential transformer $\Delta x_{LVDT}$ as a function of time. $P_f^{\text{HDO}}$ and $\Delta x_{LVDT}$ data are shown for phase (III) and (IV) only, that is, stages where fluid pressure changes were actually applied to the sample. Black dot indicates differential stress at initial H₂O contact ($\sigma_d^{\text{init}}$) and black circles the differential stress at stages of the experiment that define the frictional slip envelope ($\sigma_d^{f}$). Parts of phase (II) in subfigure (a) are not shown (data between 1,000 and 6,000 s). (b, d) Differential stress $\sigma_d$ as a function of $\Delta x_{LVDT}$ for phase (III). Color code corresponds to fluid pressure $P_f^{\text{HDO}}$. Black circles correspond to the first data point after $P_f^{\text{HDO}}$ was increased, black numbers indicate the actual $P_f^{\text{HDO}}$ in MPa for points mentioned in Text S3 in Supporting Information S1. Dotted black lines indicate the slope defined by the piston relaxation factor $PRF$ (Equation 9). Different behavior during phase (III) are indicated with (III-init), (III-a), and (III-b), see text for further information.

3.2.2. Distributed Strain Sensing Data

DSS data were acquired for a decane injection friction experiment (Exp. 2.3) and a H₂O injection friction experiment (Exp. 3.2). Figure 5 shows strain-rate data of the decane injection experiment when the fluid pressure was increased from 1.0 to 1.5 MPa (phase III-a in Figure 3b) for 5 s before and 15 s after the beginning of the fluid-pressure increase. Note that portions of the data are not shown in Figure 5 for visualization purposes, but the full data set of the same time period is shown in Figure S9 in Supporting Information S1.
The DSS data show that once the fluid pressure was increased, the BER exhibited relatively homogeneous extensional strain rate along both, vertical and saw-cut-parallel parts of the cable (Figures 5a, 5c, and 5d). At the same time, the OPA was compressed in vertical direction and showed relatively homogeneous extensional strain rate parallel to the saw cut but with a lower magnitude than in the BER (Figures 5b, 5c, and 5d). The data of the second vertical parts of the cables (Figure S9 in Supporting Information S1) are not significantly different to the data shown in Figures 5a and 5b. Data for the same fluid-pressure step but for the H₂O injection experiment are shown in Figure S10 in Supporting Information S1, demonstrating similar behavior as seen in the decane injection experiment. For all the presented DSS data, it should be noted that sharp changes in the strain rate may appear to begin earlier and last longer than they actually do, due to the applied moving slope with a window length of 50
time steps, that is, 2.5 s. As a consequence, time discrepancies in the order of ~1 s may occur when comparing the DSS data to the macroscopic mechanical data.

Figure 6 shows the DSS data of the decane injection experiment when the fluid pressure was increased from 7.5 to 8.0 MPa (phase III-b in Figure 3b) for 5 s before and 15 s after the beginning of the fluid-pressure increase. The complete data set for this time period is shown in Figure S11 in Supporting Information S1.

The DSS data for this fluid-pressure increase can be separated into two phases. At time $t_1$, the BER showed relatively homogeneous extensional strain rate parallel to the vertical part of the cable (Figures 6a and 6d), whereas the OPA showed relatively homogeneous compressive strain rate (Figures 6b and 6d). At the same time, the strain rate along the saw-cut-parallel parts of the cables was heterogeneous and non-symmetrically distributed.
(Figures 6c and 6d), with the BER exhibiting almost neutral (zero) strain rate between about 0.041 and 0.079 m and extensional strain rate with slightly varying magnitude along the other portion of the saw-cut-parallel part of the cable. In contrast, the OPA showed compressive strain rate between about 0.006 and 0.040 m and between about 0.151 and 0.199 m, and extensional strain rate between about 0.040 and 0.070 m. Also, the strain rate at the heel of the OPA, where the cable is sub-horizontal, was slightly extensional.

Between times $t_1$ and $t_2$, the magnitude of the strain rate increased relatively homogeneous along the vertical part of the cable on the BER, while the strain rate along the vertical part on the OPA transitioned into relatively homogeneously distributed extensional strain rate. The saw-cut-parallel parts of the cables at time $t_2$ showed neutral and slightly compressive strain rates at the heels of the BER and OPA, respectively, and relatively homogeneous extension in the rest of the saw-cut-parallel part of the cables. When comparing the DSS data to the macroscopic mechanical data, time $t_1$ falls within the period where the fluid pressure was increased, and time $t_2$ marks the beginning of the differential stress decrease and $\Delta \nu^\text{ref}_{LVDT}$ increase. The second vertical parts of the cables exhibited similar behavior for both the BER and OPA (Figure S11 in Supporting Information S1). Figure S12 in Supporting Information S1 shows the DSS data for the water injection experiment during the same fluid-pressure increase (from 7.5 to 8.0 MPa), which revealed behavior similar to the decane injection experiment.

Figure S13 in Supporting Information S1 shows the DSS data of the decane injection experiment when decane was initially injected at a fluid pressure of 0.1 MPa for 50 s before and 150 s after the beginning of injection. Extensional strain rate that migrates upward was observed in the vertical and saw-cut-parallel parts of the cable on the sandstone and in the saw-cut-parallel part of the cable on the OPA, indicating the migration of the fluid front. On the other hand, complex processes took place when deionized water was initially injected at a fluid pressure of 0.1 MPa (phase III-init in Figure 4b), indicated by the DSS data 50 s before and 150 s after the beginning of injection shown in Figure 7. The complete data set of this time period is shown in Figure S14 in Supporting Information S1.

The behavior during the initial $\text{H}_2\text{O}$ injection is illustrated based on the four discrete times $t_1$ to $t_4$. At time $t_1$, the vertical part of the cable on the BER showed extensional strain rate over about 1–2 mm (Figures 7a and 7d), which migrated upward with a rate of about 0.6 mm/s, and exceeded the uppermost point of the vertical part of the cable between time $t_2$ and $t_3$. In contrast, the vertical part of the cable on the OPA showed a relatively homogeneous extensional strain rate at time $t_1$ (Figures 7b and 7d). Along the saw-cut-parallel cables, a compression-extension pattern on the BER and the opposite pattern on the OPA migrated upward from time $t_1$ to $t_3$ with a rate in the order as described above for the extensional strain rate along the vertical part of the cable on the sandstone (Figures 7c and 7d). The compressive part in the saw-cut-parallel part of the cable on the BER was interrupted by an extensional strain rate over a distance of about 1–3 mm.

The vertical part of the cable on the BER at time $t_2$ showed compressive strain rate between the lowermost point and the extensional part described for time $t_1$. From time $t_2$ to $t_3$, the compressive strain rate tended to extend over a longer part of the cable and migrated upward (Figures 7a and 7d). The vertical part of the cable on the OPA showed an upward migrating compression-extension pattern at time $t_2$ (Figures 7b and 7d). Between time $t_2$ and $t_3$, extensional strain rate was again observed at the bottom of the vertical part of the cable, which extended upward with evolving time. Around time $t_4$, relatively homogeneous extensional strain rate took place in vertical direction in both the BER and OPA (Figures 7a, 7b, and 7d). At the same time, the saw-cut-parallel parts of the cables showed a slightly extensional strain rate at the uppermost part of the cable on the BER, extensional strain rate at the heel of the OPA, and compressive strain rate along the other part on the OPA (Figures 7c and 7d). Comparing the DSS and macroscopic mechanical data, a first change in the strain rate based on the DSS data occurred about 9 s after the beginning of $\text{H}_2\text{O}$ injection (Figures 7a and 7b). Time $t_1$ corresponds roughly to the beginning of differential stress decrease, and the increase in $\Delta \nu^\text{ref}_{LVDT}$ appears to be related to the process described for time $t_4$. Note that a small drop in fluid pressure prior to time $t_4$ is related to the technical operation of the pump rather than to processes in the sample assemblage.

### 3.3. Post-Mortem Sample Characterization

Figure 8 shows the OPA sample surfaces after an experiment without fluid injection, a decane injection experiment, and an $\text{H}_2\text{O}$ injection experiment. The surfaces are represented by color maps based on the gray-scale intensity of photographs. The original photographs are provided in Figure S15 in Supporting...
Figure 7. Strain rate based on distributed strain sensing data of a H$_2$O injection friction experiment (Exp. 3.2) during initial fluid injection at ambient fluid pressure ($P_f = 0.1$ MPa). Inlets indicate fiber optics (FO) cable location of corresponding subfigure with black arrow(s) (see also Figure 1b–1d). Data of additional parts of FO cables are shown in Figure S14 in Supporting Information S1. (a) Upper panel: Non-smoothed macroscopic mechanical data (corresponding to right y-axes of subfigure b), fluid pressure and axial linear variable differential transformer data are shown only after fluid injection started; Lower panel: Strain rate in a time versus distance plot for a vertical part of FO cable on Berea sandstone (BER) (colorbar in subfigure c). (b) Upper panel: Macroscopic mechanical data as in subfigure (a); Lower panel: Strain rate in a time versus distance plot for a vertical part of FO cable on Opalinus claystone (OPA) (colorbar in subfigure c). (c) Strain rate in a distance versus time plot for
Information S1 together with the corresponding BER surfaces. Curved linear structures were present in the OPA saw-cut surface after experiments without fluid injection, which result from imprinting the saw grooves of the BER saw-cut surface (i in Figure 8b, see also Figures S15a and S15b in Supporting Information S1). Furthermore, slickenlines oriented sub-parallel to the expected sliding direction were observed (ii in Figure 8c).

After dismantling the sample assemblage of decane injection friction experiments, decane evaporated from the OPA saw-cut surface within minutes, leading to brighter and darker patches where more or less decane evaporated, respectively (iv and v in Figure 8d). The relatively fast drying of the OPA saw-cut surface indicates that the fluid did not penetrate deep into the rock. Slickenlines sub-parallel to the expected sliding direction were also observed after the decane injection friction experiments (v in Figure 8e), but they were less pronounced than in the experiments without fluid injection and not recognizable throughout the surface. Furthermore, material appeared to have been trailed during slip, leading to linear structures at the end of accumulated material striking sub-perpendicular to the expected sliding direction (vi in Figure 8e).

After H₂O injection friction experiments, the uppermost layer of the OPA saw-cut surface was an incohesive mud (Figure 8f and Figure S15e in Supporting Information S1). It should be noted that the increased stickiness of the mud led to clay material sticking to the BER when separating the two sample halves (Figure S15f in Supporting Information S1), which may have altered the OPA saw-cut surface prior to photography. Darker colors close to the saw cut relative to the rest of the OPA indicated water penetration of a few millimeters. Slickenlines were not observed after H₂O injection friction experiments. Instead, material appeared to have been trailed during slip, which led to linear structures (steps) at the end of the accumulated material striking sub-perpendicular to the expected sliding direction (viii in Figure 8g). These trailing structures were more pronounced than after the decane injection experiments.

After experiments without fluid injection and H₂O injection friction experiments, intersection lineations were observed resulting from the intersection between foliation parallel cracks and the saw-cut surface (ii in Figure 8b and ix in Figure 8g).

4. Discussion

4.1. Frictional Properties of the Non-Wetted Interface

The objective of the friction experiments without fluid injection was to comprehend the frictional characteristics of the non-wetted OPA-BER interface. We have observed pronounced stress drops from peak to steady state stress at the first confining pressure stage of an experiment (Figure 2 and Figure S5 in Supporting Information S1). These observations are in agreement with direct shear experiments on OPA gouge by Orellana et al. (2018b), which displayed similar stress drops at comparable normal stresses. While such stress drops may be related to over-consolidation (e.g., shown for OPA by Wild & Amann, 2018), this appears not to explain our results as the stress drop is less pronounced at Pₑ = 4 MPa (over-consolidated) compared to the stress drops at Pₑ = 10 MPa (closer to normal consolidated). Rather, these stress drops are related to the energy density dissipated during strength loss with evolving strain, defined as the breakdown work (Cocco et al., 2023; Tinti et al., 2005). Our experiments show that breakdown work was highest during the initial confining pressure stage, indicating smoothing of asperities by smearing, while breakdown work was negligible in subsequent stages. The reappearance of minor peak stresses appears to be related to axial unloading prior to confining pressure decreases, which was not applied prior to confining pressure increases. Throughout the experiments without fluid injection, we observed strain hardening, which is typically associated with work dissipated for microstructural evolution and is a prevalent phenomenon in clay-rich materials such as OPA (e.g., Haines et al., 2013; C. A. Morrow et al., 1982, 2017; Orellana et al., 2019; Schuster et al., 2023).

saw-cut-parallel part of FO cable on OPA (left and right part) and on BER (middle part). Strain for two discrete points at distance d₁ and d₂ is shown in Figure 11. (d) Sketch of strain rate on cylindrical sample at time t₁ to t₂ (indicated with dashed lines in subfigures a–c). Blue bold lines indicate compressive strain rates, green, and red bold lines indicate extensional strain rates in BER and OPA, respectively.
Figure 8. (a) Sketch indicating location, orientation, and relative movement of Opalinus claystone (OPA) saw-cut surface (blue ellipse) and Berea sandstone (BER) saw-cut surface (black dotted ellipse). (a–g) Color maps generated based on gray-scale intensity of photos taken from OPA saw-cut surfaces after a (b, c) friction experiment without fluid injection (Exp. 1.4), (d, e) decane injection friction experiment (Exp. 2.2), and (f, g) H₂O injection friction experiment (Exp. 3.2). The colorbar in subfigure (b) indicates relative intensities and is valid for all color maps. Circles in subfigures (a, d, and f) indicate position of 2x magnified cutouts shown in subfigures (c, e, and g). Roman numbers refer to white markings on the photos: (i) imprints of BER surface, (ii) intersection between saw cut and foliation, (iii) slickenlines, (iv) brighter patch, (v) darker patch, (vi) steps from material accumulation, (vii) slickenlines, (viii) steps from material accumulation, (ix) intersection between saw cut and foliation.

Figure 9a shows peak and steady state stress data of the experiments without fluid injection in a Mohr plot. The steady state data were used to determine the non-wetted frictional slip envelope of the BER-OPA interface based on the linear model (i.e., Coulomb failure envelope)

\[ \tau = \mu_f \sigma_N + S_0 \]  

and the exponential model

\[ \tau = (\mu_f \sigma_N + S_0) e^{-\beta \sigma_N}, \]

as applied in previous studies on the mechanical properties of OPA (e.g., Minardi et al., 2021; Orellana et al., 2018b; Winhausen et al., 2022). For the linear model, \(\mu_f\) is the friction coefficient and \(S_0\) is the inherent shear strength (Lockner & Beeler, 2002). In the exponential model, the friction coefficient \(\mu_f\) and inherent shear strength \(S_0\) provide an approximation of the initial slope and intercept of the failure envelope (i.e., at \(\sigma_N = 0\), respectively, and the exponential fitting parameter \(\beta\) controls the extent of slope decrease (weakening) with increasing normal stress. The linear model revealed a friction coefficient of 0.35 (Figure 9b and Table 2), which is consistent with previous studies using OPA gouge (Bigaroni et al., 2023; Orellana et al., 2018b) and intact OPA samples.
Figure 9. (a) Peak and steady state data of friction experiments without fluid injection (Exp. 1.1–1.4) in a normal stress $\sigma_N$ versus shear stress $\tau$ plot. Legend in lower right panel. Mohr circles are shown for the steady state data only. Non-wetted frictional slip envelope for the steady state case is shown based on an exponential fit, shaded gray area corresponds to the 95% prediction interval. Two reference slopes (dashed gray lines) are shown for comparison. (b) Linear fit for steady state data (red), shaded red area corresponds to the 95% prediction interval. Exponential fit (black and shaded gray) and two reference slopes (dashed gray lines) are shown for comparison.

(Marschall & Giger, 2016; Minardi et al., 2016), whereas the inherent shear strength of 1.5 MPa in our study is relatively high (Orellana et al., 2018b). The low observed frictional strength, that is, much less than Byerlee's law (Byerlee, 1978), in any case indicates that the OPA dictates the strength of the two-material interface. This is in agreement with past research linking clay minerals to reduced frictional strength (e.g., Crawford et al., 2008; Ikari et al., 2007; Kohli & Zoback, 2013; Saffer & Marone, 2003; Saffer & Tobin, 2011; Tembe et al., 2010).

The exponential model revealed an initial friction coefficient of 0.48, an initial inherent shear strength of 0.5 MPa, and an exponential fitting parameter of 0.008 MPa$^{-1}$ (Figure 9a and Table 2). At normal stresses up to 40 MPa, the exponential frictional slip envelope lies within the 95% prediction interval of the linear one and vice versa (Figure 9b). Nevertheless, we favor the exponential model because it has been suggested in previous studies for claystones (e.g., Petley, 1999; Winhausen et al., 2022) and the lower inherent shear strength better reflects the predefined slip surface of the saw-cut set-up. We further assume that the steady state frictional slip envelope also serves as a yielding criteria for initial slip. This appears to be justified since the peak stress data fall within the 95% prediction interval of the exponential fitting curve and the differences in peak and steady state stresses of an individual experiment are relatively small compared to the scatter of the data across different experiments (Figure 9a).

4.2. Fluid-Induced Fault Slip

This section discusses fluid-induced fault slip and focuses on the mechanical response during fluid-pressure increases (phases III-a and III-b), which is similar for the injection of decane (Test 2) and H$_2$O (Test 3). The behavior during the initial H$_2$O injection (phase III-init), on the other hand, is discussed in Section 4.4.
Phase (III-a) occurred at fluid pressures at which the stress state remained stable (i.e., no slip occurred). The \( \Delta T_{W/D} \) decreases indicate poro-elastic extension each time the fluid pressure was increased (Figures 3 and 4, Figures S6 and S7 in Supporting Information S1). This is supported by the DSS strain-rate data, which showed extensional strain in the BER in all measured directions as a response to a fluid-pressure increase (Figure 5, Figures S9 and S10 in Supporting Information S1). The extensional strain rate in the BER occurred almost uniformly over time, indicating that the change in fluid pressure occurred at a higher rate than temporally resolvable in the presented data. Extensional strain rate was also observed in the saw-cut-parallel cable on the OPA showing that the fluid (both decane and \( \text{H}_2\text{O} \)) penetrated the OPA close to the interface. This is also evident from the post-mortem sample characterization, particularly after \( \text{H}_2\text{O} \) injection. The axial compressive strain rate on the OPA is interpreted as the response of the non-fluid penetrated matrix to the poro-elastic extension in the BER. It should be noted that the expected differential stress increase due to poro-elastic extension competes with differential stress relaxation that may extend beyond phase (II) (Figures 3a, 3c, 4a, and 4c). Stress relaxation by creep is expected to occur predominantly in the OPA (Naumann et al., 2007; Schulze, 2011), where creep may be enhanced with increasing relative humidity (Liu et al., 2018) and could be driven by deformation along intergranular water films (C. Zhang & Rothfuchs, 2004). Therefore, we interpret the absence of the stable phase (III-a) in the water injection experiment at \( P_f = 25 \) MPa (Figures 4c and 4d) to be a result of enhanced creep due to the transition of the OPA interface into an incohesive mud (Figures 8f and 8g, and Figure S15e in Supporting Information S1).

Phase (III-b) occurred at fluid pressures under which the stress state was unstable (i.e., slip took place), where slip was indicated by a differential stress drop and axial compression at each fluid-pressure increase (Figures 3 and 4, Figures S6 and S7 in Supporting Information S1). The DSS strain-rate data provide additional information on the slip initiation suggesting that two mechanisms took place (Figure 6, Figures S11 and S12 in Supporting Information S1). At time \( t_1 \), the strain rate along the axial cables is similar as for fluid-pressure increases where no slip was initiated, indicating poroelastic extension in the BER. Along the saw-cut-parallel cables, the strain rate was not homogeneously extensional, as would be expected for purely poroelastic extension. In contrast, the strain rate pattern at time \( t_1 \) suggests the presence of interlocked segments while pre-slip or creep occurred on other segments (Cebry et al., 2022; X. Wang et al., 2023). At time \( t_2 \), the axial extension in both BER and OPA indicates elastic relaxation of the rock matrix as slip takes place. This is supported by the saw-cut-parallel cables, which showed extensional strain rates where the cables are sub-parallel to the saw-cut dip (i.e., dominated by axial strain). The opposite was observed where the cables are sub-horizontal, as expected for axial relaxation according to Poisson’s ratio. Despite limitations in temporal synchronization between DSS and macroscopic mechanical data
Figure 10. (a) Frictional slip envelopes based on decane injection experiments (Exp. 2.1–2.3) in an effective normal stress $\sigma_n$ versus shear stress $\tau$ plot. Non-wetted frictional slip envelopes and two reference slopes are shown for comparison. Except for Exp. 2.3 at confining pressure $P_c = 10$ MPa, only data points defining the frictional slip envelope ($\sigma_n^{\text{eff}}$ vs. $\tau^\text{eff}$) are shown. For Exp. 2.3 at confining pressure $P_c = 10$ MPa, data points defining stable stress state ($\sigma_n^{\text{eff}}$ vs. $\tau^\text{eff}$) and transitional stress state ($\sigma_n^{\text{trans}}$ vs. $\tau^\text{trans}$) are shown. (b) Frictional slip envelopes based on H$_2$O injection experiments (Exp. 3.1–3.3) in an effective normal stress $\sigma_n$ versus shear stress $\tau$ plot. Non-wetted frictional slip envelopes and two reference slopes are shown for comparison. Only data points that define the frictional slip envelope ($\sigma_n^{\text{eff}}$ vs. $\tau^\text{eff}$) are shown. Colorbar indicating fluid pressures is shown in subfigure (a). Stars indicate stress state at initial contact with water ($P_{\text{water}} = 0.1$ MPa, colors not related to colorbar). (c) Sketch showing shift of Mohr circle with increasing fluid pressure $P_f$ without reaching the frictional slip envelope (stable stress state). (d) Sketch showing shift of Mohr circle with increasing fluid pressure $P_f$ when the frictional slip envelope is reached. This leads to slip and a differential stress decrease, achieving stable conditions again. (e) Sketch indicating stress state at initial contact with H$_2$O for Exp. 3.3, where the data point lies on the wet frictional slip envelope defined by data points at higher fluid pressures. As the yielding criteria is reached, a differential stress decrease occurs until a stable stress state is achieved. (f) Sketch indicating stress state at initial contact with H$_2$O for Exp. 3.1 and 3.2. Differential stress decrease occurs although the Mohr circle does not touch the wet frictional slip envelope defined by data points at higher fluid pressures.

(Section 3.2.2), the macroscopic mechanical data support a pre-slip stage at time $t_1$ with no stress drop and a slip stage at time $t_2$ with decreasing differential stress and increasing axial LVDT (Figures 6a and 6b).

4.3. Frictional Properties of the Wet Interface

To estimate the frictional properties of the wet interface, we calculated the initial stress state at the beginning of each fluid-pressure stage as effective normal stress and shear stress at the frictional interface (Equations 1–3). For the decane injection friction Exp. 2.3, the stress data of all fluid-pressure stages are plotted in a Mohr plot (Figure 10a), while for the other fluid injection experiments only the stress data defining the frictional slip
envelope are shown (Figures 10a and 10b). At low fluid pressures (phase III-a), increasing fluid pressures resulted in the Mohr circle being shifted to lower normal stress without touching the frictional slip envelope, that is, the stress state was stable ($\sigma_N^{\nu}/$ vs. $\tau^\nu$ in Figure 10a, schematically shown in Figure 10c). At higher fluid pressures (phase III-b), the Mohr circle was shifted to lower normal stress as the fluid pressure was increased, so that it touched (or crossed) the frictional slip envelope. The occurrence of slip was accompanied by a shortening of the sample assemblage and a decrease in differential stress, reducing the size of the Mohr's circle until a steady state was achieved (Ye & Ghassemi, 2018). This mechanism is described schematically in Figure 10d. With further fluid-pressure increments, slip was initiated at progressively lower differential stresses, providing data points to estimate the frictional slip envelope of a single experiment (Figure S16 in Supporting Information S1; Rutter & Hackston, 2017). These data points are denoted as $\sigma_N^{\nu}/$ and $\tau^\nu$ in Figures 10a and 10b. However, a “transitional” stress state occurred between stable and unstable stress states ($\sigma_N^{\nu}/$ vs. $\tau^\nu$ in Figure 10a), where slip was indicated by decreasing differential stress and increasing axial LVDT, but the stress states did not define a linear relationship together with the distinctly unstable fluid-pressure stages. Note that the method for deriving the friction slip envelopes of the wetted interface is based on the assumption that any frictional weakening due to the presence of fluid occurs when the interface is initially wetted or at low fluid pressures (phase III-init or III-a), while at higher fluid pressures (phase III-b) the incrementally increasing fluid pressure only changes the effective stress (i.e., $\sigma_N^{\nu}/$) and not the frictional strength.

Coulomb frictional slip envelopes (Equation 10) were defined for all decane and H$_2$O injection friction experiments based on linear regression using the $\sigma_N^{\nu}/$ versus $\tau^\nu$ data (Figures 10a and 10b, Table 2). For the fluid injection experiments, we chose to use linear (i.e., Coulomb) rather than exponential frictional slip envelopes. This because the deviation from linearity would be minor over the relatively small range of effective normal stress studied in Test 2 and 3, and because it allows a better comparability with previous studies. The wet frictional slip envelopes based on the decane injection experiments (Exp. 2.1, 2.2, and 2.3) revealed friction coefficients of 0.35, 0.44, and 0.46 and inherent shear strengths of 1.87, 0.81, and 0.87 MPa, respectively (Figure 10a and Table 2). Our experiments do not support a significant change in frictional strength in the presence of decane comparing to the non-wetted interface. This is indicated by similar friction coefficients and inherent shear strengths for the decane-wetted and non-wetted interface (Table 2), where the range of possible values for the non-wetted case can be approximately defined between the exponential model at a normal stress of zero (the steepest possible envelope) and the linear model (the envelope that suitably describes the data at normal stresses of about 15–20 MPa). Furthermore, all data points defining the wet frictional slip envelopes based on the decane injection experiments lie within the 95% prediction interval of the non-wetted frictional slip envelope.

The wet frictional slip envelopes based on the H$_2$O injection experiments (Exp. 3.1, 3.2, and 3.3) resulted in friction coefficients of 0.37, 0.30, and 0.30 and inherent shear strengths of 0.92, 1.04, and 0.25 MPa, respectively (Figure 10b and Table 2). The friction coefficients are higher than reported by Orellana et al. (2019) for wet OPA, while our range of inherent shear strength includes their result. The data points defining the frictional slip envelope of Exp. 3.1 and 3.2 are within the uncertainty of the non-wetted frictional slip envelope, while those of Exp. 3.3 are progressively lower toward higher effective normal stresses. In two out of three H$_2$O injection experiments, the friction coefficients are lower compared to the decane and non-wetted frictional slip envelopes, while the inherent shear strengths are in the same range (Table 2). Although our data may be affected by sample variability, frictional weakening in the presence of water is further indicated by the stress state at initial H$_2$O contact in Exp. 3.3, which is stable under non-wetted conditions (prior to fluid injection) albeit within the range of the frictional slip envelope determined for the wetted interface (Figures 10b and 10e). Overall, H$_2$O tends to weaken the OPA-BER interface, which does not appear to be the case for decane, showing that the polar character of H$_2$O may influence the frictional strength. This is in agreement with previous studies, which found that the adsorption of polar fluids between negatively charged phyllosilicate sheets reduces the attracting force between them and thus reducing the shear resistance (e.g., Moore & Lockner, 2004; C. A. Morrow et al., 2000; Ikari et al., 2007; Scholz, 2019). The macroscopic observation of the OPA surface showed the presence of striations after experiments without fluid injection and decane injection experiments (Figures 8b–8e), similar to what was observed after friction experiments in which OPA was used for both the footwall and hanging wall (Schuster et al., 2023). The observed trailed material after the H$_2$O injection friction experiments (Figure 8g) shows that the presence of H$_2$O alters the interface to behave like a hydroplastic fault (Doblas, 1998; Petit & Laville, 1987), causing a rheological change that may weaken the surface in addition to phyllosilicate lubrication.
4.4. Hydro-Mechanical Response During Initial Claystone Hydration

This section discusses processes occurring during initial hydration of the OPA, that is, at the first contact of water with the OPA saw-cut interface (phase III-init). Note that these processes are restricted to the $H_2O$ injection friction experiments (Exp. 3.1–3.3). The macroscopic mechanical data suggest the initiation of slip once $H_2O$ was injected, which is indicated by the differential stress drop and increase in axial LVDT similar to that observed at higher fluid pressures (phase III-b) but with higher magnitudes (Figures 4a and 4b). However, the DSS strain-rate data suggest the involvement of further physical mechanisms compared to fluid-induced slip (compare Figures 6 and 7). On the other hand, it should also be taken into account that these processes may manifest more clearly due to their slower progression.

The extensional strain-rate pattern migrating upward along the vertical cable on the BER indicates the fluid front (Figures 7a and 7d), which agrees with the similar pattern in the decane injection experiment that migrates upward with a similar rate (Figures S13a and S13b in Supporting Information S1). In the saw-cut-parallel cable on the BER, we interpret the position of the fluid front based on the 1–3 mm interruption of the compressive part (Figures 7c and 7d) that also migrates upward on a similar time scale as the fluid front in the decane injection experiment (Figure S13c in Supporting Information S1). Similarly, the upward migration of the compression-extension pattern on the OPA and the opposite pattern on the BER appears to be governed by the migration of the fluid front (see time $t_2$ and $t_3$ in Figure 7). This leads to two possible interpretations for processes observable in the DSS data at around time $t_2$ and $t_3$.

1. The OPA hydration rapidly reduces the strength up to the failure criteria and the upward migration of this weak zone can be considered as (slow) propagation of a damage zone similar to a shear rupture front. Accordingly, the OPA experiences fault-parallel extension ahead of the propagation front and compression behind, while the opposite takes place in the BER, as described by X. Wang et al. (2023) for rupture initiation based on numerical experiments. Figure 11 shows the strain based on DSS data of the saw-cut-parallel cables for two discrete points as a function of time (data of more points shown in Figure S17 in Supporting Information S1). The propagation of the saw-cut-parallel strain peaks (BER) and troughs (OPA) qualitatively agrees with off-fault strain measured in laboratory experiments simulating earthquake rupture front propagation (Svetlizky & Fineberg, 2014; Xu et al., 2019), although the propagation velocity in our experiment is at least six orders of magnitudes slower. Nevertheless, the mechanics used to explain these ruptures are still applicable to slow ruptures in laboratory tests (Selvardurai et al., 2017) and are also proposed to explain large-scale slow earthquakes on natural faults (e.g., Bartlow et al., 2011). In our test, the residual saw-cut-parallel strain after rupture propagation in the OPA was similar or higher than the initial value, while the residual strain in the BER was lower (Figure 11 and Figure S17 in Supporting Information S1). This different behavior may be partly due to irreversible (inelastic) deformation of the OPA between the saw-cut-parallel cable and the saw cut as it is transformed into an incohesive mud.

Figure 11. Strain as a function of time based on distributed strain sensing data of an $H_2O$ injection friction experiment (Exp. 3.2) during initial fluid injection. Data shown for two discrete points along the saw-cut-parallel fiber optics cables on the Opalinus claystone and Berea sandstone, location of discrete points at distance $d_1$ and $d_2$ indicated in Figure 7 and qualitatively in top right inlet (black and gray dots). Data for further points along the saw-cut-parallel cable shown in Supporting Figure S17 in Supporting Information S1.
Figure 12. Estimation of swelling stress required so that the normal and shear stress lie on the frictional slip envelope, assuming a swelling stress anisotropy of 0.02 (gray) and 0.42 (black) according to Thury and Bossart (1999) and Bossart and Thury (2008), respectively. For frictional slip envelopes and apparent effective normal stress and shear stress (stars) see also Figure 10b. (a) Exp. 3.1. (b) Exp. 3.2.

2. An alternative interpretation could be that the water-clay interaction during hydration leads to swelling and thus extension in the OPA approximately at the position of the fluid front. The build-up of swelling stress in turn results in local microfracturing (Voltolini & Ajo-Franklin, 2020; C. L. Zhang, 2011) and breaking of diagenetic bonds (Delage & Tessier, 2021) that may lead to a transition from a solid to a locally incohesive material (Corkum & Martin, 2007), causing compression behind the fluid front by local compaction. The opposite strain-rate pattern on the BER can be understood as a response to the processes taking place in the OPA.

Irrespective of the two interpretations for the processes around time \( t_2 \) and \( t_3 \), slip along the entire interface appears to have taken place approximately after the fluid has wet the entire surface (around time \( t_4 \)), as indicated by the axial relaxation in both BER and OPA (Figures 7a, 7b, and 7d). This is supported by the axial LVDT that showed the most pronounced increase at around the same time (Figures 7a and 7b). While the data along the axial cables are qualitatively similar to that during slip initiation at higher fluid pressures (compare Figures 6 and 7), the saw-cut-parallel cable of the OPA indicates the opposite behavior, that is, compression in axial direction close to the interface. We hypothesize that this is related to the transition of the OPA into an incohesive mud (see above), which could affect the DSS data already at an earlier stage, but appears to become apparent at least around time \( t_4 \).

The alteration of the OPA saw-cut interface changes its frictional strength, which is illustrated by the stress state changing from stable to unstable during the initial contact with H2O in Exp. 3.3 (see Section 4.3 and Figures 10b and 10e). However, in Exp. 3.1 and 3.2, the stress state at initial H2O contact was below the wet frictional slip envelope, suggesting that the alteration of the frictional strength cannot be the only explanation for initial slip and that the effective stress at the saw-cut interface cannot be fully described by the measured mechanical data (Figures 10b and 10f). Possibly, the build-up of swelling stress has to be considered in order to represent the stress conditions at the OPA-BER interface (Bigaroni et al., 2023; Paterson & Wong, 2005; C. L. Zhang, 2017, 2018).

To estimate the required swelling stress for slip initiation, we assumed that the foliation perpendicular swelling stress \( \sigma_{SWL}^e \) acted against the lowest principal stress \( \sigma_1 \) and the foliation parallel swelling stress \( \sigma_{SWL}^f \) against the highest principal stress \( \sigma_3 \). Assuming a swelling stress anisotropy \( \sigma_{SWL}^f \div \sigma_{SWL}^e \) that ranges between 0.02 (Thury & Bossart, 1999) and 0.42 (Bossart & Thury, 2008), the required foliation perpendicular swelling stress to reach failure \( \sigma_{SWL}^f \) ranges from 1.9 to 2.3 MPa for Exp. 3.1 (Figure 12a), and from 1.6 to 2.0 MPa for Exp. 3.2 (Figure 12b). This is in the range of foliation perpendicular swelling stresses determined for OPA (Bossart & Thury, 2008; Thury & Bossart, 1999; C. L. Zhang et al., 2010; Ziegler et al., 2019). Based on a laterally unconstrained uniaxial swelling test, C. L. Zhang et al. (2010) even measured a swelling stress of \( \sim 3.5 \) MPa, but desaturation previous to testing may have caused higher values than can be expected under naturally saturated conditions (Schmitt et al., 1994).
4.5. Implications for Natural Processes and Anthropogenic Subsurface Applications

Our data imply that H₂O-clay interactions at initial fluid contact initiate slip because of (a) changes in mechanical properties (Moore & Lockner, 2004; Orellana et al., 2019; Petit & Laville, 1987) and (b) the build-up of swelling stress. The latter shows that the commonly applied effective stress law (Equation 4) does not hold in certain cases (e.g., Handin et al., 1963; Skempton, 1961), and that the swelling stress may have to be integrated to describe the effective stress state (C. L. Zhang, 2017, 2018). This may become relevant in critically stressed fault zones in clay-rich sedimentary rocks. For example, in subduction zones and accretionary wedges, where fluid flow is thought to be transient and the pore water chemistry changes with depth (Labuena et al., 1997; Saffer & Tobin, 2011; Solomon et al., 2009; Torres et al., 2004), which may affect the build-up of swelling stress (e.g., Abdullah et al., 1999).

From an environmental perspective, the permeability of faults in clay-rich formations, especially in rocks such as OPA, is of paramount importance for the disposal of nuclear waste and CO₂ repositories. Wenning, Madonna, Kurotori, et al. (2021) and Cappa et al. (2022) showed, based on laboratory experiments, that the permeability of OPA increases during fault slip, which agrees with slickenfibrres on fault planes described by Akker et al. (2023). Swelling strain reduces the permeability by closing of cracks (e.g., Bock et al., 2010; Fang et al., 2017; Voltolini & Ajo-Franklin, 2020; Wenning, Madonna, Kurotori, et al., 2021; C. L. Zhang, 2011), but on the other hand, our data show that swelling stress may contribute to the reactivation of critically stressed faults. Therefore, estimating the overall sealing capacity of OPA should also include potential fault reactivation.

4.6. Limitations

The following limitations in our experimental approach should be noted:

1. Artifacts in swelling stress investigations may occur due to humidity loss during sample preparation, requiring saturation of the sample prior to testing (Schmitt et al., 1994). This was taken into account by equilibrating the OPA samples with 90% relative humidity (Figure S1 in Supporting Information S1), but the degree of saturation may have decreased due to interaction with air in the pore-pressure pipes or during air evacuation prior to fluid injection. However, C. L. Zhang et al. (2010) showed that the water content of OPA at a relative humidity of 100% increased from the natural level of 7% to >14%. Therefore, we assume that the swelling stresses in our experiments are not higher than if we could have used perfectly preserved natural samples.

2. The use of deionized water results in osmotic swelling stresses at the upper end of the spectrum (Delville & Laszlo, 1990; Madsen & Müller-Vonmoos, 1989), which is consistent with the increase in pore pressure measured in clay-rich rocks when in contact with deionized water (Bossart & Thury, 2008; Hale et al., 1993; Noy et al., 2004; Q. Zhang et al., 2015). On the other hand, swelling strain and swelling stress determined for OPA revealed a minor effect on pore water chemistry (Thury & Bossart, 1999; Wenning, Madonna, Kurotori, et al., 2021).

3. Fluid injection friction tests under undrained conditions, or when using low-permeability claystones, may result in higher fluid pressures within the sample than measured externally, leading to an overestimation of the effective stress (e.g., Crisci et al., 2022; Faulknner et al., 2018; Saffer & Marone, 2003). While we cannot rule out this effect occurring within the OPA, we do not expect fluid overpressures at the OPA-BER interface because the fluid pressure is controlled via the highly permeable sandstone.

4. Similarly to what was observed at the initial H₂O injection, swelling stresses may occur later in the experiment, which would affect our estimation of the wet frictional slip envelopes. However, we assume that the swelling stress was largely dissipated due to the elastic relaxation of the rock matrix during initial slip. This is indicated by (a) the discrepancy between the stress state at initial fluid contact and the wet frictional slip envelope for Exp. 3.1 and 3.2 (Figure 12) and (b) the similar response during the increasing fluid pressure steps regardless of whether decane or H₂O was injected (phase III-a and III-b in Figures 3 and 4).

5. Conclusions

We performed triaxial saw-cut friction tests with OPA as the upper half of the sample and BER as the lower half. Experiments without fluid injection were conducted at confining pressures of 4–25 MPa with constant axial displacement rate of 0.1 mm/min. We obtained a non-wetted frictional slip envelope showing that the OPA dominates the frictional strength, and that the experimental set-up is representative for clay-rich rocks.
Fluid injection experiments were carried out under confining pressures of 10 and 25 MPa, with the piston held in a constant position, and an initial differential stress set at approximately 70% of the anticipated yield stress. The experimental procedure involved a step-wise increase in fluid pressure, utilizing deionized (a non-polar fluid) in one test series and deionized water (a polar fluid) in another. The initiation of slip at progressively higher fluid pressures and lower differential stresses allowed us to estimate frictional slip envelopes of single experiments. These revealed a trend toward lower frictional strengths when a polar fluid was injected compared to a non-polar fluid. As suggested in previous studies, this indicates that the polar water is adsorbed between negatively charged phyllosilicate sheets, which has a lubricating effect. In addition to phyllosilicate lubrication, the frictional weakening may have been supported by the alteration of the OPA surface, which exhibits similarities to hydrophobic plastic faults.

Another difference when injecting a polar fluid compared to a non-polar fluid is a differential stress drop that occurs at initial hydration of the OPA surface (at ambient fluid pressure of ~0.1 MPa). DSS at this stage of the experiment indicated (a) a progressive alteration of the OPA-BER interface governed by the upward migration of the fluid front, and (b) slip initiation approximately when the entire interface was hydrated. In two out of three water injection experiments, slip was initiated at initial hydration although the apparent effective stress state was below the frictional slip envelope. This shows that the simplified effective stress law \( \sigma_N^e = \sigma_N - P_f \) may not hold in similar cases. However, if the swelling stress is taken into account, which may occur due to clay-water interaction but not clay-decane interaction, the stress state is in accordance with the frictional yield stress.

In summary, our data suggest that water-clay interactions may contribute to reactivation of critically stressed faults by (a) alteration of the frictional strength and (b) the build-up of swelling stress. This has implications for understanding fault zones in clay-rich rocks, such as in the shallow subduction zone, where fluid dynamics and fluid chemistry changes may lead to effects similar to those observed in our experiments. In addition, fault reactivation due to water-clay interactions should be considered when estimating the sealing capacity of OPA with respect to nuclear waste deposits and CO2 storage facilities.

Data Availability Statement

The data set presented in this manuscript is available on the ETH research collection at https://doi.org/10.3929/ethz-b-000636543 (Rast et al., 2023).

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