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RESEARCH ARTICLE

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Key Points:

- A model clay system shows velocity-strengthening behavior following the Herschel-Bulkley model
- Despite the velocity-strengthening friction, shear band is formed at low strain rates
- The stacking structure of clay platelets is reduced during shear and undergoes logarithmic healing with time in the system at rest

Supporting Information:

Supporting Information may be found in the online version of this article.

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Rheology and Structure of Model Smectite Clay: Insights From Molecular Dynamics

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Abstract The low frictional strength of smectite minerals, such as montmorillonite, is thought to play a critical role in controlling the rheology and the stability of clay-rich faults. In this study, we perform molecular dynamics simulations on a model clay system. Clay platelets are simplified as oblate ellipsoids interacting via the Gay-Berne potential. We study the rheology and structural development during shear in this model system, which is sheared at constant strain rates for 10 strains after compression and equilibrium. We find that the system exhibits velocity-strengthening behavior over a range of normal stresses from 1.68 to 56.18 MPa and a range of strain rates from 6.93×10^5 to 6.93×10^8 /s. The relationship between shear stress and strain rate follows the Herschel-Bulkley model. Shear localization is observed at lower strain rates despite the velocity-strengthening friction, while homogeneous shear is realized at higher strain rates. The structure change due to shear is analyzed from various aspects: the porosity, particle orientation, velocity profile, and the parallel radial distribution function. We find that particle rearrangement and compaction dominate at the early stage of shear when the shear stress increases. The shear band starts to form in the later stage as the shear stress decreases and relaxes to a steady-state value. The structural development at low strain rates is similar to previous experimental observations. The stacking structure is reduced during shear and restores logarithmically with time in the rest period.

Plain Language Summary Smectite clay minerals have relatively low frictional strength compared to other geomaterials. The strength and stability of clay-rich faults depend on the shear behavior of clay minerals. In this study, we perform a series of simulations to investigate the shear behavior and structure of a simple clay model. The complex shape and interactions of natural clay are simplified as disc-shaped ellipsoid and a generalized potential. The system is sheared at constant strain rates for 10 strains after compression and relaxation. The simulations focus on the effect of normal stress and strain rate on shear stress and clay structure. We find that the shear stress increases with strain rate for all conditions tested. Two shear scenarios are observed with shear localization in a narrow region at low strain rate while shear is homogeneous at high strain rate. The structural development at low strain rates is similar to previous experimental observations. We observe that the stacking structure of clay platelets weakens during shear. In the rest period after shear, the stacking structure restores logarithmically with resting time and can be associated with the healing of clay-rich fault.

1. Introduction

Smectite minerals, such as montmorillonite, are abundant in the top part of the upper crust and are commonly found in shallow subduction zones (Carpenter et al., 2011; Collettini et al., 2019; Deng & Underwood, 2001; Ikari et al., 2011; Jeppson & Tobin, 2015; Lockner et al., 2011; Wojatschke et al., 2016; Wu et al., 1975). During the March 2011 Tohoku-Oki earthquake, the large slip in the shallow part of subduction zone caused a devastating tsunami (Chester et al., 2013; Ujiie et al., 2013). Drilling investigations revealed a clay-rich fault zone with a composition of 60%–80% smectite (Kameda et al., 2015). The shear resistance of smectite is generally lower than that of other geomaterials, and therefore it has been associated with the weakness of some specific faults as well as shallow subduction plate boundaries.

Montmorillonite, a typical smectite at low temperatures, consists of platelets with high aspect ratios and a thickness of ~ 1 nm. Natural montmorillonite platelets are highly polydisperse in diameter with irregular shapes (Michot et al., 2004; Mouzon et al., 2016). This mineral is well known for its high activity in water. In aqueous environments, montmorillonite absorbs and exchanges cations, and the surface of platelets is negatively charged. The charge of platelet edges depends on the ionic condition: they are negatively charged under alkaline condition but positively charged under acidic condition (Paineau et al., 2013). Water molecules are also attracted to the

surface, forming interlaminar water layers, which contribute to swelling (Bird, 1984; Ho et al., 2019; Shen & Bourg, 2021).

The rheology and stability of the fault zone are governed by the mechanical properties of the fault gouges. Thus, the frictional strength and fabric development in clay and simulated fault gouges with different clay contents have been extensively studied in friction experiments (Behnsen & Faulkner, 2013; Bird, 1984; K. M. Brown et al., 2003; Haines et al., 2013; Ikari et al., 2007; Moore & Lockner, 2007; C. B. J. B. Morrow et al., 1992; Saffer & Marone, 2003; Summers & Byerlee, 1977; Tembe et al., 2010; Tesei et al., 2012). The saturation state of sample and the corresponding experimental procedures are crucial for measuring of frictional properties of montmorillonite, as summarized in C. A. Morrow et al. (2017). The coefficient of friction increases with the effective normal stress when montmorillonite is fully saturated with controlled pore pressure. For other cases including dry, unsaturated, or overpressured cases, the coefficient of friction decreases with normal stress.

Both field and laboratory studies have revealed interesting fabric evolution that may affect the frictional strength of clay-rich faults (Haines et al., 2013; Kenigsberg et al., 2019, 2020; Logan & Rauenzahn, 1987; Niemeijer et al., 2010; Saffer & Marone, 2003). For example, scaly clay foliation was observed in a core sample from the shallow source of the 2011 Tohoku-Oki earthquake (Chester et al., 2013). Laboratory experiments on crushed montmorillonite produced Riedel shears and boundary-parallel shears, which developed systematically with the shear strain (Kenigsberg et al., 2019). Scanning electron microscope (SEM) observations suggested that clay particles were reoriented on the plane perpendicular to the maximum principal stress (Haines et al., 2013). However, the origin of the above features is not fully understood.

Clay minerals can carry charge and absorb water, and they can affect the frictional properties in unconventional manner. Namely, understanding based on surface contacts alone may not be sufficient for clay minerals. To account for these effects, more effort is needed to link microscopic physical processes with macroscopic mechanical properties. In this respect, the nature of interaction force between clay particles is of particular importance. The classical description of clay interactions is based on the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory for charged parallel planes, which includes van der Waals attraction and electrostatic diffuse double layer repulsion (Derjaguin, 1941; Verwey, 1947). This is later improved by considering the ion density fluctuations between two plates which were neglected in the DLVO model (Pellenq et al., 1997). The new ion correlation force contributes to the attraction between clay particles of the same charge and leads to the stacking of platelets.

The understanding of clay interaction and clay swelling has greatly advanced due to the development of molecular dynamics (MD). The clay platelets can be built with the explicit atomistic structure together with the ions and solvent molecules. However, fully atomistic modeling is often limited to small systems due to high computational cost (Underwood & Bourg, 2020). Typical simulations of clay swelling and sliding behavior focus on only two clay platelets in bulk water (Ho et al., 2019; Wei et al., 2021).

Attempts have been made to simplify the complex interactions between the clay platelets using the disk-like shape. For early exploration, an infinitesimally thin disk with a quadrupole potential was used as the clay platelet with the electric double layers (Dijkstra et al., 1995). The next attempt involved constructing a platelet from multiple spheres together with some charged sites interacting via electrostatic potential (Delhorme et al., 2014; Kutter et al., 2000; Odriozola et al., 2004; Ruzicka et al., 2011; Thuresson et al., 2017). The resulting interaction can be viewed as a generalization of the DLVO potential for platelets. While the early models focused on dilute dispersions of synthetic clay, similar ideas are recently applied to denser systems (Aminpour & Sjöblom, 2019; de Bono & McDowell, 2022). In particular, the Gay-Berne (GB) potential has been used to study clay aggregation with the shape simplified to oblate ellipsoids (Bandera et al., 2021; Ebrahimi et al., 2014). The GB potential incorporates both short-range repulsion and mid-range attraction. It is commonly used in previous liquid crystal studies and has the ability to account for both the shape and interaction anisotropy of clay platelets.

Simulations with simplified shapes and interactions have proven to be useful tools in recent studies on compressed clay systems (Asadi et al., 2022; Bandera et al., 2021; Bayesteh & Hoseini, 2021; de Bono & McDowell, 2022; Ebrahimi et al., 2016). However, few studies have applied this method to investigate the rheology or friction of clay under large deformation. Yamamoto et al. (2005) studied the suspension rheology of disk-like particles under simple shear using the GB potential. This study predicted shear thinning behavior in viscosity

and particle alignment in the flow direction at high strain rates. To study clay in a fault zone condition, systems with low porosity and higher normal stress must be examined.

In this paper, we report the MD simulation of a simple model clay from a mesoscopic level. We aim to provide insights into the rheology and structure of clay and clay-rich fault gouges using simple non-spherical particles with the GB potential. We focus on the influence of strain rate and normal stress on shear behavior at room temperature. Another aim is to provide some direct evidence of the connection between shear and microstructure. To this end, we examine the structure development during shear through porosity, particle orientation, velocity profile, and the parallel radial distribution function (PaRDF).

2. Computational Model and Methods

The MD enables us to simulate the time evolution of particles under different physical conditions. The simulations are carried out using the LAMMPS simulation package (W. M. Brown et al., 2009; Thompson et al., 2022). The molecules are visualized using the OVITO software (Stukowski, 2009).

The temperature and the pressure can be controlled to the desired values using specific algorithms referred to as “thermostat” (temperature control) and “barostat” (pressure control), respectively. In this study, the temperature is always controlled using the thermostat, while the barostat is optional. System with active barostat have constant pressure and changeable volume (constant pressure condition). If the barostat is not active, the system volume is constant, and the pressure can change (constant volume condition).

2.1. Gay-Berne Potential

Particle interactions in MD are often defined with the potential energy as a function of separating distance and the interacting force are calculated from the negative slope of the potential. In the present study, smectite clay platelets are simplified as rigid oblate ellipsoids interacting via the GB potential defined by (Coussaert & Baus, 2002; Gay & Berne, 1981; Luckhurst & Simmonds, 1993):

$$U = 4\epsilon_0 \times \epsilon_1(\kappa) \times \epsilon_2(\kappa') \left\{ \left(\frac{\sigma_{ff}}{h_{12} + \sigma_{ff}} \right)^{12} - \left(\frac{\sigma_{ff}}{h_{12} + \sigma_{ff}} \right)^6 \right\} \quad (1)$$

where ϵ_0 determines the energy scale, ϵ_1 characterizes the shape anisotropy defined by the aspect ratio κ , ϵ_2 characterizes the orientation anisotropy in terms of the energy ratio κ' . The aspect ratio κ is the ratio between thickness σ_{ff} and diameter σ_{ee} of a disc-like ellipsoid. Here σ_{ee} sets the length scale of σ_0 . The energy ratio κ' is the ratio of potential well depth for two different configurations: end-to-end (ee) and face-to-face (ff). With $\kappa' < 1$, the potential favors ff over ee configuration and promotes stacking. h_{12} is the approximated distance between two interacting particles. The positive term $\left(\frac{\sigma_{ff}}{h_{12} + \sigma_{ff}} \right)^{12}$ represents the short-range repulsion and the negative term $-\left(\frac{\sigma_{ff}}{h_{12} + \sigma_{ff}} \right)^6$ represents the mid-range attraction between two particles. More details on the GB potential are given in Text S1 and Figure S1 in Supporting Information S1.

Previous studies show that the GB potential is a good approximation for the interaction between two clay platelets derived from the full atomistic MD for end-to-end and face-to-face configurations (Ebrahimi et al., 2014). For simplicity, here we do not pursue the perfect match with the full atomistic MD results but use the parameter values assumed in some recent MD studies. Based on the interaction between two montmorillonite platelets with diameter of ~ 6 nm in face-to-face configuration (Shen & Bourg, 2021), the potential well for aspect ratio $\kappa = 1/5$ is -0.5 eV, which corresponds to the free energy minimum at the salinity of $0.3\sim 0.5$ M NaCl. The spacing between two hydrated smectite at free energy minimum is approximately 1.55 nm with two interlaminal water layers. The soft contact distance is also defined from $\sigma_{ff} = 1.5$ nm.

The system consists of ellipsoids of three different aspect ratios, $1/5$, $1/6$, $1/7$. They have the same population, and referred to as particle type A, B, and C. They have the same thickness (1.5 nm) but different diameters (7.5, 9.0, 10.5 nm) determined by the aspect ratio. The energy ratio κ' is set to $1/5$ for all the particles. The parameter used in the GB potential is summarized in Table S1 in Supporting Information S1.

2.2. Configuration and Setup of Simulation

The simulation is performed using 4,096 particles inside a triclinic box with periodic boundary conditions. Translational and rotational equations of motion are integrated using the velocity Verlet algorithm with a dimensionless time step $\delta t^* = 0.00025$ if not specified. To optimize the computational time, the interaction potentials are truncated at a cutoff of $1.6\sigma_0$.

Initially, each platelet is placed on a simple cubic lattice with the number density of 0.2. The initial orientation and the initial velocity are assigned randomly. For the following time evolution, Langevin thermostat is used to control the reduced temperature at $T^* = 1.75$ (equivalent to 300.41 K). The total force acting on each particle is then (Schneider & Stoll, 1978):

$$F = F_c + F_f + F_r \quad (2)$$

where F_c is the force calculated from the interaction potential, $F_f = \Gamma mv$ is a frictional drag force, and F_r is a random force resulting from collisions with solvent particles. Frictional drag is given as $F_f = \Gamma mv$, where Γ is the damping constant, m is the particle mass, and v is the particle velocity. The amplitude of the random force is $\sqrt{2m\Gamma k_B T}$, where k_B is the Boltzmann constant and T is the target temperature.

In the earliest stage of time evolution (duration of $t^* = 1,000$ in the dimensionless unit), the system evolves at constant volume to randomize the platelet configuration. Subsequently, the system is compressed and equilibrated under the constant pressure condition using the Nosé-Hoover style barostat. This method assumes the flexible simulation cell. In other words, each dimension of the simulation cell is adjusted to control each normal stress in the X, Y, and Z directions, respectively (Shinoda et al., 2004). The target values are $P^* = 100, 300, 1,000, 3,000, 6,000$, and $10,000$ in the dimensionless unit, which correspond to 0.56, 1.69, 5.62, 16.85, 33.71, and 56.18 MPa. We assume a fully saturated and fully drained system and therefore the normal stress in the present simulation should be interpreted as the effective normal stress that is, $P = \sigma_n = \sigma_{\text{eff}}$. For each stage of pressure elevation, the pressure first increases linearly in $t^* = 200$ and is kept constant in the subsequent equilibration process of $t^* = 2,000$ similar to Ebrahimi et al. (2014).

After this compression and relaxation process, the system is sheared at a constant strain rate. The shear is applied by deformation of the simulation cell instead of driven by the movement of plate. Flow velocity is in the X direction with the velocity gradient in the Z direction. The Lees-Edwards boundary condition is used in the Z direction in order to adapt to the shear strain (Evans & Morriss, 2007). The particle velocity in the X direction is updated when one goes across the $\pm Z$ boundaries as:

$$v_{x_new} = v_x \pm \dot{\gamma} L_z \quad (3)$$

Here L_z is a system dimension in the Z direction and $\pm$ are for particles leaving from the bottom/top boundary, respectively. In addition, the particle interaction across the Z boundary is computed with the additional distance of $\pm \dot{\gamma} L_z$ in the X direction. Standard periodic boundary conditions are applied in the X and the Y directions.

The frictional drag force in the Langevin thermostat is applied with the bias of linear velocity profile during shear in the flow direction x as:

$$F_{f_x} = \Gamma m(v_x - \dot{\gamma} z) \quad (4)$$

where v_x is the x component of a particle's velocity, $\dot{\gamma}$ is the strain rate, and z is the z coordination of a particle.

During shear, the Nosé-Hoover barostat is only applied in the Z direction to control the normal stress. The dimensionless strain rates are given as $\dot{\gamma}^* = 1, 0.4, 0.1, 0.04, 0.01$, and 0.001 ($6.93 \times 10^8, 2.77 \times 10^8, 6.93 \times 10^7, 2.77 \times 10^7, 6.93 \times 10^6$, and $6.93 \times 10^5/\text{s}$) at the dimensionless normal stresses of $P^* = 300, 1,000, 3,000, 6,000$, and $10,000$ (1.69, 5.62, 16.85, 33.71, and 56.18 MPa). A reduced time step $\delta t^* = 0.000125$ is used in the case with $P = 56.18$ MPa and $\dot{\gamma} = 6.93 \times 10^5/\text{s}$. We pay more attention to the system at $P = 5.62$ MPa, which is comparable to the effective normal stress of 7.0 MPa at the shallow subduction zone that accommodated the large slip in the 2011 Tohoku-Oki earthquake (Chester et al., 2013).

To examine the structural change after the flow cessation, the sheared system is held again and relaxed without shear for the duration of $t^* = 1,000$. In this hold period, we consider two different conditions: constant volume and constant pressure. At the constant pressure condition, the normal stress in the Z direction is controlled at the same value throughout the shear and the hold. The result is reported in the real unit for comparison with the previous studies.

2.3. Structural Analysis

Structural analysis tools on the bulk system can be used with the application of periodic boundary. The extend of particle alignment in the system can be determined from the orientational tensor:

$$Q = \frac{1}{2N} \sum_{i=1}^N (3u_i \otimes u_i - I) \quad (5)$$

where u_i is the unit vector along the symmetry axis of particle i (i.e., the normal vector of platelet i), $\otimes$ denotes the tensor product, I denotes the identity matrix, and N is the total number of ellipsoids in the system. The nematic order parameter S is then defined as the largest eigenvalue of the orientational tensor Q . When the nematic order parameter vanishes ($S = 0$), the orientation of each platelet is randomly distributed, whereas $S = 1$ means that all the platelets are perfectly aligned. The corresponding eigenvector is referred to as the *director* of the system, which is regarded as the averaged symmetry vector of the platelets.

The PaRDF is used to characterize the spatial structure of the platelet mixture (Busselez et al., 2014; Caprion et al., 2003; Cienega-Cacerez et al., 2014). The PaRDF is the radial distribution function along the symmetry axis of particle i (i.e., the normal vector of platelet i) $\hat{u}_i$ and expresses the extent of stacking structure (tactoid formation) of platelets. It is defined as:

$$g^{\parallel}(r) = \frac{V}{N^2 2\pi(\sigma_{ee})^2 h} \left\langle \sum_i \sum_{j \neq i} \delta(r - r_{ij}^{\parallel}) \theta(\sigma_{ee}/2 - |r_{ij}^{\perp}|) \right\rangle \quad (6)$$

where V is the volume of the simulation box, N is the number of ellipsoid particles, h is the cylinder height used to discretize the volume. $r_{ij}^{\parallel} = |\mathbf{r}_{ij}^{\parallel}| = |\mathbf{r}_{ij} \cdot \hat{u}_i|$ is the center-to-center distance between particle i and j along the symmetry axis of particle i , and $r_{ij}^{\perp} = |\mathbf{r}_{ij}^{\perp}| = |\mathbf{r}_{ij} - \mathbf{r}_{ij}^{\parallel}|$ is the corresponding perpendicular component. $\theta(x)$ is the Heaviside step function. We use the diameter of the smallest particle type A (aspect ratio 1/5) for σ_{ee} in the mixture system. The sum regarding j is taken only for the particles within the perpendicular distance of the particle radius ($\sigma_{ee}/2$). The PaRDF gives the normalized probability density of finding another particle along the symmetry axis direction of the center particle at the distance of r . The search is restricted inside a column region where σ_{ee} is the diameter of the searching column. A schematic figure of PaRDF is shown in Figure S2 in Supporting Information S1.

3. Results

3.1. Porosity and Nematic Order Under Compression

Examples of the system configuration are shown in Figure 1 at the normal stress of 5.62 MPa. A similar configuration at a higher normal stress of 33.71 MPa is shown in Figure S3 in Supporting Information S1. The system is first compressed to reach equilibrium at the given normal stress. The target values are 0.56, 1.69, 5.62, 16.85, 33.71, and 56.18 MPa, respectively. The compaction process is monitored via the porosity $\phi = (1 - V_{\text{particle}}/V_{\text{system}})$, which decreases from 0.48 at $P = 0.56$ MPa to 0.33 at $P = 56.18$ MPa (Figure 2a). The range of porosity in our simulation is similar to some experimental data: for example, oven-dried Na-smectite (Likos & Lu, 2006) and partially saturated bentonite at higher normal stress (Baille et al., 2010), while Na-Ca-smectite (MX80) saturated with pure water at low normal stress showed higher porosity (Marcial et al., 2002).

Figure 2b shows that the nematic order increases from 0.37 to 0.47 during compression, indicating increasing order in orientation at higher normal stress. A similar trend is observed in both experimental and simulation

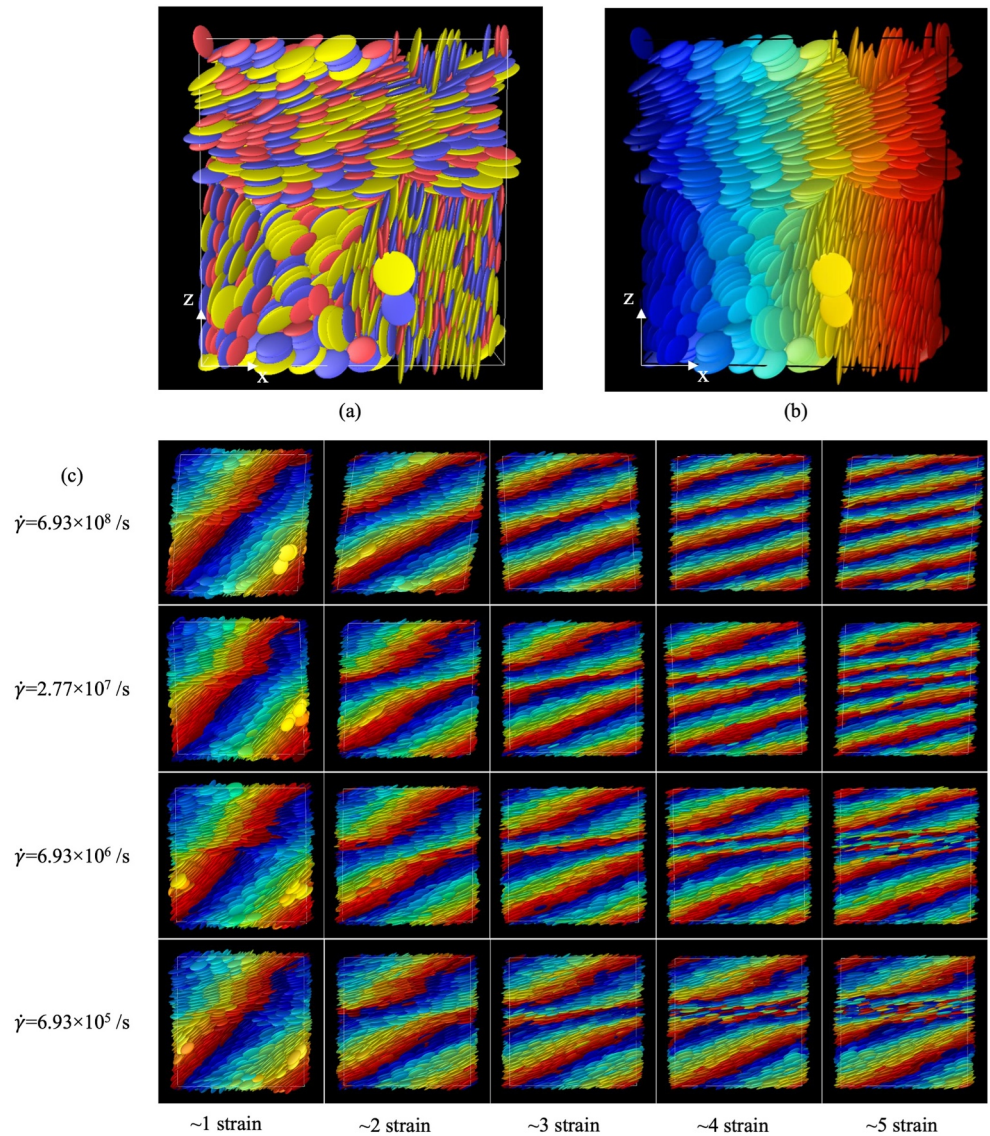


Figure 1. Examples of compression and shear configurations at normal stress $P = 5.62$ MPa, a view on the XZ plane. (a) Compressed system colored by different particle aspect ratio, where red, blue, and yellow correspond to particle aspect ratio of 1/5, 1/6, and 1/7. (b) Mixture system colored by the x coordinate of particles at strain of 0, the same coloring is used for (c). (c) Two scenarios are observed with homogeneous shear at high strain rate ($\dot{\gamma} \geq 2.77 \times 10^7$ /s) and the shear band at low strain rate ($\dot{\gamma} \leq 6.93 \times 10^6$ /s). Shear localization appear after the strain of 2 and is more obvious at lower strain rate.

studies (Ebrahimi et al., 2016; Perdigon-Aller et al., 2005). Recent mesoscale simulations on clay matrix also suggest less orientational anisotropy for the larger system size (Asadi et al., 2022; Bandera et al., 2021).

3.2. Shear

The stress-strain curves are shown in Figure 3a. Each curve shows a peak and the subsequent weakening with increasing shear strain until a residual strength is reached. The stress peaks around the 200% shear strain for strain rates below 2.77×10^8 /s. At higher strain rates, both the peak stress and the steady-state value increase. This trend is common to experiments on crushed montmorillonite with a prominent peak in shear stress (Haines et al., 2013; Kenigsberg et al., 2019).

The evolution of porosity is shown in Figure 3b. At lower strain rates, the porosity decreases with strain before reaching the steady-state value. At higher strain rates, the system first dilates and then compresses, with the peak

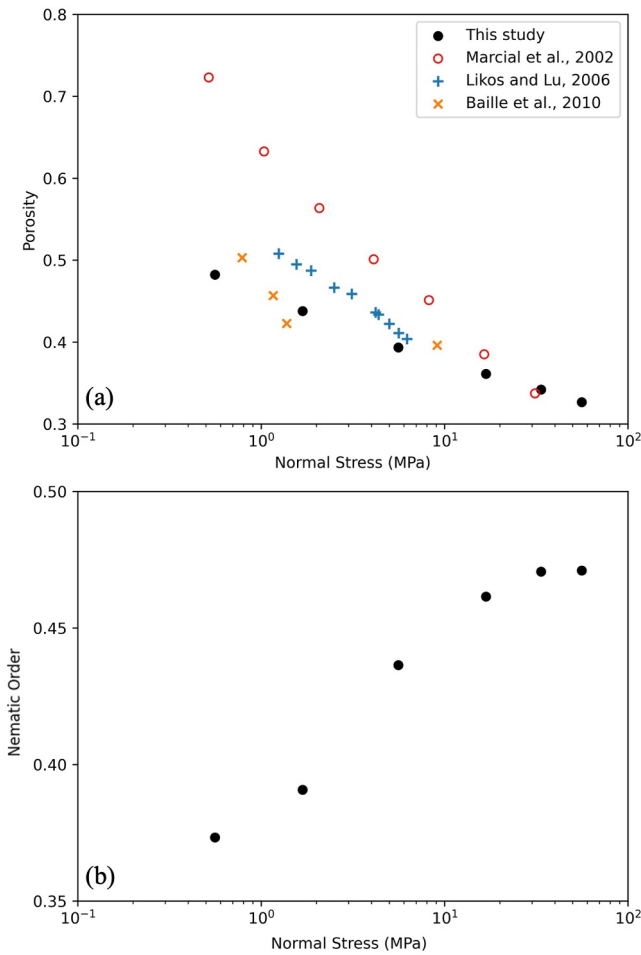


Figure 2. Porosity and nematic order as a function of normal stress during compression. Solid circle: present study, compressed to the normal stress of 0.56, 1.69, 5.62, 16.85, 33.71, and 56.18 MPa. Hollow circle: saturated Na-Ca-smectite (Marcial et al., 2002). Plus: oven-dried Na-smectite (Likos & Lu, 2006). Cross: partially saturated bentonite (Baille et al., 2010).

stress (Figure 4c). Note that the correction of strain rate with shear band does not significantly affect the stress-strain rate relationship as shown in Figure S4 in Supporting Information S1.

The steady-state coefficient of friction is defined by:

$$\mu = \tau / \sigma_n \quad (8)$$

The coefficient of friction is apparently small in the range of 0.025–0.187 and decreases with increasing normal stress and increases with strain rate (Figure 5a). The porosity decreases with normal stress and increases with strain rate (Figure 5b). This is consistent with experiments where dilation is observed in response to the increasing shear velocity (Ikari et al., 2009).

Visual inspection shows that shear banding is more apparent at lower strain rates, while homogeneous shear is more common at higher strain rates (Figure 1). The velocity profile at $P = 5.62$ MPa is shown in Figure 6 with samples divided into 20 layers along the Z direction. In each velocity profile, the flow velocity is averaged over each strain increment of 0.5. At higher strain rates ($\dot{\gamma} \geq 2.77 \times 10^7/\text{s}$), the velocity profile is relatively linear. At lower strain rates ($\dot{\gamma} \leq 6.93 \times 10^6/\text{s}$), shear localization is observed.

of dilation occurring during the first 200% strain. The porosity is larger at higher strain rates, but it may take even larger shear strains (larger than 10) to reach the steady state. We find that the peaks in shear stress and porosity at early strains coincide at higher strain rates.

Figure 3c shows that the nematic order parameter S increases from 0.4 to near 1.0 for all strain rates. These results suggest that the system becomes more orientationally ordered during shear, consistent with particle alignment analysis from shear experiments (Haines et al., 2009). A closer look at a region above $S = 0.9$ (Figure 3d) shows that the nematic order increases rapidly to above 0.9 within the 200% shear strain and then continues to increase slowly until reaching the steady-state value. Systems at higher strain rates reach the similar nematic order above $S = 0.98$, while relatively low nematic order is realized at the two lower strain rates (6.93×10^6 and $6.93 \times 10^5/\text{s}$).

The shear stress averaged over the strain of 8–10 in each run is used as the steady-state shear stress for further comparison. The result for different strain rates and different normal stresses is summarized in Figure 4a. The steady-state shear stress can be characterized by the following equation:

$$\tau = \tau_0 + K\dot{\gamma}^n \quad (7)$$

where the yield stress τ_0 denotes the shear stress at the zero-strain rate limit, K is a constant and n is the exponent. Equation 7 is referred to as the Herschel-Bulkley (HB) model, which is an empirical law describing non-Newtonian rheology such as the flow of mud and clay suspensions (Coussot & Proust, 1996; Manga & Bonini, 2012). The HB model is reduced to the Newtonian fluid with $\tau_0 = 0$ and $n = 1$, and K is then the dynamic viscosity of the fluid.

The fitting parameters at each normal stress is presented in the Table S2 in Supporting Information S1. The yield stress τ_0 increases with the normal stress (Figure 4b). An approximation with the linear dependence on normal stress gives the coefficient of friction, which turns out to be very small: 0.0135. The yield stress at the vanishing normal stress is about 0.25 MPa. The exponent n is below unity in all the cases, indicating the shear thinning behavior. We find the tendency that the exponent n decreases with normal

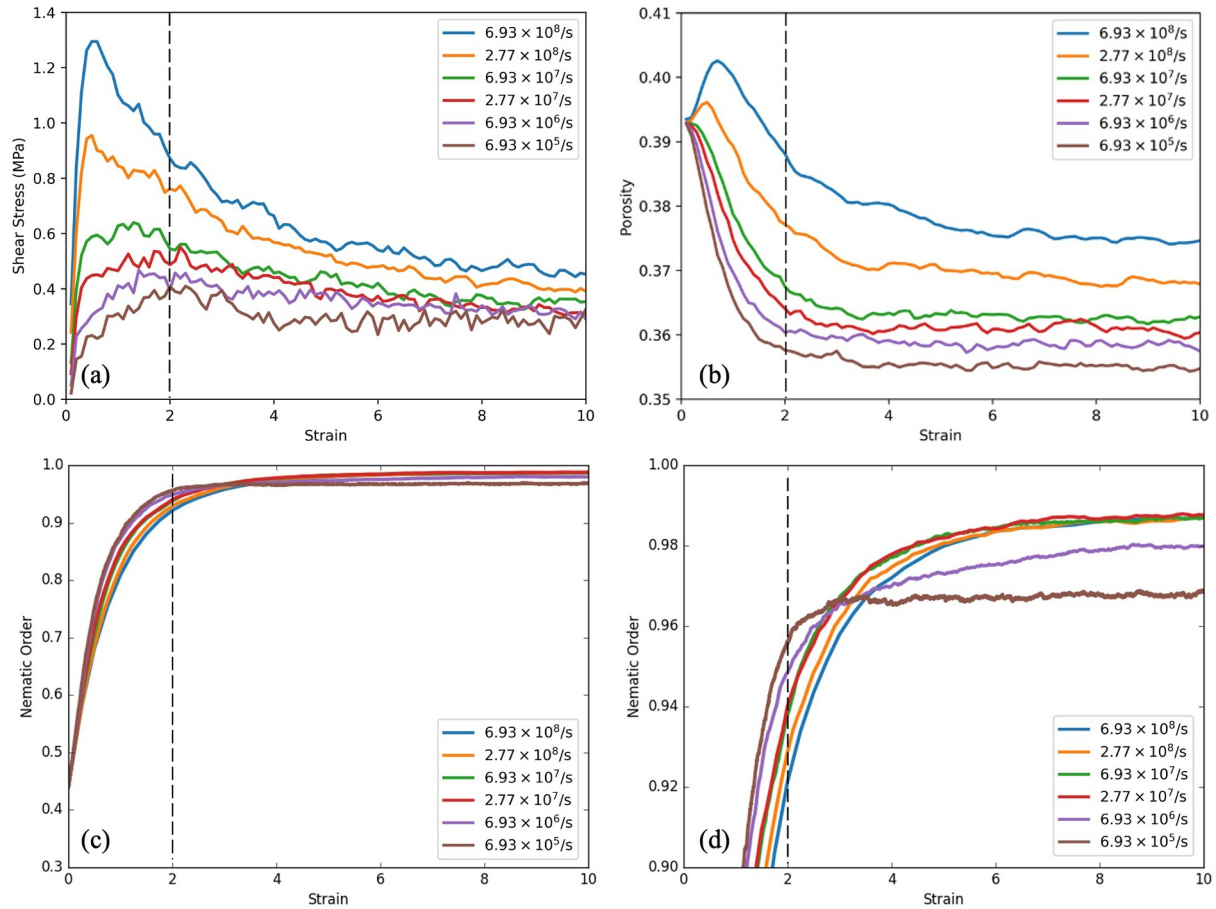


Figure 3. Example of (a) shear stress, (b) porosity, and (c, d) nematic order parameter as a function of shear strain for all strain rates at the normal stress of 5.62 MPa. Dashed line is a guide line of the position of peak shear stress around the strain of 200% for lower strain rate cases. The two highest strain rates experienced the peak stress at early strain, close to the position of dilation in porosity. (d) A zoom at larger nematic order shows the two lowest strain rates reached slightly lower order.

The average angle between the symmetry axis of the particle and the Z axis is used to represent the structural rearrangement during shear deformation (Figure 6). The samples are divided into 20 layers along the Z direction. The black line at strain 0 shows the initial orientation profile. At high strain rate ($\dot{\gamma} \geq 2.77 \times 10^7/\text{s}$), the velocity profile is linear, and the orientation profile becomes homogeneous at large strains. The dashed line indicates the steady-state value is about 10° . In the presence of shear band ($\dot{\gamma} \leq 6.93 \times 10^6/\text{s}$), the particle orientation is approximately 10° inside the shear band, while it is larger outside the band. A preferred orientation of 10° in the XZ plane and 0° in the YZ plane are observed at high strain rate (see Figure S5 in Supporting Information S1). Recent studies show that the preferred orientation is controlled by particle shape at low porosity (Börzsönyi et al., 2012; Marschall et al., 2020; Nagy et al., 2017). Shear banding also explains the low nematic order at low strain rates shown in Figure 3d, the overall orientation order is lower than in the homogeneous cases.

Figure 7a shows the change in PaRDF with increasing strain. The initial configuration at zero strain has a short-range correlation with seven pronounced peaks. The peak height decreases with the parallel distance and diminishes to unity around $r = 1.5$. Both the peak height and the peak number decreases during shear; that is, the decrease in the parallel order. At the shear strain of 10, the peaks have a much lower magnitude and only the first three peaks are recognizable. Namely, the parallel stacking of clay particle is reduced by the shear.

In each RDF, peaks larger than 1.05 can be fitted with the exponential function:

$$g_{\text{peak}} - 1 = A * e^{-r/B} \quad (9)$$

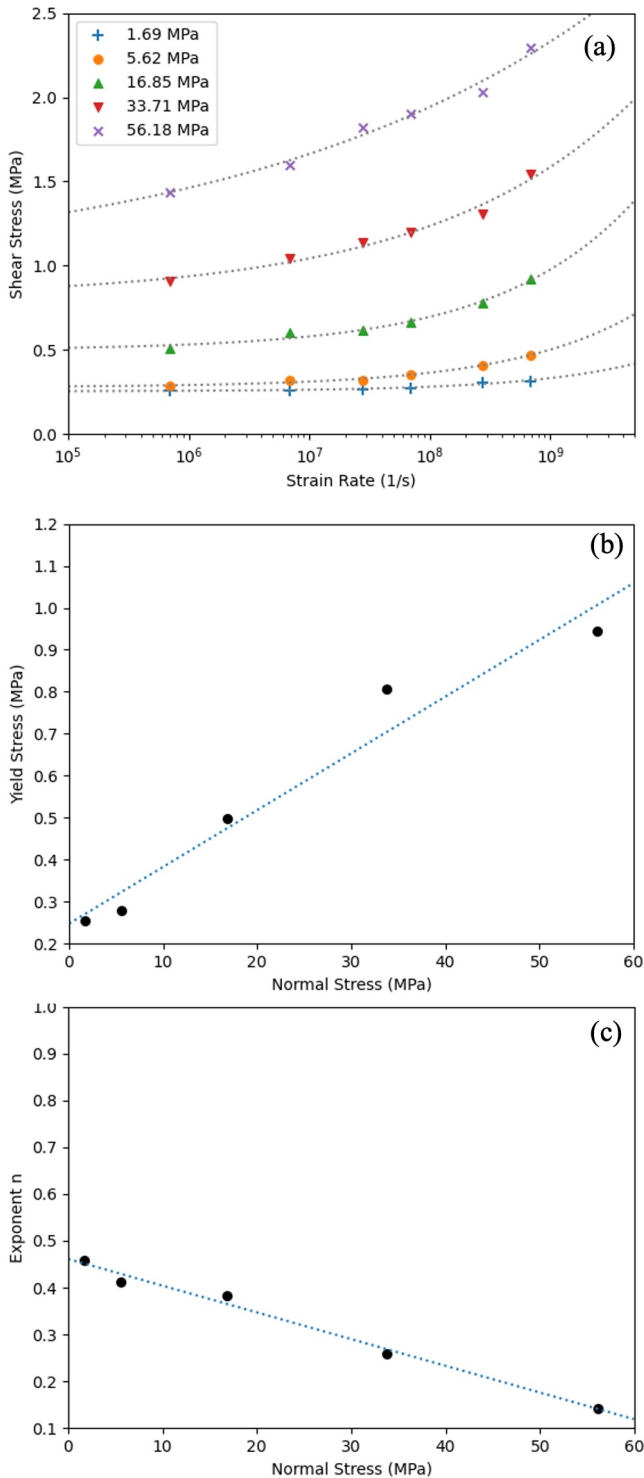


Figure 4. (a) Shear stress as a function of strain rate at different normal stresses, fitted with Herschel-Bulkley model. (b) The yield stress at zero strain rate limit increases with normal stress. The dotted line is the approximation with a linear model. (c) The exponent decrease with normal stress. The dotted line is the approximation with a linear model.

where B defines the characteristic length for parallel stacking. The fitting result for $P = 5.62$ MPa and $\dot{\gamma} = 6.93 \times 10^8$ /s is shown as an example in Figure 7b in a semi-log plot. The peaks at zero strain do not strictly follow the exponential line with higher values at greater distances. After the shear is applied, the peaks first decrease rapidly and reach the lowest curve around the shear strain of 2. Then, in the following 8 strains the peaks increase again gradually but only slightly. In other words, the parallel stacking order does not decrease monotonically with the shear strain. However, since the number of appreciable peaks decreases with the shear, the fitting here may not give a sufficient estimate. The fitting parameters are shown in Table S3 in Supporting Information S1.

The same analysis is conducted for cases with the strain rate larger than 2.77×10^7 /s at $P = 5.62$ MPa. In Figure 7c, the characteristic stacking size B is shown as a function of shear strain at different strain rates. The result is approximated by an exponential function of the strain:

$$B = C * e^{-\gamma/D} + E \quad (10)$$

where γ is the shear strain, D denotes the characteristic strain, and E denotes the residual value at $\gamma = +\infty$. All the fitting parameters are listed in Table S4 in Supporting Information S1. The fitting Function 6 generally provides a good estimation but underestimates the stacking size at the strain of 10 for all strain rates. The steady-state stacking size E decreases with strain rate, this again shows the stacking order is reduced by the shear (Figure 7c inset).

3.3. Aging at Rest

Clay can form complex microscopic structure and exhibits thixotropy. Thixotropy is a time-dependent increase of viscosity depending on the shear stress. From a microscopic view, thixotropy is caused by a time-dependent restructuring of fabric of the particles in a suspension referred to as aging. Since thixotropy acts as a positive feedback, stick-slip behavior is expected in clay-rich materials (Kameda & Hamada, 2022).

Here, we study such time-dependent restructuring in our model clay. Structural analysis using PaRDF is conducted on a system at rest after shear at the normal stress $P = 5.62$ MPa. Shear rate is set to zero after the shear strain of 10 is applied. For the system at rest, we test two different conditions: constant normal stress and constant volume. These two conditions may correspond to drained or undrained conditions at the laboratory scale, respectively.

The characteristic stacking size B increases during rest, indicating the restoration of the parallel stacking structure during rest (Figure 8). Previous experiments on samples taken from a phyllosilicate-rich fault have observed frictional healing with the hold time, but the healing rate is much smaller than that in phyllosilicate-free materials (Tesei et al., 2012). For clay suspensions and analog yield-stress materials, the logarithmic increase of peak stress with the waiting time is predicted (Bonn et al., 2017). Here we follow these ideas to use the logarithmic function to describe the restoration of stacking size B as a function of the resting time:

$$B = a + b * \log_{10}(t_{\text{rest}}) \quad (11)$$

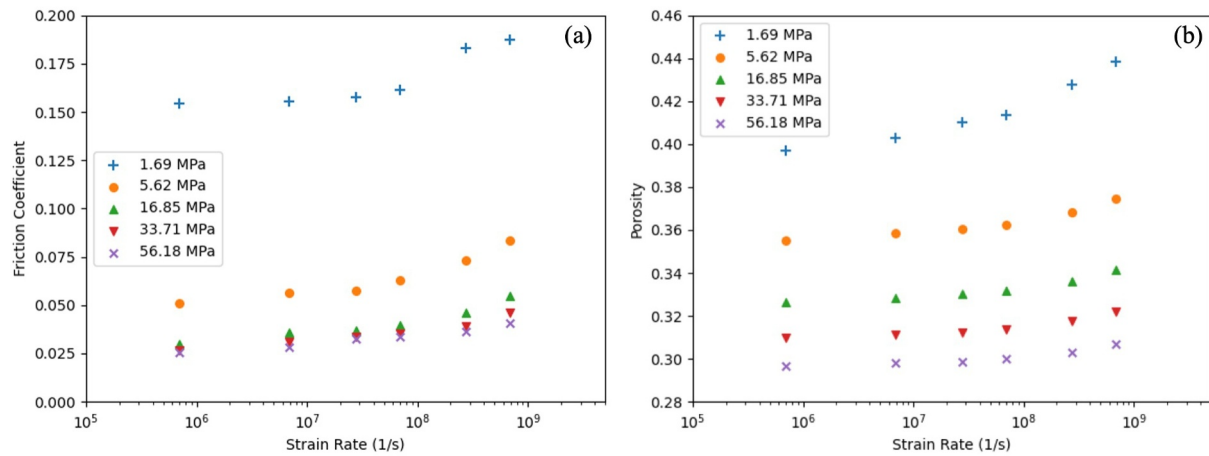


Figure 5. Friction coefficient and porosity as a function of strain rate at different normal stresses.

Here the coefficient b is interpreted as the restoration tendency of stacking size. This logarithmic model serves as a reasonable representation of the data for the constant volume condition in Figure 8a, whereas the logarithmic behavior is less apparent for the constant pressure condition in Figure 8b. The fitting parameters are given in Table S5 in Supporting Information S1.

Figure 8c shows the slope b as the function of the porosity at the end of the rest. Although the rest period starts from the same porosity for the two different conditions, the system is further compressed to decrease the porosity at the constant pressure condition. Contrastingly, the porosity is kept fixed during the rest period under the constant volume condition. The restoration coefficient b is larger at higher porosities. Namely, the restoration of stacking structure is faster with more space to reorganize. However, at constant pressure condition, the pore volume decreases after the cessation of shear. Due to little space for reorganization, the restoration is hindered. The result suggests the extent of restoration is more controlled by the porosity of system at rest.

4. Discussion

4.1. Comparison With Previous Friction Data

The strain rate dependence of shear stress in this study follows the HB model with a finite yield stress at the zero-strain rate limit. This yield stress increases linearly with normal stress, with μ of 0.0135. In comparison, similar linear fitting for yield stress has been used for natural cation-exchanged montmorillonite, which suggests a larger μ of 0.11–0.26 (Behnsen & Faulkner, 2013). C. A. Morrow et al. (2017) found that μ increased from 0.1 to 0.28 between 10 and 600 MPa for wet montmorillonite. The limitation of the model is further discussed in Section 4.4.

Our model clay system exhibits a velocity strengthening behavior, which is consistent with previous experimental studies on wet smectite (Behnsen & Faulkner, 2013; K. M. Brown et al., 2003; Ikari et al., 2007; Moore & Lockner, 2007; C. B. J. B. Morrow et al., 1992). In these experiments, the velocity dependence of steady-state friction is estimated in the framework of the rate and state dependent friction law:

$$(a - b) = \frac{\Delta\mu_{ss}}{\ln\left(\frac{V_2}{V_1}\right)} \quad (12)$$

where $\Delta\mu_{ss}$ is the change in steady-state friction, and V_1 and V_2 are different steady-state shear velocity. In the present simulation, the steady-state shear velocity is given as $\dot{\gamma}L_{z-ss}$, where L_{z-ss} is the average box height in the strain of 8–10. As shown in Figure 9, the estimated $(a-b)$ values are all positive in the investigated range. They generally increase with strain rate and decreases with normal stress. At the lowest strain rate of $6.93 \times 10^5/s$, the $(a-b)$ values are in the range of 0–0.003. This is larger than the experimental values for wet montmorillonite in the range of 0–0.001 at the effective normal stress below 100 MPa (C. A. Morrow et al., 2017). Similarly, Tembe et al. (2010) reported the value of 0.0005 at the effective normal stress of 40 MPa. The discrepancy may be

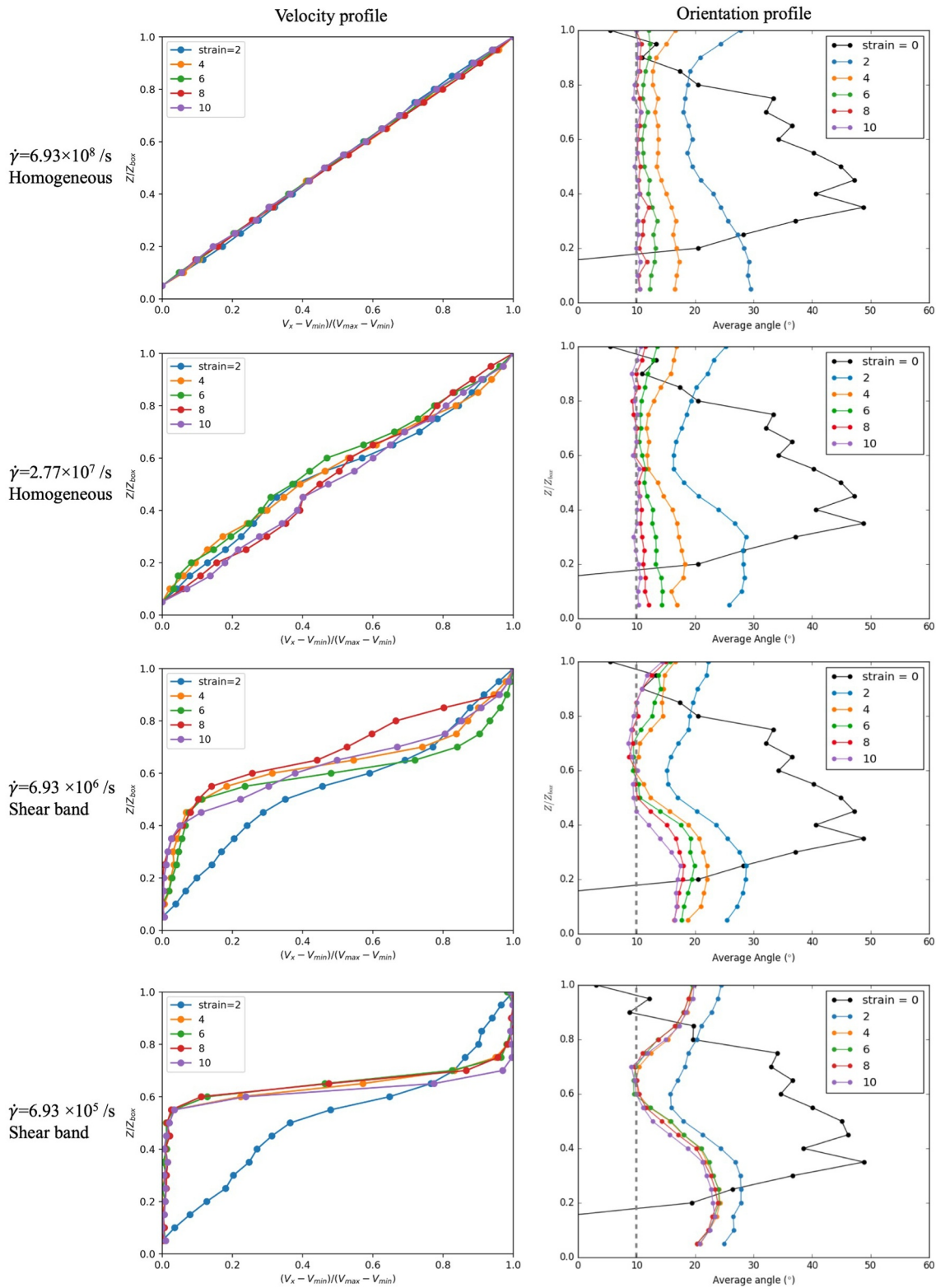


Figure 6.

attributed to the strain rate much larger than the experimental values. Recalling the HB model of Equation 7, the (a–b) value is then $\frac{d\mu_{ss}}{d \ln(V)} = \frac{1}{\sigma_{eff}} \frac{d\tau_{ss}}{d \ln(\dot{\gamma})} = \frac{Kn}{\sigma_{eff}} \dot{\gamma}^n$, suggesting the (a–b) value is overestimated at higher strain rate. This tendency is consistent with the increase in (a–b) with sliding velocities for partially hydrated montmorillonite (Ikari et al., 2007). The (a–b) value extrapolated from the HB model fit at 56.18 MPa using sliding velocities of 0.1–1.0 $\mu\text{m/s}$ is 2.85×10^{-4} , which is comparable to the above experimental data.

4.2. Shear Band Formation and Its Relevance to Experiment and Natural Faults

The formation of shear band can be further quantified using the velocity profile and the particle orientation. Here we quantify the deviation from the linear velocity profile using the quantity below:

$$R^2 = 1 - \frac{\sum_i (v_i - \bar{v})^2}{\sum_i (v_i - \bar{v})^2} \quad (13)$$

where i denotes the discretized Z coordinate with 20 layers, v_i is the average X velocity in layer i , $\bar{v}$ is the X velocity averaged over the entire system, and f_i is the X velocity calculated from the linear velocity profile. When $R^2 \simeq 1$, the velocity profile is close to linear: that is, homogeneous shear is realized.

Since the velocity outside the shear band is not strictly zero, determining the clear shear band from the velocity profile alone can be difficult. Therefore, we also use the information on the particle orientation. When the particles are sheared, their orientation angles tend to be near 10° . As shown in Figure 6, the region with preferred orientation angle ($8^\circ \sim 12^\circ$) overlaps with the larger strain-rate zone shown in the velocity profile. In the following analysis, the thickness of preferred orientation zone ($8^\circ \sim 12^\circ$) is used to define the final shear band width.

The evolution of R^2 and the shear band width is shown in Figure 10, where the band width is divided by the system height and shown as a percentage. At the lowest strain rate, shear banding starts around the strain of 2~3 as seen in the decrease of R^2 . The normal stress does not significantly affects the onset of shear banding. In addition, the final band width of 30% is independent of the normal stress. Comparing to the nematic order change shown in Figure 3d, the particle orientation does not change significantly after the formation of shear band. At intermediate strain rate, the final band width increases and is wider at lower normal stress.

Interestingly, at high normal stress and high strain rate conditions ($\dot{\gamma} \geq 2.77 \times 10^7/\text{s}$), R^2 first decreases and then increases. This nonmonotonic behavior suggests that the system tends to form the shear band initially, but then returns to homogeneous shear. Simultaneously, more particles are aligned in the preferred orientation and the orientation band expands. The nematic order shown in Figure 3d also suggests that particle rearrangement continues in the later stage of shear. Restoration to homogeneous shear is not observed at lower strain rates where the shear band appears to be stable.

At higher strain rate and lower normal stress, the linear velocity profile ($R^2 \simeq 1$) is maintained throughout the shear. The homogeneous shear is defined if the system satisfies the two criteria: (a) R^2 is above 0.9 throughout the shear, and (b) the zone of preferred orientation angle extends to the entire system (100%) at the end of simulation (strain of 10).

The phase diagram of shear banding is shown in Figure 11. At lower strain rates and/or higher normal stress, shear is accommodated in the shear band. Conversely, lower effective normal stress and higher strain rate can facilitate the particle rearrangement to increase the shear band width. This tendency is common to general yield stress materials (Berthier, 2003).

Figure 6. Inspection on shear banding from velocity profile (left column) and particle orientation profile (right column) at the normal stress of 5.62 MPa. The velocity profile is close to linear at high strain rates ($\dot{\gamma} \geq 2.77 \times 10^7/\text{s}$), whereas shear band is formed at lower strain rates ($\dot{\gamma} \leq 6.93 \times 10^6/\text{s}$). The orientation profile shows average angle between the symmetry axis of platelet and the Z axis along the height. The black line represents the initial structure. The particle orientation approaches 10° uniformly at high strain rate while only particles inside certain region approaches 10° at low strain rate. The region of shear band coincides with the region of preferred particle orientation.

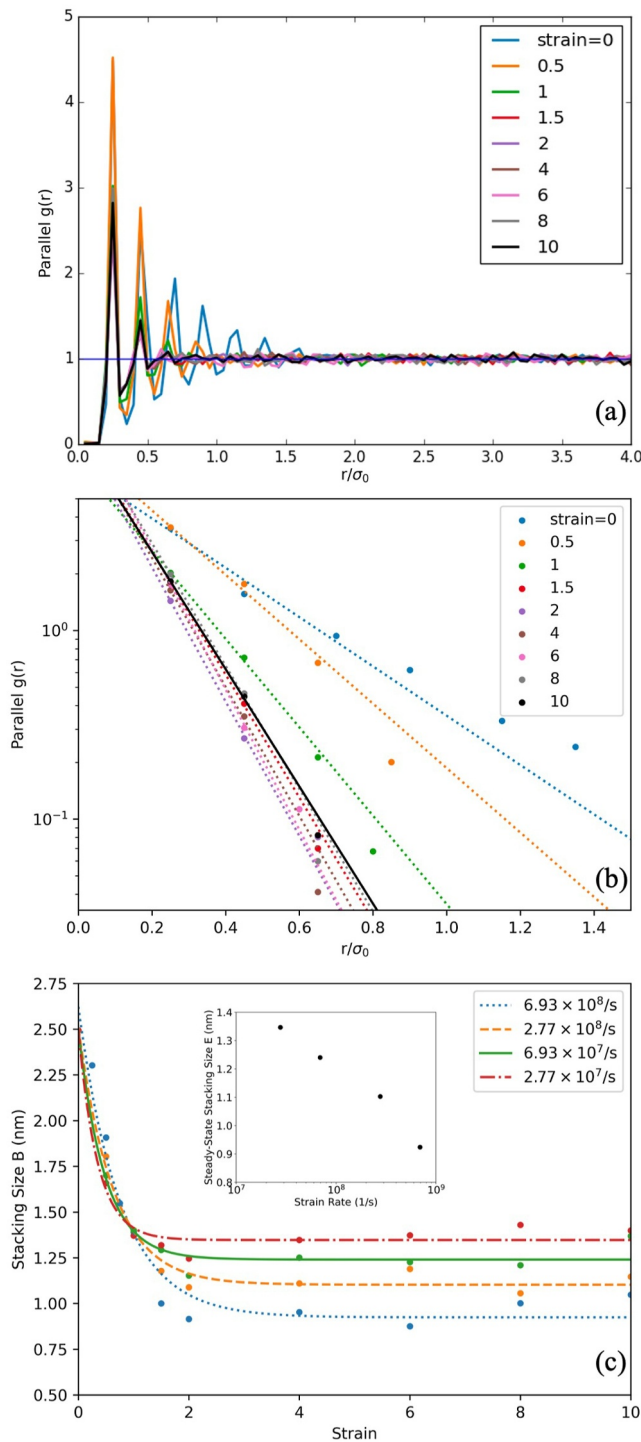


Figure 7. Change in the parallel stacking structure during shear. (a) The parallel stacking structure is first probed with parallel radial distribution function (PaRDF) for the system at $P = 5.62$ MPa and $\dot{\gamma} = 6.93 \times 10^8$ /s. Each curve represents a PaRDF at a certain strain. The peak height and the peak number decrease with shear strain. (b) The peak value as a function of distance is fitted with an exponential function from Equation 9. (c) A characteristic stacking size B is determined from the change in PaRDF and decreases to a residual value during shear following Equation 10. The inset shows the steady-state stacking size E decreases with strain rate.

The “low” strain rate in the simulation is higher than typical experimental values. This means that shear bands should be ubiquitous in natural clay minerals irrespective of their velocity strengthening friction. To support this idea, the tendency of shear banding has been observed in experiments (Haines et al., 2013; Moore & Lockner, 2007). The thickness of the shear band formed in the simulation is comparable to the size of a single particle, which is typically tens of nanometers. Although shear band thickness is difficult to measure in practice, experiments on clay-rich gouges showed a smooth boundary layer of sub-micron thickness, where particles are perfectly parallel to the shear surface (Haines et al., 2013). Saturated montmorillonite samples showed sharp, polished, planar surfaces even for Riedel shear, which accommodates a small amount of strain (Moore & Lockner, 2007). This may suggest that the thickness of the shear band in experiments can be as small as the size of a single particle.

Given the high strain rate adopted in the simulation, the shear in clay-rich fault must be predominantly controlled by the localized shear band at realistic strain rates in natural faults.

4.3. Structure Development During Shear

In this section, we compare changes in shear stress with structural changes. At lower strain rates, porosity decreases rapidly in the initial stage of shear as shown in Figure 3b. This is accompanied by significant changes in the nematic order and the PaRDF (Figures 3c and 7c). Visual inspections (Figure 1), the velocity profile, and the orientation angle profile (Figure 10) indicate homogeneous shear for the first 200% strain (i.e., $\gamma \leq 2$) and the formation of a shear band at later strains ($\gamma > 2$).

The system behavior during shear at low strain rates can be divided into two stages. In Stage I ($\gamma \leq 2$), shear stress increases along with pronounced structural rearrangements, including compaction, particle reorientation, and alignment. During this stage, the parallel stacking structure declines and the shear is relatively homogeneous. Subsequently, in Stage II, the system gradually approaches a steady state. The shear stress decreases to a steady-state value with the formation of shear band. The changes in particle orientation and porosity in Stage II are relatively small.

Kenigsberg et al. (2019) proposed a micromechanical process for clay-rich gouge under shear, following pioneering work of Haines et al. (2013) and Logan and Rauenzahn (1987). Particle reorientation and compaction occur when the shear strain is relatively small. Fabrics such as Riedel shear, boundary shear, and Y shear start to form after the stress peak and contribute to shear stress reduction. Further weakening with progressive shear is controlled by the development of multiple fabrics and compaction. The structural evolution observed in the present simulation at low strain rates is somewhat consistent with these experimental findings, though the present system may be too small to accommodate the development of various shear fabrics and spatial structures.

At the lowest strain rate, the above scenario may hold irrespective of normal stress. Figure S6a in Supporting Information S1 shows that the overall shape of the stress-strain curve remains similar across different normal stress. Additionally, the decrease in shear stress and shear localization occur simultaneously, independent of normal stress (Figure 10). Thus, the timing of shear localization may not be controlled by normal stress.

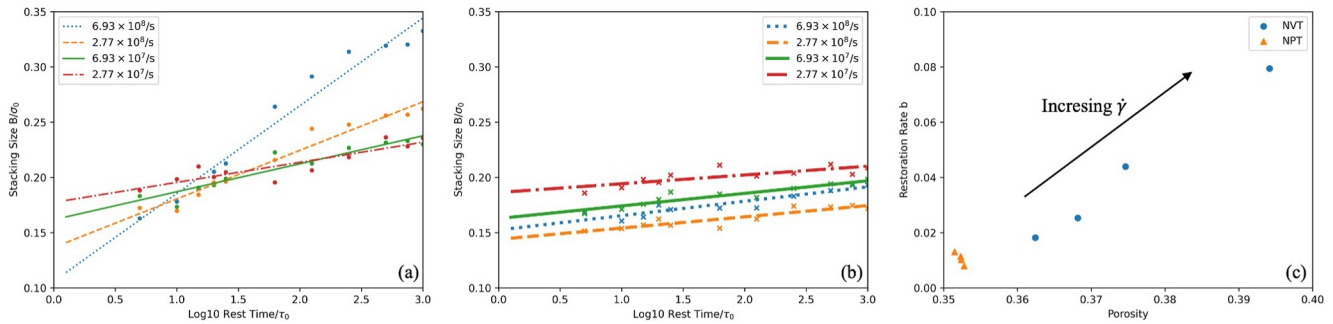


Figure 8. The characteristic stacking size increases with the rest time under (a) the constant volume condition and (b) the constant normal stress condition. The lines represent the logarithmic Equation 11. (c) The slope b , the restoration rate of the stacking size, increases with the system porosity.

In contrast, the above scenario for structure development does not apply to higher strain rates. As shown in Figure 3a and Figure S6b in Supporting Information S1, the shear stress peaks occur at smaller strain with lower normal stress. Compaction continues to larger strains depending on normal stress. Therefore, stage I cannot be simply defined according to the stress peak. The shear is also more homogeneous at high strain rate without apparent shear banding. We suggest the uniform shear can extend the period of compaction and overall structure rearrangement.

4.4. Limitations of the Model

4.4.1. Shape and Interactions

Natural clay platelets have a much larger aspect ratio than those used in the present model. A typical size distribution for Na-montmorillonite ranges from 5 to 750 nm in diameter, with an average of 250 nm (Michot et al., 2004). Thus, the present study only covers smaller end of the natural clay platelet size distribution. As a result, the shear stress may be underestimated during the initial stage of shear (stage I) as the initial rearrangement and rotation of larger platelets would be more difficult. A larger aspect ratio may also lead to more prevalent shear localization.

Regarding particle interactions, the GB potential is used in the present study with implicit solvent and ions. Although calibrated from the full atomistic MD simulation under equilibrium conditions, this potential may not fully capture the effects of dynamic shear. Various structural changes, such as the reduction of interlamellar water layers, may occur due to shear, which would reduce lubricity and increase shear resistance (Moore & Lockner, 2007).

4.4.2. Limitations in Time and Size

In general, MD simulations have inherent limitations in terms of time and length scales compared to real systems. The timescale limitation results in an unrealistically high strain rate (Ewen et al., 2018). The strain rates investigated in this study are therefore much higher than those in typical experiments, where the gouge thickness is a few mm and the loading rate is on the order of $\mu\text{m/s}$. At sufficiently high strain rates, various thermal effects may occur, such as thermal pressurization of pore fluid (Rice, 2006). These effects are not considered in the present simulation, where the temperature is kept constant during shear.

The system size in this study is limited to the nanometer scale. Previous experimental studies suggest that the formation of various fabrics and shear bands plays a key role in reducing the macroscopic frictional strength in clay-rich gouges (Collettini et al., 2019; Haines et al., 2013; Kenigsberg et al., 2019). Such large-scale fabrics cannot be formed in small simulation

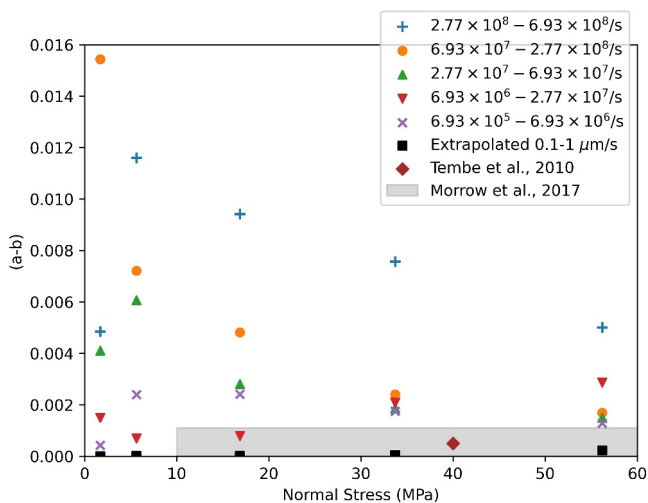


Figure 9. The velocity dependence of friction, (a-b), as a function of normal stress for different strain rate. The average (a-b) values are given in black circles.

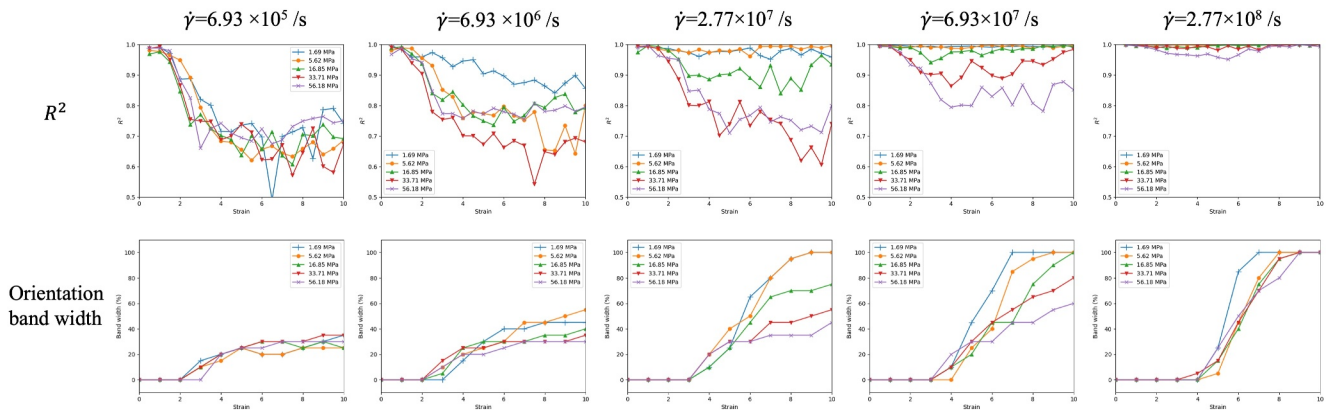


Figure 10. The change in deviation from linear velocity profile characterized by R^2 (top row) and the estimated final shear band width determined from preferred orientation band (bottom row) at different strain rate for each normal stress. The preferred orientation is normalized by the sample height and shown in percentage. A low R^2 and finite band width at lower strain rates shows the shear is localized. At higher strain rates, R^2 is closer to 1 and band width can reach the full sample height suggesting a more homogeneous shear.

systems. Fabric formation also depends on the presence of larger and stronger minerals, such as quartz (Colletini et al., 2019; Tesei et al., 2012). Such mineral heterogeneity is not considered in the current system.

Since the computational cell in the present simulation is only tens of nanometers, it can be interpreted as a magnification of a region including a shear band in an experimental specimen or in a natural fault. The true strain rate at the shear band, estimated from experiments or natural faults, can be as large as 105/s when the slip velocity is 1 mm/s, and the shear band thickness is 10 nm. Therefore, high strain rates in the present simulation may not be so unrealistic. In such a situation, the frictional heat may not be negligible as discussed above. However, at least in the present parameter range, the comparable (a–b) values with experiments may partially support the validity of the present simulation.

5. Conclusions

The rheology and structure of a model smectite clay are studied using MD. The particle shape is simplified to an oblate ellipsoid, and the GB potential is adopted as a simplified interaction model. Despite the simplifications, the present exploratory model is able to capture key features of a clay system. The system exhibits velocity-strengthening behavior described by the HB law. The estimated (a–b) values are comparable to those obtained in experiments and increase with strain rates.

Shear localization is observed at low strain rate conditions despite velocity-strengthening friction. Since the “low” strain rate in simulation is still much higher than typical experimental values, shear localization is expected to be ubiquitous in experiments on smectite. The shear is accommodated in a narrow region where the particles align with the preferred orientation. More uniform shear is achieved at high strain rates.

The structure development at low strain rates aligns with the micromechanical process hypothesized in previous experimental studies. In the first stage, the particles rearrange themselves to align with shear ($\gamma \leq 2$), causing the shear stress to rise to a maximum. In the following stage, a shear band starts to form, and the shear stress gradually decreases to a steady-state value.

We introduce the PaPDF to examine changes in parallel stacking structure. The characteristic stacking size decreases with the shear strain and undergoes logarithmic healing over time during the rest period. The amplitude of the restoration rate increases with the porosity of the resting system. Although it may give some insights to the healing process in natural fault, one must note

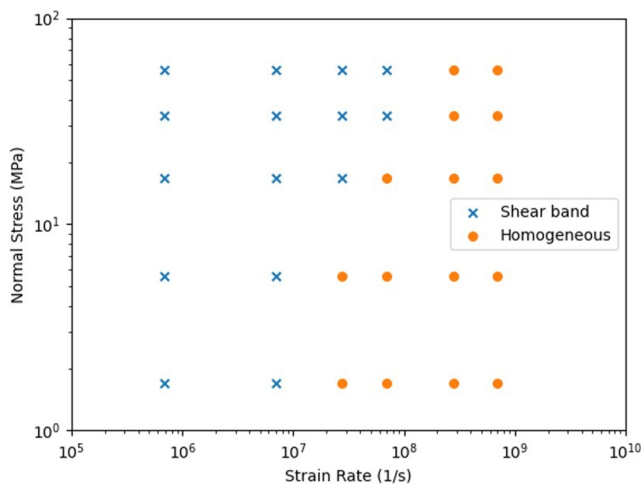


Figure 11. The phase diagram of shear band in the range of normal stress and strain rate investigated. Shear band is more likely to occur at lower strain rate and higher normal stress.

that the timescale is very different. Further studies are required to connect the healing behavior in simulation and the healing processes in natural clay-rich faults.

Although simple, the present clay model with oblate ellipsoids and the GB potential can be useful for studying the rheology and microstructure of clay systems. For future improvement, a broader size distribution can be adopted in larger systems to study the large-scale structure. While steady state shear is adopted in the current study, other shear configurations should also be considered to provide more information on the stability and rheology of the model clay system. Understanding the effects of particle shape and interaction may also help generalize the present findings to other phyllosilicate minerals.

Data Availability Statement

The computer codes for LAMMPS simulations of this paper with LAMMPS version information is available through the Zenodo digital repository at (Lin & Hatano, 2024). Figures are made with the Python library Matplotlib version 3.6.1 (Caswell et al., 2022; Hunter, 2007), available under the Matplotlib license. Visualization in Figure 1 and part of the data analysis are carried out using the visualization and analysis software OVITO (Stukowski, 2009). The Supporting Information contains Text S1, Tables S1–S5, and Figures S1–S6 in Supporting Information S1.

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