Atomic-scale interlayer friction of gibbsite is lower than brucite due to interactions of hydroxyls

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\section*{ABSTRACT}

To investigate the role of atomic-scale structure on the frictional properties of gibbsite, a dioctahedral-type aluminum hydroxide, we calculated the atomic-scale interlayer shear properties using the first-principles method based on density functional theory. We found that the presence of vacant sites within the octahedral sheet of gibbsite enables hydroxyls to move to more stable positions and reduce the repulsive force, leading to a lower atomic-scale shear stress of gibbsite compared with brucite, a trioctahedral-type magnesium hydroxide. We also estimated the macroscopic single-crystal friction coefficient of gibbsite with the assumption that only the atomic-scale interlayer friction controls macroscopic friction. The estimated single-crystal friction coefficient for gibbsite is 0.36(6), which is clearly lower than the experimentally obtained friction coefficient of the powdered gouge of gibbsite (0.74). This difference between the interlayer friction coefficient and gogue friction coefficient suggests the presence of additional mechanisms that affect the frictional strength, such as microstructures within a fault gouge.

\textbf{Keywords:} Layered structure minerals, interlayer friction, gouge friction, gibbsite

\section*{INTRODUCTION}

Frictional strength is a fundamental physical property that controls the localization of deformation within a fault zone. Layered structure minerals have lower friction coefficients than common rock-forming minerals (Byerlee 1978; Morrow et al. 2000; Moore and Lockner 2004). The presence of layered structure minerals can cause creep in the San Andreas fault (Carpenter et al. 2011; Lockner et al. 2011), activate low-angle normal faults (Viti and Collettini 2009; Collettini et al. 2019), and reduce frictional strengths of plate boundaries in subduction zones (Ikari et al. 2018; Okuda et al. 2021b). The low-friction coefficients of layered structure minerals have been explained by their characteristic crystal structure (Morrow et al. 2000; Moore and Lockner 2004; Behnsen and Faulkner 2012; Kawai et al. 2015; Sakuma and Suehara 2015; Niemeijer 2018; Okamoto et al. 2019; den Hartog et al. 2020). The frictional strength between the layers of layered structure minerals critically affects the low-friction coefficients of these minerals, as demonstrated by friction experiments of single-crystal phyllosilicates (Kawai et al. 2015; Niemeijer 2018; Okamoto et al. 2019). Based on the importance of interlayer shear sliding properties, we recently focused on the atomic-scale interlayer interactions and quantitatively evaluated its influence of interlayer sliding on macroscopic frictional properties (Sakuma et al. 2018, 2020, 2022; Okuda et al. 2019). These studies are hereinafter referred to as SKKS18, SKK20, SLSD22, and OKS19, respectively. In SKKS18, the estimated macroscopic friction coefficient of muscovite was found to be nearly identical to the experimentally obtained friction coefficient of single-crystal muscovite. OKS19 and SKK20 estimated slightly higher and lower single-crystal friction coefficients for brucite and pyrophyllite, respectively, compared with that for muscovite. SLSD22 found that the friction coefficients for interlayer sliding of montmorillonite positively depend on the ionic radii of interlayer cations. These studies suggested that the difference in frictional properties for the interlayer sliding potentially plays a role in the difference in macroscopic friction coefficients of layered structure minerals.

In this study, we simulated the single-crystal friction coefficient of gibbsite using density functional theory. Gibbsite [Al(OH)$_3$] has a similar crystal structure as that of brucite [Mg(OH)$_2$], which was studied in OKS19. Experimentally obtained friction coefficients of the gouges of gibbsite and brucite were quite different; the value for gibbsite is 0.74, whereas that for brucite is 0.39 (Moore and Lockner 2004; Okuda et al. 2021a). To evaluate their interlayer frictional properties and their roles in their single-crystal and gogue friction coefficients, herein, we discuss the difference in friction coefficients of gibbsite and brucite and other layered structure minerals based on theoretical consideration of the atomic-scale shear deformation.

\section*{MATERIALS}

Al(OH)$_3$ gibbsite is a dioctahedral-type layered structure mineral (Fig. 1a). The space group is $P2_1/n$ with lattice constants of $a = 8.684(1)$ Å, $b = 5.078(1)$ Å, $c = 9.736(2)$ Å, and $\beta = 94.54(1)^\circ$ under ambient-pressure conditions (Saalfeld and Wedde 1974). The primitive unit cell of gibbsite contains two sheets of Al octa-