



Research paper

Crystal-chemistry control of the mechanical properties of 2:1 clay minerals



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ABSTRACT

Clay minerals are the main constituents of the clay matrix of a wide variety of sedimentary deposits. When subjected to burial, some of these minerals undergo phase transitions accompanied by atomic substitutions which ultimately impact the cohesive interactions between their constitutive layers. The most common among such transitions is smectite illitization, which is also highly relevant for oil and gas exploration and production from source rocks. The impact of this transition on the mechanical properties of clay minerals as well as clay bearing rocks remains, however, to be properly addressed. To this end, a set of macroscopic single 2:1 clay minerals (pyrophyllite, talc, vermiculite, phlogopite, muscovite, and clintonite) representative of the two octahedral fillings and the variation in surface charge densities was investigated. A hybrid experimental-modeling approach is proposed, which combines nanoindentation in orthogonal directions (x_1 and x_3 -directions) with analytical derivations of the cohesive energy (U_i) using XRD and TEM-EDS measurements. The results highlight that the interlayer energy defines the elasticity (modulus M), strength (hardness H) and ductility behavior (M/H) of these materials; and not the bond energy stored into the constituent layers. This finding permits the derivation of predictive stiffness and strength functional relations as derivatives of the interlayer energy that account for the arrangement of nanoscale layers and the interlayer composition. These relations suggest that as the cohesion increases with the coulombic interactions through progressive smectite illitization, the system loses its capacity to dissipate applied energy by dislocation mechanisms in between the layers, entailing an increase in brittleness of the clay particles with burial. By way of conclusion, the geophysics and geochemistry implications of these results for predicting the macroscopic mechanical performance of clay bearing geomaterials, such as economically valuable source rocks, are discussed.

1. Introduction

Clay minerals are among the major components of the fine grained matrix of sedimentary deposits such as shales and mudrocks (Chamley, 2013), and play an important role in the overall mechanical behavior of economically valuable source rocks (Ulm and Abouseleman, 2006; Bobko and Ulm, 2008; Ortega et al., 2009; Deirieh et al., 2012; Abedi et al., 2016a). Among the clay minerals encountered in these deposits, smectite and illite contents evolve with respect to increases in temperature and pressure experienced during burial (Meunier and Velde, 1989; Stixrude and Peacor, 2002; Kennedy et al., 2006; Peltonen et al., 2009; Ferrage et al., 2011). Smectites settle as detrital inputs within the water column in sedimentary basins (Chamley, 2013). Upon burial, they undergo a mineral transition toward illite according

to a reaction called smectite illitization (Meunier and Velde, 1989; Lanson et al., 2009; Ferrage et al., 2011). This reaction can be summarized as an evolution of the permanent layer charge, driven by atomic substitutions, which ultimately impacts the interlayer cohesive interactions. It involves the release of a significant amount of silica and contributes to the changes in physical properties of mudstones recorded on well logs (Peltonen et al., 2009). This process also occurs concomitantly with the maturation of organic matter that leads to the yield of oil and gas (Seewald, 2003). Therefore, understanding the sole impact of smectite illitization on the mechanical response of shale's clay matrix is of prime importance to link geochemistry with geophysics and rock mechanics.

Clay minerals display pseudo bi-dimensional crystal structures resulting in the junction of tetrahedral (Si_2O_5) and octahedral (R^{2+}_3 ,

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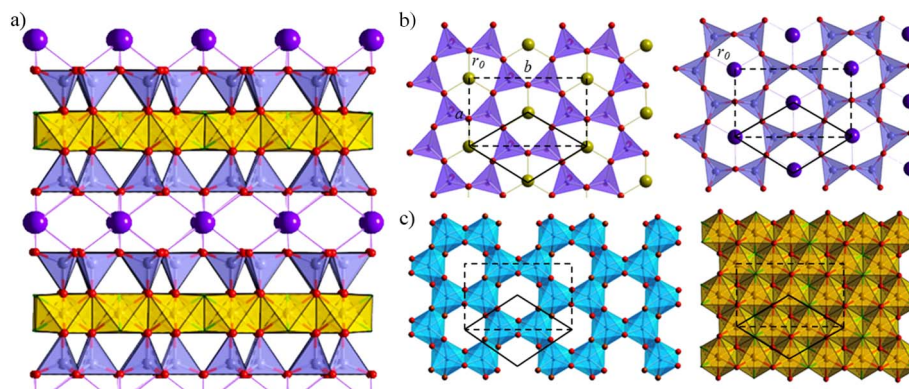


Fig. 1. Crystal structure of 2:1 phyllosilicates. a) Projected crystal structure of the 2:1 layers according to $\langle 100 \rangle$ constituted by the tetrahedral (blue) and octahedral (yellow) sheets and the interlayer cations (purple). b) Top-views of the tetrahedral sheets in di-octahedral (left, showing ditrigonalization) and tri-octahedral (right) configurations. c) Top-views of the octahedral sheets in di-octahedral (left) and tri-octahedral (right) configurations. The C-centered Bravais unit cell and the primitive unit cell are displayed in dashed and solid lines, respectively. The dimensions a and b are the Bravais unit-cell parameters, and r_0 is the distance between the interlayer cation and its nearest neighbor (apical oxygen in red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

$R^{3+}_2(O, OH)_6$ sheets, see Fig. 1. The wider clay mineral family encountered in sediments is commonly referred to as 2:1 clay minerals. They are composed of a succession of layers of two tetrahedral sheets sandwiched by an octahedral sheet, and separated by an interlayer space. Isomorphic substitutions inside the different sheets drive the evolution of cohesive bonds between successive layers, which can be viewed as long-range interactions. The whole spectrum of the permanent charges is commonly used to classify the 2:1 clay minerals from talc and pyrophyllite to brittle micas (Bleam and Hoffmann, 1988; Guggenheim et al., 2006). Within this classification, smectite and vermiculite display low charges that allow hydration of the interlayer cations. This ultimately impacts the basal spacing (d_{001}), entailing an expansion. In contrast, illite which is often described as altered mica, is non-expansive.

The irregularities of a material's micro to nano structure (porosity, grain boundary, dislocation, or atomic bonding) are usually thought to be responsible for its hardness. The elasticity of bulk materials is also controlled by the atomic bonding (ionic, covalent, or metallic bonds). The elastic deformation of the loaded bulk causes reversible bond stretching, while inelastic deformation usually involves bond rearrangement or breakage (Baker et al., 2002). In clay minerals, the interlayer interactions are generally much weaker than the ionic-covalent bonds building the 2:1 layers (Giese, 1974). The weak cohesive bonding along the stacking direction allows the rigid 2:1 layers to act almost as dislocation walls. Thus, any compositional variations within the crystalline framework of the 2:1 clay minerals are expected to affect their mechanical properties (Kunz et al., 2009; Zartman et al., 2010; Fu et al., 2011).

The elastic properties of 2:1 clay minerals have been studied through a wide range of techniques, such as Brillouin scattering (Vaughan and Guggenheim, 1986; McNeil and Grimsditch, 1993); nanoindentation (Zhang et al., 2009a, 2009b, 2010, 2013); molecular dynamics simulations (Carrier et al., 2014; Hantal et al., 2014); and digital image correlation (Hedan et al., 2015). Among the reference values thus available in the literature, it is worthy that the results obtained using nanoindentation on an ensemble of 2:1 clay minerals probed normal to the crystal planes suggested a linear relationship between the reduced elasticity modulus and the layer charge (Zhang et al., 2009a, 2010, 2013, see also discussion by Bobko et al., 2009). Although sound, the approach, however, did not account for distinct mechanical and crystallographic features of such systems, namely their elastic anisotropy, their resistance to plastic deformation (hardness), and the specific interactions between successive layers. To address these issues, the focus of this study is to quantitatively pin down the link between mechanical properties (elasticity, hardness, ductility) of 2:1 clay minerals and the relevant bonding energies with respect to

their anisotropy.

To this end, six single crystals representative of the wide range of layer charges and octahedral compositions typically found in the 2:1 clay minerals family, were studied. First, a characterization of their crystal chemistry was performed by coupling X-ray diffraction and energy dispersive spectroscopy. Second, the mechanical properties of these materials were assessed by means of nanoindentation in orthogonal directions representative of the crystallographic directions. In fact, owing to their layered structure, clay minerals are expected to exhibit elastic and plastic anisotropy comparable to orthotropic solids (Delafargue and Ulm, 2004). Finally, by linking the cohesive coulombic energy with mechanical properties, the proposed approach aims at identifying the impact of the arrangement of nanoscale layers and interlayer composition on fundamental physical and mechanical properties. Last, the implications of these results for predicting the macroscopic mechanical performance of clay bearing materials such as economically valuable source rocks are discussed thanks to the consideration of theoretical and experimental data available in the literature (Carrier et al., 2014; Hantal et al., 2014; Abedi et al., 2016a).

2. Materials and methods

2.1. Crystal-chemical characterization

Six single crystals of 2:1 clay minerals (pyrophyllite, talc, vermiculite, phlogopite, muscovite, and clintonite) from the mineral collection of Aix-Marseille University (CINaM/CNRS) were studied. Their unit cell dimensions were defined using X-ray diffraction (XRD) on homogenized powders with a Bruker D8 Focus diffractometer (Co $K\alpha$ radiation) equipped with a Lynx'Eye detector operating with an aperture of $1^\circ 2\theta$. Diffracted intensities were collected at $0.02^\circ 2\theta$ step intervals from 4 to $60^\circ 2\theta$, using a 1 s counting time per step. The divergence slit and the two Soller slits were 0.3 , 2.5 , and 2.5° respectively.

The structural formulas of the studied samples were assessed from raw micro analytical data recorded by a transmission electron microscope (TEM, JEOL JEM 2011) coupled with an energy-dispersive X-ray spectrometer (EDS, Bruker X-Flash Silicon Drift Detector 5030). Small fractions of the samples were ground in an agate mortar, and drops of particle suspensions in water were deposited onto a carbon coated TEM copper slot. A magnification of 50,000; angular tilt of 20° toward the detector; time constant of 60 kcp s^{-1} ; energy range of 40 keV; and corrected counting time of 30 s were used as acquisition parameters. The beam diameter was set to approximately 20 nm in order to reach the smallest particles. The constant beam density was about 63.5 pA cm^{-2} . Analyses were collected from a minimum of twenty-five individual particles per sample. The main elements (O, Na, Mg, Al,

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