Applied Clay Science 116–117 (2015) 248–256

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Research paper

Measurement of the elastic properties of swelling clay minerals using the digital image correlation method on a single macroscopic crystal

Stephen Hedan [a,*], Fabien Hubert [a], Dimitri Prêt [a], Eric Ferrage [a], Valéry Valle [b], Philippe Cosenza [a]

[a] Université de Poitiers, CNRS UMR 7285 IC2MP, HydrASA, Ecole Nationale Supérieure d'Ingénieurs de Poitiers, 1 rue Marcel Doré, 86022 Poitiers Cedex, France
[b] Université de Poitiers, CNRS UPR 3346 Institut PPRIME, S.P. 2 M.I., Bd. M. et P. Curie, Téléport 2, B.P. 30179, 86962 Futuroscope Chasseneuil Cedex, France

A R T I C L E I N F O

Article history:
Received 7 January 2015
Accepted 2 April 2015
Available online 21 April 2015

Keywords:
Vermiculite
Swelling clay mineral
Digital image correlation
Elastic modulus

A B S T R A C T

This paper presents a new methodology for determining the elastic moduli of swelling clay minerals based on the combination of two original elements: (a) a natural multi-millimetric vermiculite crystal considered as a model system of swelling clay minerals and (b) a mechanical device that allows for mechanical compression in a controlled hydric state while concomitantly measuring the resulting strain field using the digital image correlation (DIC) method. Following this approach, the DIC method can (a) identify anomalous areas of the strain field to be excluded from an elastic analysis and (b) not introduce any assumptions for Poisson's ratio, as often required in other techniques (e.g., nanoindentation).

This approach was applied to a vermiculite crystal, with dimensions of 5×5 mm^2 and a thickness of 1 mm; the value obtained for the undrained uniaxial Young's modulus was 32.3 ± 3.9 GPa. To the authors' knowledge, this is the first value for Young's modulus proposed in the literature for a natural and pure swelling clay mineral and is consistent with those obtained from synthetic samples and molecular dynamics simulations.

1. Introduction

Quantifying the elastic properties (e.g., Young's modulus, Poisson's ratio or equivalent shear modulus and bulk modulus) of swelling clay minerals is significant for several scientific issues. In mineral physics, investigating how the mineral elasticity of nanoscale clay minerals and the distribution of water molecules in the interlayer space affect the mechanical behavior of the clay crystals or tactoids is of scientific interest (e.g., Ulm et al., 2005; Ichikawa et al., 2004; Carrier et al., 2014; Ebrahimi et al., 2014). In geophysics, because clay minerals are an integral part of many geological formations, reservoir characterization using seismic waves requires the elastic parameters of solid components and pore fluids to be known (e.g., Wang et al., 2001; Vanorio et al., 2003; Mondol et al., 2008; Mavko et al., 2009). In geomechanics, micromechanical or multiscale modeling of the mechanical behavior of clayey geomaterials uses the values of elastic constants of clay minerals at the lowest scale as input data (e.g., Giraud et al., 2007; Ortega et al., 2007; Cariou et al., 2013).

However, the elastic properties of clay minerals and *a fortiori* those of swelling clays, unlike many other minerals, minerals are rarely found in handbooks (e.g., Schon, 1996; Mavko et al., 2009). This scarcity of values stems from two facts: (a) clay minerals are not sufficiently large for conventional mechanical techniques and (b) for swelling clay minerals, the hydric state, i.e., the amount of the interlayer water molecules in crystals, must be correctly controlled. Despite these difficulties, several attempts have been made to estimate elastic properties of clay minerals by using experimental and theoretical approaches. These attempts can be divided into three groups based on the nature of the clay system.

In the first group, the elastic properties are indirectly obtained from water–clay composites or mixtures using ultrasonic measurements (e.g., Wang et al., 2001; Vanorio et al., 2003; Mondol et al., 2008), Atomic Force Acoustic Microscopy (Prasad et al., 2002), nanoindentation (Bobko and Ulm, 2008) or by coupling these techniques (Deirieh et al., 2012). These approaches can be considered indirect methods because they most often use homogenization models and a fitting procedure to back-calculate the elastic tensor of the clay minerals. However, they have at least two limitations. First, they often introduce assumptions in their back analysis. For instance, Vanorio et al. (2003) and Mondol et al. (2008) extrapolated their elastic data to zero porosity to estimate the elastic properties of clay minerals. Prasad et al. (2002) assumes a priori a Poisson's ratio value of 0.3 for their clay samples that were supposed to be isotropic. Second, most studies did not control the hydric state of the studied water–clay composites, and thus, the resulting estimates of elastic constants were not associated with a bulk water content or known interlayer water content, a key parameter for swelling clay minerals.

In the second group of approaches, the elastic constants of clay minerals are determined on a single clay mineral by using Brillouin spectrometry (Vaughan and Guggenheim, 1986; McNeil and Grimsditch, 1993) or nanoindentation (Basu et al., 2009; Zhang et al., 2009). These

* Corresponding author. Tel.: +33 5 49 45 35 48; fax: +33 5 49 45 36 34.
E-mail address: stephen.hedan@univ-poitiers.fr (S. Hedan).

http://dx.doi.org/10.1016/j.clay.2015.04.002
0169-1317/

approaches have from the same assumptions than the previous approaches: the nanoindentation technique used on a single crystal requires back-calculations and corrections to obtain the elastic constants. Zhang et al. (2009) postulated an a priori Poisson's ratio value of 0.25 for their samples that were assumed to be isotropic i.e., their actual layered structure of clay minerals was not taken into account. Moreover, to our knowledge, these approaches have primarily been applied to non-swelling clay minerals (mainly muscovite).

The third group of approaches addresses theoretical clay systems. Here, the elastic properties are primarily obtained from molecular dynamics (MD) simulations. Although these types of calculations have been applied to clay minerals (e.g., Shroll and Smith, 1999), the theoretical estimation of elastic properties as a function of hydration is rather recent (Militzer et al., 2011; Carrier et al., 2014). The results emphasized the crucial role of the interlayer water molecules on the mechanical response of the swelling clay crystal. However, the values obtained by MD simulations for the swelling clay still remain to be cross-validated as the elastic moduli reported for the dehydrated dioctahedral montmorillonite (7 GPa) are really lower than the previous experimental values reported for dioctahedral mica (50 to 150 GPa).

The new methodological approach described in this work aims to provide values of elastic moduli directly obtained on a single swelling clay crystal in a controlled hydric state, with no assumption for the elastic constants and without using sophisticated homogenization techniques or idealized isotropic behavior. The originality of this approach stems from the coupling of two elements: (a) an original object of study, i.e., a multi-millimetric vermiculite crystal considered as a model swelling clay system and (b) an original mechanical device that allows for mechanical load (compression) in a controlled hydric state and to concomitantly spatialize the resulting strain field using the digital image correlation (DIC) method. This non-invasive optical method allows determining the displacement and strain fields (i.e. maps) of an interest zone from deformation of speckle (variations of gray level). First, this methodological approach is described, and then preliminary results on elastic modulus are presented and compared to previous estimates from the literature.

2. Materials and methods

2.1. Material description and sample preparation

The swelling clay sample investigated in this work was a centimeter-sized crystal of a natural vermiculite from Santa Olalla (Huelva, Spain). This macrocrystal of vermiculite is considered in this work to be an interesting analog of a nanometric swelling clay particle constituted in 2:1 clay layer and interlayer water.

The unit formulas of vermiculite reported by Argüelles et al. (2010) and in agreement with previous reports (De la Calle et al., 1977, 1978 and De La Calle and Suquet, 1988, Mareschal et al., 2009 and Marcos et al., 2003) have layer charges of 0.82 and 0.80 per $O_{10}(OH)_2$, respectively, resulting from Al-for-Si substitutions in the tetrahedral sheet. This vermiculite is mainly Mg^{2+}-saturated in the interlayer corresponding to almost 0.4 interlayer cations per $O_{10}(OH)_2$.

The pluri-millimetric vermiculite monocrystal was first prepared by gentle sawing to obtain a sub-parallelepiped shape (Fig. 1). It was then immersed in hydrochloric acid (1 g of sample in 20 mL of 10^{-4} mol L^{-1} HCL solution for 5 min) to dissolve carbonates and then washed three times in distilled water. Most of the sub-millimetric oxides were removed from the suspension using a magnetic stick. The sample was originally Mg-saturated, indicating the presence of two water sheets in the interlayer space (De la Calle and Suquet, 1988).

Our model swelling clay system, i.e., the macrocrystal of vermiculite is a natural material and could thus present some viewable cleavages due to its millimeter size. These heterogeneities could affect the macroscopic strain measurements and thus must be checked. As explained below, the DIC method is suitable for recovering spatialized kinematic parameters (displacement fields and strains) and hence to discard any mechanical effects induced by possible heterogeneities.

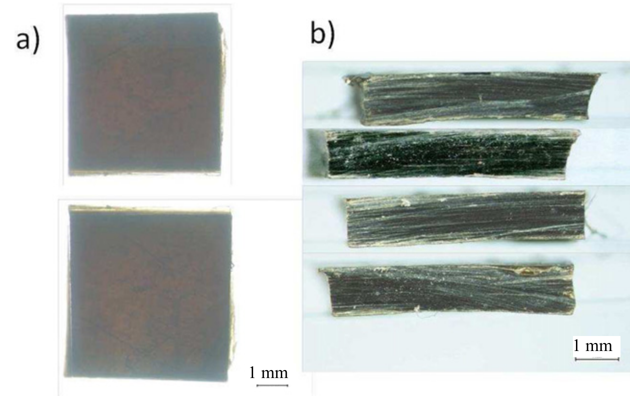

Fig. 1. Pluri-millimetric monocrystal of vermiculite. Right (a): The basal surfaces. Left (b): The four lateral surfaces of monocrystal.

2.2. Digital image correlation method

DIC is intrinsically a non-invasive and non-destructive approach (e.g., Bruck et al., 1989; Hild et al., 2002; Bornert et al., 2009). This approach is simple to use and can be applied in many experiments on materials and for various scales. In the geosciences, the ability of the DIC method to observe strain localization in geomaterials (e.g., Bésuelle et al., 2000) and to detect cracks in clay rocks (e.g., Hédan et al., 2012, 2014) has been demonstrated. Moreover, this optical method coupled with environmental scanning electron microscopy (ESEM) has recently been applied to clay-rocks (Wang et al., 2013) and clay films (Carrier et al., 2013).

To obtain the displacement field of an area undergoing mechanical transformation, the DIC method monitors the positional changes of a speckle pattern located on the analyzed surface. The speckle pattern is typically the result of a random spatial variation of light intensity and can also be obtained artificially by painting the surface of the sample. In our case, a natural speckle was generated during the sawing of the sample (Fig. 1b).

To obtain the full-field displacements from DIC, two images are necessary, a first gray-level function $f(x,y)$ acquired at the initial state t_0 and a second gray-level function $g(x^*,y^*)$ acquired at an actual state t. The principle of this method is to minimize a correlation function CF on a given small and square subset S:

$$CF\left(u,v,\frac{du}{dx},\frac{du}{dy},\frac{dv}{dx},\frac{dv}{dy}\right)=1-\frac{\int_S f(x,y)g\left(x^*,y^*\right)dxdy}{\sqrt{\int_S\left(f(x,y)-\overline{f_s}\right)^2dxdy\int_S\left(g\left(x^*,y^*\right)-\overline{g_s}\right)^2dxdy}} \quad (1)$$

where u and v are the horizontal and the vertical components of the displacement vector, respectively; $u=x^*-x-\frac{du}{dx}x-\frac{du}{dy}y$ and $v=y^*-y-\frac{dv}{dx}x-\frac{dv}{dy}y$.

The kinematic transformation is fully characterized by simple in-plane translations (u, v) and the first gradients $\left(\frac{du}{dx},\frac{du}{dy},\frac{dv}{dx},\frac{dv}{dy}\right)$. The components of the displacement vector (u, v) are obtained in the center of the considered subset S. $\overline{f_s}$ and $\overline{g_s}$ are the averaged gray level on the considered subset in the initial and actual states, respectively. This procedure is repeated for all subsets constituting the image.

In our study, the strain fields were calculated from displacement fields/maps by considering two aspects. First, the strain fields ε_{ij} $(i,j =$