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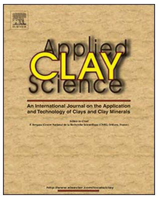

Research paper

Heating-freezing effects on the orientation of kaolin clay particles

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ABSTRACT

The effects of temperature changes on the particle orientation of a consolidated kaolin are studied using XRD experiments. Two sets of equipment were utilized in this study: a benchtop equipment, and a synchrotron beamline at the National Synchrotron Light Source II (NSLS-II) at Brookhaven National Laboratory. The kaolin specimens tested in the benchtop XRD were subjected to elevated and freezing temperatures ex-situ, while those used for the NSLS-II experiment were exposed to the temperature changes in-situ. The temperatures considered in this study range from freezing (− 10 °C) to elevated temperature below boiling (90 °C). The thermally-induced reorientation of clay mineral particles is highly dependent on the relative orientation of the clay mineral particles with respect to the applied thermal gradient. For example, kaolin samples with kaolinite particles oriented perpendicular to the thermal gradient, and to the expected thermally-induced pore water flow, experience much higher particles reorientations compared to samples with particles initially oriented parallel to the thermal gradient. Moreover, freezing kaolin preserved its microstructure as ice crystals form.

1. Introduction

The behavior of earth surface materials, i.e., soils and rocks, subjected to combined thermal and mechanical loads, commonly referred to as thermo-hydro-mechanical behavior, has gained an increasing interest over the last few decades. Depending on the considered application and the expected temperature range, the focus of each study in the literature varied; some concentrated on freezing-thawing (Alkire and Morrison, 1983; Broms and Yao, 1964; Chamberlain and Gow, 1979; Czurda and Hohmann, 1997; Garham and Au, 1985; Konrad, 1989; Norrish and Rausell-Colom, 1962; Ogata et al., 1985; Othman and Benson, 1993; Tang and Yan, 2014; Wang et al., 2007a; Wang et al., 2007b; Wang and Lei, 2017; Yong et al., 1985) while others considered heating-cooling (Abuel-Naga et al., 2007a,b,c, 2005, 2006; Akagi and Komiya, 1995; Bergado et al., 2007; Burghignoli et al., 2000; Campanella, 1965; Campanella and Mitchell, 1968; Cekerevac and Laloui, 2004; Cui et al., 2009; De Bruyn and Thimus, 1996; Delage et al., 2000; Demars and Charles, 1982; Duncan and Campanella, 1965; Eriksson, 1989; Houston et al., 1985; Hueckel and Baldi, 1990; Hueckel et al., 2009; Hueckel and Pellegrini, 1992; Jefferson and Rogers, 1998; Kuntiwattanakul et al., 1995; Laguros, 1969; Laloui, 2001; Mitchell, 1969; Moritz, 1995; Murayama, 1969; Noble and Demirel, 1969; Paaswell, 1967; Plum and Esrig, 1969; Sherif and Burrous, 1969; Sultan et al., 2002; Tanaka et al., 1997; Tang et al., 2008; Tidfors and Sälfors, 1989; Uchaipichat and Khalili, 2009; Vega et al., 2012; Villar, 2004). Understanding the evolution of the thermo-hydro-mechanical behavior of soils over the full temperature domain, i.e., from freezing to elevated temperatures, is currently absent.

Treating the temperature as a continuous domain over which the soil behavior evolves is critical to, for example, incorporate temperature variations in stability analysis of natural and man-made slopes and bluffs (Gariano and Guzzetti, 2016; Mickelson et al., 2004; Paranunzio et al., 2016; Ravanel and Deline, 2011, 2015; Shibasaki et al., 2016; Stoffel and Beniston, 2006). Furthermore, the effect of full thermal cycles on the top landfill clay liners is critical to ensure that the liners maintain a low permeability throughout their life time (Rowe, 2008; Rowe, 2012; Rowe et al., 2010). Finally, understanding the thermo-hydro-mechanical behavior of earth surface materials subjected to freeze-heat temperature variations will pave the road to explaining the behavior of planetary soils subjected to freeze-heat diurnal temperature variations (Molaro et al., 2015; Molaro et al., 2017).

Despite the extensive studies focused on the macroscale physical

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and mechanical properties of soils subjected to either freezing or elevated temperatures, the evolution of these macroscale properties over the entire temperature range is unclear. While different temperature changes apply during freezing-thawing and heating-cooling, comparable macroscale responses of cohesive soils were reported after heating-cooling cycles and after freezing-thawing cycles. For instance, the yield stress of normally consolidated raw clays increases after heating the raw clay and cooling it back to its original temperature (Plum and Esrig, 1969); a comparable increase in the yield stress occurs when raw clays are subjected to freezing-thawing cycles (Chamberlain, 1981). Furthermore, the permeability of cohesive soils increases at elevated temperatures (Cho et al., 1999; Mitchell, 1969) as well as after freezing-thawing cycles (Benoit and Voorhees, 1990; Chamberlain and Gow, 1979; Konrad, 1989; Othman and Benson, 1993; Qi et al., 2008; Viklander, 1998). Moreover, heating-cooling cycles cause contractive thermally-induced volumetric strains of normally-consolidated raw clays (Baldi et al., 1988; Hueckel and Baldi, 1990; Tanaka, 1995), which is the same behavior observed in normally-consolidated raw clays after freezing-thawing cycles (Othman and Benson, 1993). While these observed responses were explained in the literature using different micro-scale phenomena that occur at freezing and elevated temperatures, the impact of the soil microstructural changes occurring at one side of the temperature domain on the soil response at the other end of the temperature domain is not understood making the interpretation of the evolution of the macroscale behavior over the full temperature range challenging.

Understanding the evolution of the soil microstructure over the full temperature range will facilitate (1) justifying the observed macro-scale thermo-mechanical behavior, and (2) developing robust thermo-mechanical constitutive relations that mimic the soil response over the full temperature range. Such understanding will provide the basic knowledge needed to, for example, learn how the increase in pore sizes and cracks that occur upon freezing (Chamberlain and Gow, 1979; Konrad, 1989; Othman and Benson, 1993) will impact the hydraulic conductivity, compressibility, and strength at elevated temperatures. Also, it will help determining whether or not the thermal-softening due to heating an already frozen-thawed raw clay (De Bruyn and Thimus, 1996; Hueckel et al., 2009; Kuntiwattanakul et al., 1995; Noble and Demirel, 1969; Sherif and Burrous, 1969; Uchaipichat and Khalili, 2009) will cause additional reductions in the already reduced shear strength after freeze-thaw (Broms and Yao, 1964; Czurda and Hohmann, 1997; Garham and Au, 1985; Ogata et al., 1985).

Therefore, the overall goal of this study is to determine the microstructural evolution of raw clays over the full temperature range from freezing to elevated temperatures. In general, three parameters are considered to explain the effect of temperature on clay microstructure; these parameters are the clay particle orientation, pore size distribution, and pore connectivity. This paper focuses on the characterizing experiments performed to understand the temperature effects on the former parameter, i.e., clay particle orientation.

2. Background

X-ray diffraction (XRD) was identified as a viable technique to study the change in clay mineral particle orientations due to one-dimensional loading (Davis-Smith, 2004), sample preparation technique (Gibbs, 1965; Krizek et al., 1975; Sachan and Penumadu, 2007), and freeze-thaw cycles (Norrish and Rausell-Colom, 1962). These studies relied on the fact that the platy nature of clay mineral particles impacts the intensities of the X-ray reflected measurements (Brindley and Kurtossy, 1961, 1962; Lim et al., 2016; Martin, 1966). This section provides a brief background on XRD, and the approach followed to infer about clay mineral particles orientation.

2.1. Basics of X-ray diffraction (XRD)

X-ray diffraction (XRD) technique relies on exposing the material of interest to x-ray radiation and detecting the reflected radiations. The reflected radiations occur in all directions; of interest is the in-phase radiations that strengthen one another producing a strong reflection in the detected X-ray pattern. According to Bragg's law ($n\lambda = 2d\sin\theta$), in-phase radiations occur when X-rays with (λ) wave length radiating the material at an angle (θ) are reflected on two parallel atomic planes at a spacing (d) without a phase shift between the reflected radiations. When $n = 1$, the reflection is known to be of first order. Since each mineral has unique atomic arrangements (i.e., lattice structure) and therefore unique X-ray reflection pattern, XRD is typically used for mineral identification (Grim, 1968; Mitchell and Soga, 2005). The intensity of the detected X-ray reflections from any given plane of a given mineral is proportionally correlated to the amount of the mineral in the soil and the relative orientation of the planes with respect to the radiating X-rays (Mitchell and Soga, 2005).

The atomic planes are typically classified into basal planes and prism planes. Basal planes are those perpendicular to the major principal axis of the crystal, while all other planes (i.e., not perpendicular to the major principal axis) are known as prism planes. In general, the major principal axis for clay minerals is the axis perpendicular to the silica tetrahedral sheets and the octahedral sheets as shown for kaolinite in Fig. 1. Therefore, the basal plane (001) is the plane that contains oxygen atoms as shown in Fig. 1.

2.2. XRD for particle orientations

As mentioned earlier, the intensity of the diffracted rays depends on the relative orientation of the planes with respect to the radiating X-rays. This dependency of the intensities on plane orientations forms the main basis for the use XRD to determine the particle orientations. For simplification, well-oriented basal planes as shown in Fig. 2.a result in no phase shift of the diffracted ray leading to the maximum possible intensity. Any rotation of these basal planes, in this study due to temperature variation, will cause a phase shift that either delays or advances the diffracted rays with respect to those diffracted on lower

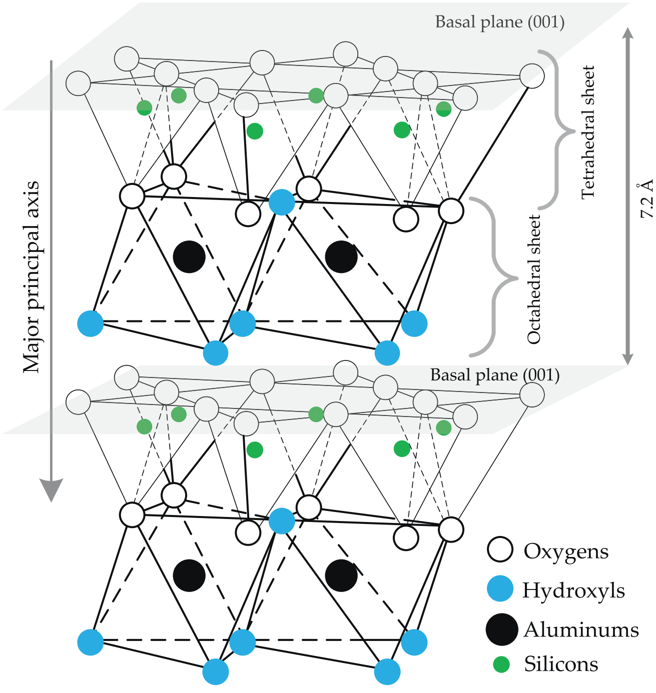

Fig. 1. Kaolinite lattice structure (after Grim, 1968).