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Membrane behavior of bentonite-amended compacted clay

Qiang Tang[a,b], Takeshi Katsumi[a,*], Toru Inui[a], Zhenze Li[c]

[a]*Graduate School of Global Environmental Studies, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan*
[b]*School of Urban Rail Transportation, Soochow University, Suzhou 215131, China*
[c]*Department of Civil Engineering, University of Ottawa, 161 Louis Pasteur, Ottawa, ON, Canada K1N 6N5*

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Abstract

The performance of landfill liners can be enhanced if they exhibit a semipermeable membrane behavior, which restricts the migration of contaminants. Consequently, enhancing the membrane property of clays used for liners is becoming increasingly important. As bentonite has already proven to be an excellent additive for improving the membrane behavior of clay, the hydraulic conductivities and membrane behaviors of a locally available clay, known as Fukakusa clay, amended with different amounts of dry bentonite (5%, 10%, 15%, and 20%) were herein evaluated. The chemico-osmotic efficiency coefficient, ω, was obtained under different concentration differences of KCl solution (0.5, 1, 5, 10, and 50 mM) for specimens in rigid-wall cells. The test results show that the ω of unamended clay is very low and can be ignored unless amended with bentonite, and that hydraulic conductivity k is suitably low (1.58×10^{-9} m/s). Additionally, ω decreased as the KCl concentration increased, which is consistent with the theory that increasing concentration causes progressively greater shrinkage of the diffuse-double layers of the clay particles. Furthermore, the mechanisms that influence the membrane behavior are discussed from the viewpoints of the diffuse-double layer and the interparticle pore size with the assistance of SEM.

Keywords: Bentonite; Fukakusa clay; Hydraulic conductivity; KCl; Membrane behavior; Chemico-osmotic pressure

1. Introduction

Compacted clay liners are a common component of bottom liner systems in waste containment facilities due to their affordability and excellent barrier performance against the migration of aqueous contaminants (e.g., Boynton and Daniel, 1985; Shelley and Daniel, 1993; Chapuis, 2002; Cui et al., 2011; Wang et al., 2013). However, the performance of these clay liners may be greatly enhanced if they exhibit a semipermeable membrane behavior, by which the liners can prevent or restrict the migration of selected substances (Tuwiner, 1962; Mulder, 1991). In the case of a chemical solution, a semipermeable clay liner can inhibit the migration of solute molecules (Mitchell, 1993), while allowing water to flow from a lower solute concentration (higher water activity) to a higher solute concentration (lower water activity) until equilibrium in the concentration on both sides of the clay has been established (Olsen, 1969, 1972; Greenberg et al., 1973; Manassero and Dominijanni, 2003; Shackelford and Lee, 2003; Henning et al., 2006).

Membrane behavior in soil is mainly attributed to two causes, namely, size restriction and electrical repulsion from a diffuse-double layer (DDL) (Van Impe, 2002). Size restriction,

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[*]Corresponding author.
E-mail address: katsumi.takeshi.6v@kyoto-u.ac.jp (T. Katsumi).
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which usually occurs in biological fields, is the process by which very large non-charged molecules, such as neutral organic compounds, are blocked when the pore size is sufficiently small (Grathwohl, 1998). A DDL is composed of two components: a negative charge on the surface of the clay particles and a distribution of counterions adjacent to the particle surface to counterbalance the surface charge (Dominijanni and Manassero, 2012b). When the interparticle distance is on the same order as the pore size under an external load, the DDLs overlap (Fritz and Marine, 1983; Fritz, 1986; Mitchell, 1993). In this case, anions cannot migrate through the pores due to the predominantly negative electrical potentials of the clay particle surface. To maintain electrical neutrality in the solution, cations tend to remain with their co-ions, which restricts their movement (Hanshaw and Coplen, 1973; Marine and Fritz, 1981; Fritz and Marine, 1983; Fritz, 1986; Keijzer et al., 1997; Shackelford, 2011).

When a membrane can completely restrict the migration of solute, the membrane is ideal. In this case, only the solvent can cross an ideal semipermeable membrane regardless of the solute concentration gradient. In contrast, if both the solvent and the solute can cross the membrane freely, then the membrane property does not exist. Actually, the membrane behavior of natural materials falls in between these two extremes; a function of the solute can pass through the membrane and change the concentration gradient. Over a sufficiently long period of time, diffusion of the solute will gradually equalize the concentrations on both sides, decreasing the membrane behavior until it completely disappears (Malusis et al., 2003; Manassero and Dominijanni, 2003; Malusis and Shackelford, 2004).

Typically, the extent to which a soil acts as a membrane is quantified in terms of a reflection efficiency coefficient, σ, (Staverman, 1952; Katchalsky and Curran, 1965; Kemper and Rollins, 1966; Spiegler and Kedem, 1966; Olsen et al., 1990) or a chemico-osmotic efficiency coefficient, ω (Mitchell, 1993; Malusis et al., 2003; Malusis and Shackelford, 2004; Yeo et al., 2005; Evans et al., 2008; Kang and Shackelford, 2010). As the symbol σ is usually used to designate stress in engineering fields, ω is more commonly used to designate the degree of membrane behavior. The value of ω ranges from 0, representing no solute restrictions, to 1, representing an “ideal” or “perfect” membrane that completely restricts the movement of the solutes (i.e., $0 \leq \omega \leq 1$). In most cases involving the membrane behavior of clay, only a fraction of the pores are restrictive. Consequently, clay materials are usually referred to as “non-ideal” or “leaky” membranes (Kemper and Rollins, 1966; Olsen, 1969; Barbour and Fredlund, 1989; Mitchell, 1993; Keijzer et al., 1997).

Membrane behavior has been observed in several types of soils used as liners or barriers (e.g., Malusis and Shackelford, 2004; Yeo et al., 2005; Evans et al., 2008; Kang and Shackelford, 2010). However, the observed membrane behavior has been too low to sufficiently prevent the migration of contaminants. Thus, in combining the advantages of clay and bentonite, bentonite is introduced to promote the barrier performance of clay towards contaminants and enhance its membrane properties (Malusis and Shackelford, 2002b; Yeo et al., 2005; Evans et al., 2008; Kang and Shackelford, 2010). According to previous studies, however, the bentonite content of the mixtures for membrane tests significantly varied, as shown in Table 1, while very few studies have evaluated the effect of the bentonite content on the previous membrane behavior, which was also addressed by Shackelford (2012). Based on the above considerations, this study utilizes bentonite as an additive to amend locally available clay for the purpose of enhancing the membrane behavior of clays. The mechanisms for the observed membrane behavior will also be analyzed and discussed.

Table 1
Bentonite proportion in clay–bentonite mixture for membrane tests in references.

Clay	Bentonite	Bentonite proportion (by dry weight)	References
Nelson farm clay	Na-bentonite	0, 5%	Kang and Shackelford (2010)
Clay–sand mixture	Na-bentonite	5%	Yeo et al. (2005)
Kaolin	Na-bentonite	20%	Van Impe (2002)
–	GCL bentonite	100%	Malusis and Shackelford (2002b)

2. Materials and experimental method

2.1. Materials

Two types of soils were evaluated: locally available natural clay and powdered sodium bentonite. The local natural clay is known as Fukakusa clay (FC); it is obtained from Kyoto, Japan. The powered sodium bentonite, referred to as “super clay”, is originally from Wyoming, USA (purchased from Hojun Co. Ltd.). Sodium bentonite is widely used as a soil mixture additive in slurry walls. Table 2 presents the physical and chemical properties of Fukakusa clay and sodium bentonite. For Fukakusa clay, the main exchangeable metal was identified as Ca^{2+} (10.5 meq/100 g), and small amount of Mg^{2+}, Na^{+} and K^{+} (3.8, 0.6 and 2.2 meq/100 g). For the bentonite, the dominant exchangeable metal was identified as Na^{+} (43.7 meq/100 g), then Ca^{2+} (22.1 meq/100 g), Mg^{2+} (13.8 meq/100 g), and only small amount of K^{+} was detected. The sum of exchangeable metals is a little higher than the cation exchange capacity (CEC), which can be ascribed to the presence of carbonate in the soils. Part of the measured cations, therefore, might have originated from the carbonates and not from the exchange complex of the soil particles (Heister, 2005).

Fig. 1 shows the XRD patterns of both Fukakusa clay and bentonite (RAD-2B, Rigaku Corporation, Japan). In the case of bentonite, it was abundant in Montmorillonite as can be observed by the characteristic peaks that appeared at $2\theta = 7.02°$, 19.78°, 26.60°, 29.84°, 34.86°, and 36.14°. In the case of the Fukakusa clay, and based on the characteristic peaks appearing at $2\theta =$ 20.94°, 26.62°, 36.62°, 39.54°, 40.32°, 42.56°, and 45.86°, it is