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## Research paper

# Determining the thickness of adsorbed water layers on the external surfaces of clay minerals based on the engineering properties of soils

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#### A R T I C L E I N F O

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## ABSTRACT

This article discusses the basic findings that enable a determination of the thicknesses of the adsorbed water layers on the external surfaces of clay minerals and their dependence on the total water content in soils. The thicknesses of the water films were first determined indirectly from tests of the undrained shear strength of clay mineral samples. A comparison between these thicknesses and the total quantity of water in the samples showed that this ratio is exactly defined and can be expressed by a non-linear function in which the type of soil is determined by two parameters, which depend on the size and the quantity of the clay minerals. A test of the correctness and usefulness of the established expression was made on two samples of clays. The thicknesses of the water films, calculated using the proposed equation for different moisture contents, were compared with the thicknesses determined from the adsorptive potential of the soils. These results showed a good agreement. The possible application of the proposed expression is shown for the case of an estimation of the soil–water characteristic curve in the range between the water content at the air-entry value and the residual water content. © 2016 Elsevier B.V. All rights reserved.

### 1. Introduction

The engineering properties of fine-grained soils depend on different factors resulting from the mineral composition and the environment. The types, quantity and physico-chemical properties of minerals determine the basic soil characteristics, whereas their final engineering properties depend on the quantity of the available water. Moreover, they are influenced by the soil density, the structure, the organic matter and the chemical composition of the pore water.

Fine-grained soils include both clay and non-clay minerals. They differ in terms of their grain size and shape, as well as in their surface properties. Firstly, non-clay minerals belong to the group of rock-forming minerals, such as quartz, feldspars, amphiboles, pyroxene, mica, olivine, calcite and dolomite. Their grain shape is mostly round-to-spherical and larger than 2 µm. In addition, they are relatively inert, and their engineering properties depend mostly on the shape, the surface texture, and the size distribution (Mitchell, 1993).

Clay minerals consist predominantly of flat grains that are smaller than 2  $\mu$ m. Their external specific surface area  $A_{Se}$  (external area per unit mass of solid material) is between 10 and 100 m<sup>2</sup>/g; and their internal specific surface area  $A_{Si}$ , which appears in swelling clays, is approximately 800 m<sup>2</sup>/g (Mitchell, 1993). On the flat layer surfaces of clay minerals a negative charge is produced due to the isomorphous substitution of cations in tetrahedral silica sheets and the octahedral alumina sheets. On the edges, where the primary bonds are broken, a positive

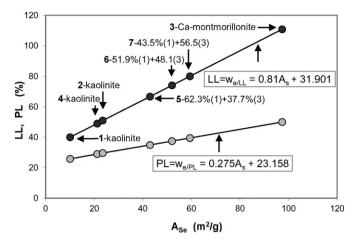
\* Corresponding author. *E-mail address:* bojana.dolinar@um.si (B. Dolinar). charge can appear that depends basically on the pH value of the pore water. The charges on such surfaces are compensated with cations, anions and water molecules. Besides the adsorbed water on the internal and external surfaces of

Besides the adsorbed water on the internal and external surfaces of clay minerals, free pore water is present in soils (except for matrix potentials < -10 MPa, Tuller and Or, 2005). Adsorbed water has a different structure and properties than pure water (Low, 1979; Sposito, 1984; Sun et al., 1986 and others). It is tightly bound, so it does not influence the strength, deformability or hydraulic permeability of soils, which was proved by the research of Nagaraj et al. (1991), Dolinar and Trauner (2005) and Dolinar (2005). Using the standard method for measuring water content, the total quantity of free pore and adsorbed water can always be determined by drying at a temperature higher than 100 °C. This is why it is not possible to define generally valid expressions for different fine-grained soils, which would then enable a definition of the relationships between the individual engineering properties and the water content.

The adsorbed water content in the soil can be determined if the specific surface area and the thickness of the adsorbed water film are known. It has been established that the thickness of this film on the external surfaces of the clay grains depends on the total amount of water in the soil, and with an increase of the latter the film thickness increases; however, this relationship has not been investigated until now. In this article a procedure for determining the thicknesses of adsorbed water layers on the external surfaces of clay minerals is described as well as the interdependences between these thicknesses and the total quantities of water in the soils. The usefulness of these findings was tested on two soil samples for which the soil–water characteristic curves







**Fig. 1.** Quantity of pore water plus the external surface water at the liquid limit *LL* (%) and the plastic limit *PL* (%) as a function of the specific surface area  $A_{Se}$  (m<sup>2</sup>/g).

(SWCCs) in the range between the water content at the air-entry value and the residual water content were estimated.

#### 2. Preliminary findings

The content of adsorbed interlayer water  $w_{ai}$  in the swelling clay minerals depends on the internal specific surface area  $A_{Si}$ , but mostly on the type of interlayer cations (Grim, 1962; Bergaya et al., 2006 and others), whereas the quantity of adsorbed water on the external surfaces of the clay grains  $w_{ae}$  (g/g) depends on the external specific surface area  $A_{Se}$  (m<sup>2</sup>/g), the thickness of the adsorbed water layer *h* (m) and the density of the water  $\rho_w$  (~10<sup>6</sup> g/m<sup>3</sup>) (Mitchell, 1993) – Eq. (1).

$$w_{ae} = A_{Se} \cdot h \cdot \rho_w \tag{1}$$

The thickness of the adsorbed water film changes, depending on the quantity of water w in the soils. It was established that this thickness for a water content at the liquid limit *LL* of the soils is the same for different clay minerals and also that different fine-grained soils have the same matrix potential  $\Psi_m$  for this water content (Russell and Mickle, 1970; Mitchell, 1993). The reason for this lies in the same surface structures of clay minerals and, consequently, also in the forces of the interaction between these surfaces and the water molecules.

The listed findings are consistent with the conclusions published by Tuller et al. (1999) and Baker and Frydman (2009), and refer to the adsorptive  $\Psi_{ad}$  and the capillary potential  $\Psi_{cp}$  as components of the matrix potential  $\Psi_m$  of soils.

Based on previous studies (Derjaguin et al., 1987; Iwamatsu and Horii, 1996) Tuller et al. (1999) defined the relationship between the adsorptive components  $\Psi_{ad}$  (Pa) of the matrix potential and the thickness of the adsorbed water film *h* (m) using Eq. (2).

$$\Psi_{ad} = A/6 \cdot \pi \cdot h^3 \tag{2}$$

They assumed that the thickness of the adsorbed water layer depends mostly on the strength of the van der Waals interactions between the clay minerals and the water. This material characteristic in Eq. (2) is expressed with the Hamaker constant *A* (J). The typical values of *A* for a soil–water system are in the range  $A = -10^{-20}$  to  $-10^{-19}$  J. Or and Tuller (1999) recommended adopting a value of  $A = -6 \cdot 10^{-20}$  J. Baker and Frydman (2009) assumed that for fine-grained soils with double porosity, the adsorptive potential  $\Psi_{ad}$  and the capillary potential  $\Psi_{cp}$  are the same, because at equilibrium, the water in the pods (adsorbed water) and macropores (free pore water) must be in the same energy state (Eq. (3)).

$$\Psi_m = \Psi_{ad} = \Psi_{cp} \tag{3}$$

Dolinar (2004) and Dolinar and Trauner (2004) found that the total water content at the liquid limit  $w_{LL} = LL$  of different soils can be divided into free pore and adsorbed water as described by Eq. (4) for non-swelling soils and Eq. (5) for swelling soils. The free pore water content  $w_{p|LL}$  and the thickness  $h_{LL}$  of the adsorbed water layer at the liquid limit of the soils were determined experimentally using clay minerals, their mixtures, and composites containing non-clay minerals.

$$LL = w_{LL} = p \cdot w_{P|LL} + w_{ae|LL} = p \cdot w_{P|LL} + h_{LL} \cdot A_{Se} \cdot \rho_w \tag{4}$$

$$LL = w_{LL} = p \cdot w_{P|LL} + w_{ae|LL} + w_{ai|LL} = p \cdot w_{P|LL} + h_{LL} \cdot A_{Se} \cdot \rho_w + w_{ai|LL} (5)$$

In the equations  $w_{p|LL} = 0.319 \text{ g/g}$  is the quantity of free pore water at the liquid limit, which depends on the quantity of clay minerals p in the soil (p is the percentage of clay minerals in the soil divided by 100;  $0 ), <math>h_{LL} = 8.1 \cdot 10^{-9}$  m is the thickness of the adsorbed water layer on the external surfaces of clay minerals,  $A_{Se}(m^2/g)$  is the external specific surface area,  $w_{ae|LL} = h_{LL} \cdot A_{Se} \cdot \rho_w$  (g/g) is the quantity of strongly adsorbed water on the external surfaces of the soil,  $w_{ai|LL}$ (g/g) is the quantity of adsorbed water on the soil's internal surfaces, and  $\rho_w$  (g/m<sup>3</sup>) is the density of the water.

The findings (Eqs. (4) and (5)) agree with previous ones and confirm that the film thickness of the adsorbed water on the external surfaces of clay minerals is equal at the liquid limit of the soils (taking into account the accuracy of the tests of the liquid limit); the content of the adsorbed water on the external surfaces of clay mineral particles depends on the external specific surface area; the free pore water content as well as total water content depends on the portion of clay minerals in the soil's composition. The latter was already established in 1964 (Seed et al., 1964) with tests using mixtures of kaolinite and sand.

Dolinar et al. (2007) also found that Eqs. (4) and (5) are valid at the plastic limit (PL) of soils where the quantity of free pore water is  $w_{p|PL} =$  0.231 g/g and the thickness of the adsorbed water layer on the external

#### Table 1

	specific surface				

Sample	KGa-1b	KGa-2	SAz-1		
Mineral composition	96% kaolinite, 3% anataz, 1% cradallit	96% kaolinite, 3% anataz, 1% cradallit	98% Ca-montmorillonite, 1% quartz, <1% plagioclase, microcline		
External specific surface area $A_{Se}$ (m <sup>2</sup> /g) Calculated internal specific surface area $A_{Si}$ (m <sup>2</sup> /g) Water content (%) at:	$10.05\pm0.02$	$23.50\pm0.02$	97.42 ± 0.58 724.22		
$s_u = 2.66 \text{ kPa} - \text{liquid limit}$	40.0	50.9	110.8 (without $w_{ai}$ )		
$s_{u} = 10.6 \text{ kPa}$	35.0	43.3	87.2 (without $w_{ai}$ )		
$s_u = 42.5 \text{ kPa}$	30.8	36.8	$68.7$ (without $w_{ai}$ )		
$s_u = 266 \text{ kPa} - \text{plastic limit}$	25.8	29.8	50.0 (without $w_{ai}$ )		

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