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CORRELATION BETWEEN SURFACE AREA AND ATTERBERG LIMITS OF FINE-GRAINED SOILS

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Abstract—As the water content is increased, the consistency of a fine-grained soil changes from a semi-solid state to a plastic state and finally to a liquid state. The plastic limit (PL) is the point at which the consistency, caused by the soil water content, is transformed from a semi-solid state to a plastic state. The liquid limit (LL) is the point at which the consistency is transformed from a plastic state to a liquid state. The plastic limit and liquid limit are often collectively referred to as the Atterberg (or consistency) Limits. Although the liquid and plastic limits are easily determined, fundamental interpretations of the limits and quantitative relationships between their values and compositional factors are more complex. Previous studies examined artificially-prepared soil samples that contained monomineralic clays and a non-clay substance (quartz sand). These studies have shown that in soils without expandable clays the PL and LL water contents were mostly related to surface area and clay content. For soils that contain expandable clays, the PL and LL values are also dependent on interlayer water content. Hence, expandable clay mineral contents are needed to calculate PL and LL values. These relationships have been presented in a general analytical form. The aim of these investigations was to identify practical applications. Mineral compositions and surface areas of five randomly selected natural soil samples were used to estimate PL and LL values. The estimated values were compared to experimentally measured liquid limits (by the 'fall-cone' test) and plastic limit (by the 'rolling thread' test) values. The measured PL values ranged from 18.77 to 44.92% and the LL values from 31.19 to 82.10%. The differences between estimated and measured Atterberg Limits were 3.0−7.1% for the PL and 2.7−7.8% for the LL. Minor differences in measured and estimated Atterberg Limits were probably due to soil organic matter (1.2−2.7%).

Key Words—Atterberg Limits, Clays, Liquid Limit, Plastic Limit, Specific Surface of Soils.

INTRODUCTION

As the water content is increased, the consistency of a fine-grained soil changes from a semi-solid state to a plastic state and eventually to a liquid state. The plastic limit is the boundary between semi-solid and plastic consistency and the liquid limit is the boundary between plastic and liquid consistency. The plastic limit and liquid limit are often collectively referred to as the 'Atterberg Limits'. They are of key importance in soil mechanics because they determine, in a simple way, the interaction between solid and liquid phases in soils, and thus provide the possibility of classifying soils into groups with similar mechanical properties. The results of these investigations, in most cases, provide a good basis for predicting other soil properties, such as deformability, expansion, hydraulic conductivity and strength.

It has long been known that Atterberg Limit values depend primarily on soil mineralogical properties. Several researchers (White, 1949; Grim, 1962; Seed *et al.*, 1964; Farrar and Coleman, 1967; Sudhaker *et al.*, 1985; Sridharan *et al.*, 1988; Muhunthan, 1991) compared soil plastic limit and liquid limit values to grain size, specific surface, clay content, and cation exchange capacity to identify relationships between mineralogy and Atterberg Limit values. The results of these research studies, however, varied considerably, were valid only for the investigated soils, and were not generally applicable.

Dolinar and Trauner (2004, 2005) explained why previous research studies to relate mineralogy to Atterberg Limits reached different conclusions. Cohesive soils contain both clay and non-clay minerals and interactions between clay minerals and water affect the water-holding capacity of soil. Water is strongly adsorbed to the external surfaces (w_e) of non-expanding clays, whereas water adsorbs to both the external (w_e) and internal surfaces (w_i) of expanding clays. Saturated clays contain free pore-water (w_{fp}) in addition to strongly adsorbed water. The total quantity of intergrain water (w_{tig}) is equal to the free pore-water plus the external surface water ($w_{tig} = w_{fp} + w_e$). A standard method for measuring water content (w_{od}) is from the weight loss after drying in an oven at 100 to 110ºC. The total water measured by oven drying (w_{od}) equals the total intergrain water plus the internal surface water ($w_{od} = w_{tig} + w_i = w_{fp} + w_e + w_i$). The amount of intergrain water, (w_{tig}), at the liquid and plastic limits is mostly dependent on soil texture and clay mineralogy, whereas the interlayer water, w_i, is mostly dependent on the interlayer cations (Grim, 1962). This explains why

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no unified criterion can relate Atterberg Limit values to soil mineralogical properties for soils with or without expandable clays.

Dolinar and Trauner (2004, 2005) found that the total intergrain water (w_{tig}) at the liquid and plastic limits depends on soil texture and clay content. The following relationships between clay mineralogy and Atterberg Limits were derived from published research:

(1) The hydraulic conductivities of different clays are about equal at the liquid limit (Nagaraj *et al.*, 1991). This means that the pore sizes that effectively control fluid flow at the liquid limit must be about the same size for all clays (Mitchell, 1993) and, hence, the quantity of free pore-water at the liquid limit is a constant.

(2) The water content is directly proportional to soil surface area and matrix suction (Tuller, 2005). Soils have similar pore-water suction values at the liquid limit (Russel and Mickle, 1970). This means that the ratio of adsorbed water to clay surface area should be about the same at the liquid limit.

(3) Seed *et al.* (1964) investigated clay mineral/sand mixtures and discovered a linear relationship between the liquid limit value and the clay content. This suggests that most of the water in soils at the liquid limit is associated with clay.

(4) At the liquid limit, different fine-grained soils have approximately equal undrained shear strength (Casagrande, 1932).

Considering these relationships, the total quantity of intergrain water in soils at the liquid limit $w_{tig|LL}$ can be expressed by equation 1

$$w_{tig|LL} = p(w_{fp|LL} + w_{e|LL}) = p\ (w_{fp|LL} + t_{a|LL} \times A_{SC}) \quad (1)$$

where $w_{fp|LL}$ is the quantity of free pore-water at the liquid limit. According to equation 1, the quantity of free pore-water ($w_{fp|LL}$) of different clays at the liquid limit is constant. The quantity of firmly adsorbed water on soil external surfaces is $w_{e|LL}$. This quantity of water depends on the sizes of the external surfaces of the clay grains A_{SC} (m^2/g) because the thickness $t_{a|LL}$ of the firmly adsorbed water on the external clay surface is constant for all clays at the liquid limit (according to equation 2). The intergrain water content depends on the quantity of clay minerals, p, in the soil (p is the percentage of clay minerals in soil divided by 100; $0 < p = 1$) assuming that all water in the soil is associated with the clay minerals (according to equation 3). If the expression 1 is valid for undrained shear strength at the liquid limit of soils (according to relationship 4), it is to be expected that it is valid at the other values of undrained shear strength also (Koumoto and Houlsby, 2001). The quantity of intergrain water at the plastic limit of soils can, therefore, be expressed by equation 2.

$$w_{tig|PL} = p(w_{fp|PL} + w_{e|PL}) = p(w_{fp|PL} + t_{a|PL} \times A_{SC}) \quad (2)$$

On the basis of experimental tests, Dolinar and Trauner (2004) reported that the quantity of intergrain water $w_{tig|LL}$ (%) at the liquid limit is linearly dependent on the specific surface and the quantity of clay minerals in soils (equation 3).

$$w_{tig|LL} = \alpha + \beta_{LL} \times A_{SC} \quad (3)$$

The parameters α_{LL} = 31.90 and β_{LL} = 0.81 were determined from values of A_{SC} and $w_{tig|LL}$ by approximation (least squares method). Equations 1 and 3 have the same structure; therefore, the first and the second terms in both have to be equal. Consequently the quantity of free pore-water at the liquid limit, $w_{fp|LL} = \alpha_{LL}$, the quantity of the firmly adsorbed water around the clay grains, $w_{e|LL} = t_{a|LL}A_{SC} = \beta_{LL}A_{SC}$, and the average thickness of the firmly adsorbed water around the clay grains $t_{a|LL} = \beta_{LL}$. Equation 3 is valid for soils which contain only clay minerals. If the non-clay minerals are present, the value of parameter α_{LL} decreases in proportion to the weight portion of clay minerals in soils, but the slope of the function remains the same. In that case equation 3 is expressed as equation 4,

$$w_{tig|LL} = p\ \alpha_{LL} + \beta_{LL} \times A_S \quad (4)$$

where p is the portion of clay minerals in soil and $A_S = pA_{SC}$ (m^2/g) is the specific surface of soil.

The quantity of $w_{tig|PL}$ (%) can be calculated by equation 5

$$w_{tig|PL} = p\ \alpha_{PL} + \beta_{PL} \times A_S \quad (5)$$

where α_{PL}= 23.16 and β_{PL}= 0.27. The coefficients in equations 4 and 5 were determined by testing artificial monomineral clay mixtures that had clearly defined chemical and mineral compositions as well as specific surface. It should be noted that in practice it is difficult to determine an accurate quantitative mineral composition. The values of Atterberg limits can also be influenced by organic substances which are often found in soils. Their influence was not investigated in the present work. The aim of the investigations described in this article is to verify practical applications for the above-mentioned findings. Five samples of cohesive soils were tested for this purpose, in which the liquid and plastic limits were determined both experimentally and on the basis of mineralogical properties (specific surface, type and portion of clay minerals in soil composition). This allowed a comparison between the measured and calculated values.

MATERIALS AND METHODS

Mineral composition

The bulk mineralogy and clay mineralogy of the soil samples were determined by the Geological Survey of Slovenia using X-ray diffraction (XRD) techniques. Samples were scanned using a Philips PW 3710 X-ray diffractometer with an 1820 goniometer, an automatic divergence slit, and a curved-crystal graphite monochromator. The instrument was operated at 40 kV and

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Table 1. Mineralogical composition (wt.%) of whole-soil samples.

Sample	1	2	3	4	5
Muscovite/illite	25	35	28	35	34
Chlorite	8	14	16	0	18
Kaolinite	5	0	0	12	0
Ca-montmorillonite	14	0	0	34	0
Quartz	34	25	42	19	43
Plagioclase	9	3	9	0	3
Microcline	5	0	4	0	3
Calcite	0	23	0	0	0

30 mA using CuKα radiation. The bulk mineralogy was determined on whole-soil powder mounts. The clay fraction was separated by centrifugation. Oriented mounts were prepared by suspending soil clay samples in deionized water and air drying the suspensions on quartz plates. The clay suspensions were later analyzed after vapor solvation with ethylene glycol for 12 h at 60ºC. Semi-quantitative mineralogical compositions of the bulk soil samples (Table 1) and clay fractions (Table 2) were calculated using the methods of Shultz (1964) and Biscaye (1965).

Chemical composition

The results of the chemical analyses were used to confirm the quantities of individual minerals in the soils. The chemical compositions of the soils (Table 3) were determined by Acma Analytical Laboratories Ltd. of Vancouver. The inductively-coupled plasma emission spectrometry method was used to determine the main elements quantitatively and qualitatively, whilst inductively-coupled plasma mass spectrometry was used to determine trace elements. Carbon and sulfur were determined using a Leco CS444 element analyzer.

Specific surface

External and internal surfaces can contribute to the specific surface area of mineral particles. Kaolinite and illite minerals are not expandable and only have external surfaces. Montmorillonite minerals are expandable and have both external and internal surfaces. Exchangeable cations and polar liquids can penetrate into the interlayers which have much larger surface areas than the external surfaces. The external specific surface areas of the investigated soils were measured using a five-point BET method with N_2 (Table 4).

Table 3. Chemical composition (%) of bulk-soil samples.

Sample	1	2	3	4	5
SiO_2	64.15	46.83	72.10	51.71	67.48
Al_2O_3	15.83	13.45	12.09	20.38	12.77
TiO_2	0.89	0.61	0.78	0.80	0.88
Fe_2O_3	5.09	5.02	4.92	9.06	6.70
FeO	1.1	1.0	1.0	0.3	0.5
MnO	0.03	0.07	0.05	0.18	0.18
MgO	1.97	2.27	1.07	1.66	1.62
CaO	0.87	12.42	0.41	1.21	0.40
Na_2O	0.97	0.15	1.19	0.16	0.28
K_2O	2.20	2.75	1.95	2.52	2.64
P_2O_5	0.13	0.12	0.15	0.13	0.07
Cr_2O_3	0.017	0.015	0.012	0.024	0.027
Total C	0.27	3.03	0.40	0.41	0.30
Total S	0.01	0.06	0.03	0.06	< 0.01
Organic matter	2.37	2.02	1.26	2.70	1.82

Particle-size analysis

The grain-size distribution was determined using a hydometer method (ISO/TS 17892-4 (2004)). In the method commonly used for engineering purposes, 20–40 g of clay soil are mixed with 1 L of water, agitated, and poured into a container. The density of the suspension is measured at various times by means of a hydrometer of special design. At any given time, the size of the largest particles remaining in suspension at the level of the hydrometer can be computed by means of Stoke's law, whereas the weight of the particles finer than that size can be computed from the density of the suspension at the same level. To disperse the soil, a deflocculating agent must be added to the water (sodium hexametaphosphate – 40 g/L). The grain-size distribution of tested soils is shown in Table 4.

Liquid and plastic limits

The liquid limits of samples were determined by the fall cone test (British Standards Institution, 1990). This test uses a standard cone with a 30º apex angle and a total mass of 80 g. A dish (55 mm diameter by 40 mm deep) is filled with soil in the consistency range of the liquid limit. The cone point is carefully brought into contact with the soil. The cone is then released so the

Table 2. The mineralogical composition (wt.%) of the <2 μm clay fraction with totals adjusted to equal % clay.

Sample	1	2	3	4	5
Illite	11	12	6	13	12
Chlorite	0	3	7	0	0
Kaolinite	4	3	0	5	7
Ca-montmorillonite	14	4	2	34	0
Mixed layers	10	19	14	19	25

Table 4. Particle-size distribution and specific surface area of soil samples.

Sample	% Clay	% Silt	% Sand	A_S (m^2/g)
1	39.1	58.1	2.8	30.1±0.4
2	40.7	56.9	2.4	28.5±0.4
3	29.2	60.6	10.2	16.7±0.1
4	70.7	27.2	2.1	54.1±0.3
5	44.2	53.9	1.9	32.6±0.2

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80 g mass produces some penetration for 5 s. The liquid limit is defined as the water content at which 20 mm of cone penetration occurs in the 5 s test time.

The plastic limits were determined by the rolling thread test (British Standards Institution, 1990). A sample of ~10 mm^3 was taken and rolled with the palm of the hand on a glass plate into a thread of ~3 mm diameter. The moisture content is adjusted until the 3 mm thread just begins to crumble. The moisture content should then be measured, as this is the plastic limit. The plasticity index (I_P) of a soil is the numerical difference between the liquid limit and the plastic limit. The I_P is usually proportional to the clay content and indicates the range of water contents over which a soil has plastic properties.

The values of Atterberg limits and I_P are given in Table 5.

The mineralogical analyses indicate that all of the soils contain montmorillonite in illite-montmorillonite (I-M), kaolinite-montmorillonite (K-M) and chlorite-montmorillonite (Ch-M) mixed-layered minerals or as Ca-montmorillonite. In this case, the portion of the interlayer water in an expanding mineral must be determined, so as to allow a comparison of the measured and calculated values of consistency limits with equations 4 and 5. The interlayer water quantity, w_i, can be calculated in accordance with equation 6 (Fink and Nakayama, 1972).

$$w_i = \frac{A_{Si}(d_2 - d_1)}{2\ 10^3} p_m(\%) \qquad (6)$$

The basal spacing in the c direction, which is d_1 = 0.96 nm for dry Ca-montmorillonite (dried at 105ºC), increases to $d_2 \approx$ 1.54 nm at a relative humidity of 80% and to d_2 = 1.9 nm in water (Brindley and Brown, 1980). In the case of a Ca exchangeable cation in montmorillonite, the adsorption of water between layers is then completed and the basal spacing is practically constant. In calculating the interlayer water quantity, w_i, with equation 6, consideration was given to the internal specific surface, A_{Si} = 626.80 m^2/g (the value adopted from the literature by Dolinar and Trauner, 2004) and adequate mass portions of montmorillonite, p_m, in the individual soils (Table 2). The assumed basal spacings (d_2) of montmorillonite at the plastic and liquid limit were 1.54 and 1.90 nm, respectively.

RESULTS AND DISCUSSION

Comparison of laboratory determined and estimated values of Atterberg limits

Measured and calculated Atterberg Limit values are compared in Table 5. The 'Cal.' symbol indicates values that were calculated from soil mineralogical properties using Equations 4–6. The 'Exp.' symbol indicates experimentally measured values.

It was evident that water quantities at the plastic and liquid limits that were calculated using specific surface area, clay mineral content, and interlayer water content were less than experimentally measured values. The differences in calculated and measured water contents were 3.0–7.1% at the plastic limit and 2.7–7.8% at the liquid limit. The lower values of the calculated consistency limits can result from the presence of organic matter in clays (1.3–2.7%). Errors can also occur because the calculation does not consider interlayer water in montmorillonite present in mixed-layer clays. An accurate quantitative analysis of the mineral composition is also debatable. In the case shown, the adopted portion of clay minerals (p) equalled the <2 μm grain quantity determined using the hydrometer method. Clay minerals with larger grains were not considered in the analysis, therefore the adopted value, p, was probably less that the real value.

Soil water contents measured at the Atterberg Limits can include water adsorbed to internal and external grain

Table 5. Soil clay fraction (p), %Ca-montmorillonite (p_m), interlayer water contents at liquid and plastic limits ($w_{i|LL}$,$w_{i|PL}$), intergrain water contents at liquid and plastic limits ($w_{tig|LL}$,$w_{tig|PL}$), liquid limit (LL), plastic limit (PL), and plasticity index (I_P). The abbreviations 'Cal.' and 'Exp.' indicate calculated and measured Atterberg Limit values.

Parameter	Sample									
	— 1 —		— 2 —		— 3 —		— 4 —		— 5 —	
	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.	Cal.	Exp.
p	0.39		0.41		0.29		0.71		0.44	
p_m (%)	14		4		2		34		0	
[1] $w_{i\|LL}$ (%)	4.12		1.18		0.59		10.02		0	
[2] $w_{i\|PL}$ (%)	2.54		0.72		0.36		6.18		0	
[3] $w_{tig\|LPL}$ (%)	36.8	43.1	36.2	42.9	22.8	30.6	66.5	72.7	40.4	43.1
[4] LL (%)	40.9	47.2	37.3	44.0	23.4	31.2	76.5	82.1	40.4	43.1
[5] $w_{tig\|PL}$ (%)	17.2	21.7	17.2	22.0	11.2	18.3	31.1	39.1	19.0	22.0
[6] PL (%)	19.7	24.3	17.9	22.7	11.6	18.7	37.3	44.9	19.0	22.0
[7] I_P (%)	21.2	22.9	19.4	21.3	11.8	12.5	39.2	37.2	21.4	21.1

[1,2] $w_i = A_{Si}(d_2 - d_1)p_m/2 \times 10^3$; [3] $w_{tig|LL} = 31.9p + 0.81A_S$; [4] LL = $w_{tig|LL} + w_i$;
[5] $w_{tig|PL} = 23.16p + 0.27A_S$; [6] PL = $w_{tig|PL} + w_i$; [7] I_p = LL − PL

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surfaces. The proposed method for calculating Atterberg Limits is simple for soils that do not contain expandable clays. In soils that contain swelling clays, the quantity of interlayer water must be determined. Interlayer water quantity is calculated from clay mineralogy, which is more difficult to determine than it is to directly measure Atterberg Limits. Therefore, this method of Atterberg Limit calculation is less applicable in practice.

It should also be noted that, in addition to mineralogy, other factors influence Atterberg Limits including temperature, organic matter, pore-water chemistry, and soil fabric. These were not investigated in the present study.

CONCLUSIONS

The first part of this paper discussed recent findings that show how soil composition influences water content at the plastic and liquid limits. It has been established that the quantity of intergrain water at both consistency limits depends on the soil clay content and mineralogy. These relationships were expressed empirically by test results obtained on artificially prepared samples composed of pure clay minerals and a non-clay substance (quartz sand).

The aim of the investigations described in this paper was to check for a practical application of established relationships between water retention, mineralogy and Atterberg Limits. We chose samples of five natural cohesive soils for this purpose and measured the chemical composition, mineral composition and specific surface area. We also performed particle-size analysis. The soil chemical composition and grain-size distribution analyses made a more accurate mineralogical determination possible. Intergrain water quantities at the plastic and liquid limits that were calculated from mineralogical properties cannot be compared directly with experimentally measured values, because Ca-montmorillonite and, consequently, interlayer water were present in all of the soils. The interlayer water content first had to be calculated using the quantity of montmorillonite and the internal specific surface area.

Due to the small number of samples tested, the general applicability of the chosen method could not be confirmed, in spite of the fact that the procedure for determining the Atterberg limits based on soil composition was performed on randomly selected soils of different composition and from different locations. This method for calculating liquid and plastic limits in geomechanics will not replace existing procedures, yet it is essential for understanding the interdependence between the quantity of water in soils and their mineralogical characteristics.

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