Fines Classification Based on Sensitivity to Pore-Fluid Chemistry
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Abstract: The 75-μm particle size is used to discriminate between fine and coarse grains. Further analysis of fine grains is typically based on the plasticity chart. Whereas pore-fluid-chemistry-dependent soil response is a salient and distinguishing characteristic of fine grains, pore-fluid chemistry is not addressed in current classification systems. Liquid limits obtained with electrically contrasting pore fluids (deionized water, 2-M NaCl brine, and kerosene) are combined to define the soil “electrical sensitivity.” Liquid limit and electrical sensitivity can be effectively used to classify fine grains according to their fluid-soil response into no-, low-, intermediate-, or high-plasticity fine grains of low, intermediate, or high electrical sensitivity. The proposed methodology benefits from the accumulated experience with liquid limit in the field and addresses the needs of a broader range of geotechnical engineering problems. DOI: 10.1061/(ASCE)GT.1943-5606.0001420. This work is made available under the terms of the Creative Commons Attribution 4.0 International license, http://creativecommons.org/licenses/by/4.0/.

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Introduction

Soil classification is intended to help engineers anticipate soil response and physical properties. Soil classification systems for geotechnical engineering purposes have evolved to properly address prevailing needs. Many geotechnical problems involve changes in pore-fluid chemistry. These range from classical geotechnical systems (e.g., dispersion in dams, rainfall driven hydro-chemo coupled erosion of slopes and scouring), geoenvironmental problems [e.g., landfills, nonaqueous phase liquids (NAPL), and salt water intrusion associated to sea level rise], and energy-related geotechnical problems (e.g., shale instability during well drilling, water flooding for oil production, and CO₂ injection for enhanced oil recovery or geological storage).

Classification systems worldwide are based on grain-size distribution and Atterberg limits (Fig. 1). In general, (1) grains are labeled fine when they are smaller than 75 μm (sieve No. 200); (2) the transition between fines-dominant and coarse-dominant behavior is at a fines content between 35 and 50; and (3) the liquid limit (LL) of 50 distinguishes between low- and high-plasticity fine-grained sediments.

The 75-μm size discriminator adequately captures differences in formation history, particle shape, and governing interparticle forces between coarse or fine grains (Santamarina et al. 2001). Further differentiation among fine-grained sediments becomes less clear. This ambivalence is in part caused by the extensive use of the term clay to refer to pastes that harden and gain strength during firing (e.g., china dishes; Mackenzie 1963), to particles made of phyllosilicate minerals such as kaolinite and smectite (not all phyllosilicates are clay minerals, e.g., mica; van Olphen 1977; Nesse 2000; Mitchell and Soga 2005), to particles smaller than 2-μm diameter or ∼1-m²/g specific surface (hence the frequent association with colloids and submicron particles that experience Brownian motion in water; Baver et al. 1972), and to soils that plot above the A-line on the Casagrande chart (Casagrande 1938, 1948; Holtz et al. 2011).

Casagrande’s plasticity chart is used in the Unified Soil Classification System (USCS) and in most other geotechnical classification systems. It properly discerns siltlike materials with high LL and plastic limit (PL) yet low plasticity index (PI = LL – PL) (such as diatoms) from sediments with high liquid limit and high plasticity index (such as bentonite). However, the chart has its limitations:

- Clay minerals often plot below the A-line and are classified as “silt” even with liquid limit as high as LL = 250 [see examples in Casagrande’s original charts in 1938 and 1948, and multiple cases in the data compilation reported in Fig. 2(a)].
- Organic soils may be found above or below the A-line (Howard 1984).
- The classification of mixtures made of plastic and nonplastic grains may be determined by the weight of the coarse nonplastic fraction, whereas the sediment hydraulic and mechanical properties remain controlled by the high-plasticity fines (further details are discussed subsequently).
- For high-plasticity clays, the plasticity index is dominated by the liquid limit, PI is strongly correlated with LL [Fig. 2(b); see also Seed et al. 1964b], and the plastic limit provides limited additional information for the purposes of clay classification (the plastic limit remains as a convenient field test to assess optimal water content during the compaction of silty and clayey soils; Wesley 2010).

Pore-fluid chemistry, i.e., pH, ionic concentration, and permittivity, is not addressed in current classification systems. Yet, pore-fluid-chemistry-dependent fabric and volumetric strains, because of changes in pore-fluid chemistry, are salient distinguishing characteristics of fine-grained sediments (Lambe 1953; Mitchell 1956; Yong and Warkentin 1966; Santamarina et al. 2002a; Mitchell and Soga 2005). In particular, the aggregation of clay platelets changes when the salt concentration exceeds the threshold of 0.01–0.1 mol/L (Palomino and Santamarina 2005).

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Fig. 1. Soil classification systems: (a) mixture qualifiers based on fines content (percent passing sieve No. 200); (b) plasticity qualifiers based on liquid limit (fraction passing sieve No. 40).

Fig. 2. Plasticity chart: (a) various soils on the plastic chart; (b) clay minerals only [extracted from the complete data set shown as Fig. 2(a)]; the linear regression (LR) between liquid limit and plasticity and ±1 standard deviation (σ = 20) boundaries plot between the A-line and the LL >> PL line.

Liquid limit data are plotted against specific surface in Fig. 3. Superimposed trends show published empirical equations, and three geometric models used to estimate the water content from (1) water adsorbed onto mineral surfaces (lower bound), (2) water held in dispersed fabrics, and (3) water contained in flocculated fabrics (upper bound). Clearly, the liquid limit test measures not only water adsorbed onto particle surfaces (i.e., proportionality with specific surface) but also held in the pore space of the chemistry-dependent fabric (Warkentin 1961; Farrar and Coleman 1967; Warkentin 1972; Wetzel 1990; Muhunthan 1991; Cerato and Lutenegger 2002; Santamarina et al. 2002a, b).

This study aims to explicitly address the effect of pore-fluid chemistry on the classification of fine grains in view of frequent field conditions that involve hydro-chemo-thermo-mechanically coupled chemistry. Several index tests were considered as potential candidates for the classification of fine grains in terms of electrical sensitivity, including sedimentation, dilation, and liquid limit tests (the complete study is reported in Jang 2014). The liquid limit did not suffer from segregation or boundary effects, provides consistent and repeatable values, and benefits from the accumulated experience in the field. Therefore, the liquid limit test was selected for the rest of this study.

Three fluids with different relative permittivity \( \kappa' \) and electrical conductivity \( \sigma_{el} \) are identified to explore distinct electrical fluid-particle interactions: deionized water (\( \kappa' = 80, \sigma_{el} = 10^{-6} \text{ S/m} \)), NaCl brine (concentration \( c = 2 \text{ M}, \kappa' = 55, \sigma_{el} = 12 \text{ S/m} \)), and kerosene (\( \kappa' = 2, \sigma_{el} = 10^{-11} \text{ S/m} \)). These common fluids are available at geotechnical laboratories worldwide.

### Test Procedure: Liquid Limit

Liquid limits were determined by using the fall cone test method to reduce experimental variability with the standard “Casagrande cup” method (Casagrande 1958; Sowers et al. 1960; Sherwood and Ryley 1970; BS 1377 1990; Dueñas and Poblete 2014). The 80-g, 30-degree- apex cone is allowed to penetrate the paste for 5 s (see Evans and Simpson (2015) for a more advanced test methodology). The liquid limit is the paste water content when the 5-s penetration equals 20 mm, and it corresponds to an undrained shear strength of 1.7 \( \sim 2.7 \text{ kPa} \) and a suction of \( \sim 6 \text{ kPa} \) (Hansbo 1957; Russell and Mickle 1970; Wroth and Wood 1978; Koumoto and Houlsby 2001; Mitchell and Soga 2005).

The presence of nonplastic coarse grains reduces the measured liquid limit as shown in Fig. 5 (see also Wintermayer 1926; Lambe 1951; Seed et al. 1964b). The trend can be explained by considering a coarse grain embedded in a clay paste with \( LL_{clay} \) (see inset in Fig. 5): water is held in the clay matrix and the mixture’s \( LL_{mix} \) is linearly dependent on the clay fraction \( LL_{mix} = LL_{clay} \cdot CF \), where clay fraction \( CF \) is defined as \( CF = W_{clay}/(W_{sand} + W_{clay}) \). The trend is the same for clay-sand mixtures (fraction between sieves No. 40 and No. 200) and for clay-silt mixtures (captured in the concept of “activity”; Skempton 1953). The goal of this study is to assess the pore-fluid chemistry dependency of fines; hence, the reported liquid limit tests are conducted on the passing sieve No. 200 fraction (this does not apply to Ottawa 20-30, which is included in the data set as an extreme case).

### Materials

Distinct soils are selected for this study, including Ottawa 20-30 sand, silica flour, diatoms, fly ash, kaolinite, illite bentonite, and ground organic matter. The salient properties for all tested fine grains are summarized in Fig. 4.
Soils are dried before preparing mixtures with kerosene. Drying during sample preparation affects test results in diagenetically modified soils such as volcanic ash [Casagrande 1932; ASTM D4318 (ASTM 2005); Herrera et al. 2007; Wesley 2010]. Soils should be dried at 60°C when such conditions are anticipated [ASTM D2216 (ASTM 2010)].

The liquid limit measured with deionized water \( LLDW \) is intended to avoid the face-to-face aggregation of clay platelets when the ionic concentration exceeds the threshold of \( 0 \text{.}01 – 0\text{.}1 \text{ mol}\text{=L} \), as noted previously. Therefore, sediments saturated with high-ionic-concentration saline water must be washed with deionized water to reduce the ionic concentration before measuring \( LLDW \).

**Results and Observations**

Liquid limits determined with deionized water \( LLDW \), NaCl brine \( LLL_{\text{brine}} \) and kerosene \( LLL_{\text{ker}} \) are summarized in Fig. 4. The ratios \( LLD/W/LL_{\text{brine}} \) and \( LLD/W/LL_{\text{ker}} \) are corrected to account for differences in water versus kerosene unit weight \( \gamma \) through specific gravity \( \gamma_{\text{ker}} = \gamma_{\text{ker}}/\gamma_{\text{ker}} \), and the precipitation of excess salts during oven drying when the NaCl brine with concentration \( c_{\text{brine}} (g/g) \) is used to run the test.

\[
\frac{LLD/W/LL_{\text{brine}} \text{ corrected}}{LLD/W/LL_{\text{ker}} G_{\text{ker}}} = \frac{LLD/W/LL_{\text{brine}} \text{ corrected}}{LLD/W/LL_{\text{ker}} G_{\text{ker}}} \text{ (1)}
\]

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\]
Fluid permittivity: The effect of permittivity is more complex. Wettability: The affinity of pore fluids to particle surfaces can affect the measured liquid limits in sediments with intragrain porosity, as wetting fluids can invade the small pores more readily and yield higher LL than nonwetting fluids. Although most natural sediments are water-wet, there are exceptions, such as fly ash, which prefer kerosene to water and exhibit \( LL_{ker} > LL_{brine} \).

Discussion: Recommended Classification for Fine Grains

Liquid Limit

Data compilations have led to valuable correlations between the liquid limit and engineering soil properties, such as hydraulic conductivity (Carrier and Beckman 1984), compressibility (Sridharan and Nagaraj 2000), and shear strength (Jamiolkowski et al. 1985; Mayne 2006; Haigh et al. 2013). Given the physical meaning and engineering usefulness of the liquid limit, it is retained in this study for the classification of fine grains.

Electrical Sensitivity

The experimental results summarized in Figs. 4 and 6 highlight the complexity of electrical interactions in fine grains whereby particle size and shape, surface and edge charges, and pore-fluid characteristics determine interparticle-electrical forces, define fabric formation, and affect sediment behavior (Yong and Warkentin 1966; Mesri and Olson 1970; Ridley et al. 1984; Chan et al. 1986; Bowders and Daniel 1987; Sivapullaiah and Sridharan 1987; Acar and Olivieri 1989; Meegoda and Ratnaweera 1994; Palomino and Santamarina 2005; Calvelo et al. 2005; Mishra et al. 2012).

The corrected liquid limit ratios are plotted as \( LL_{DW}/LL_{brine} \) versus \( LL_{ker}/LL_{brine} \) in Fig. 6 for all tested grains including additional data found in the literature. When either LL ratio is less than 1, the reciprocal value is plotted on the opposite quadrant to attain a symmetric assessment of electrical sensitivity to changes in pore fluid:

- Ottawa 20–30 sand, fly ash, silica flour, and diatoms exhibit low or no sensitivity to pore fluids, but kaolinite, illite, and bentonite display clear pore-fluid effects. Organic fines adsorb and swell with water, almost unaffected by salt concentration \( LL_{DW} \approx LL_{brine} \), but do not respond to nonpolar kerosene; therefore, \( LL_{brine} > LL_{ker} \).
- Fluid conductivity: In general, \( LL_{DW} > LL_{brine} \); hence, most soils plot on quadrants on the right. This result is consistent with changes in double-layer thickness and associated repulsion force (Mitchell and Soga 2005).
- Fluid permittivity: The effect of permittivity is more complex. High-plasticity sediments, probably with prevalent 2:1 clays, display \( LL_{brine} > LL_{ker} \). Yet, coarser sediments and even 1:1 kaolinite exhibit \( LL_{ker} > LL_{brine} \). The distinct response of kaolinite to low-permittivity fluids as compared with other clay minerals has been observed by others and attributed to edge charges and van der Waals forces (data compilation and analysis are in Santamarina et al. 2001, 2002a; see forces in Israelachvili 2011).
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Electrical Sensitivity

The electrical sensitivity \( S_E \) is defined to capture in a single parameter changes in liquid limit with pore-fluid permittivity and electrical conductivity, i.e., van der Waals and double-layer effects. For the first quadrant where \( LL_{ker}/LL_{brine} > 1.0 \) and \( LL_{DW}/LL_{brine} > 1.0 \) (the inset in Fig. 6 shows Pythagorean distance)

\[
S_E = \sqrt{\left(\frac{LL_{DW}}{LL_{brine}} - 1\right)^2 + \left(\frac{LL_{ker}}{LL_{brine}} - 1\right)^2} \tag{3}
\]

where \( S_E \) = distance from the origin at \( LL_{ker}/LL_{brine} = 1.0 \) and \( LL_{DW}/LL_{brine} = 1.0 \) to the data point [corrected LL ratios—Eqs. (1) and (2)]. The reciprocal LL ratios are used in Eq. (3) if they are less than 1.0 and data points fall in the second, third, or fourth quadrants.

Recommended Classification

A new classification chart is proposed in Fig. 7. It identifies soils on the basis of their electrical sensitivity \( S_E \) and the liquid limit measured with brine \( LL_{brine} \); the value obtained with NaCl brine is selected to minimize any ambiguity associated with existing ions in the soil. Then, the recommended procedure for the classification of fine grains is as follows:

1. Use the soil fraction that passes sieve No. 200.
2. Determine the liquid limit using the fall cone test (BSI 1990) for soil pastes prepared with the following three pore fluids: deionized water, kerosene, and 2-M NaCl brine.
3. Compute LL ratios [Eqs. (1) and (2)], and calculate the electrical sensitivity \( S_E \) [Fig. 6 and Eq. (3)].
4. Identify soil types by using the chart in Fig. 7.
5. Report the classification as no-, low-, intermediate-, or high-electrical sensitivity.

Observations

Sediments listed in Fig. 4 are classified by using the standard USCS chart and the proposed chart. Whereas there are basically four
It is recommended that fine grains are classified by using the proposed procedure and chart as no-, low-, intermediate-, or high-plasticity fine grains of low, intermediate, or high electrical sensitivity.

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