Statistical variability of the correlation plasticity index versus liquid limit for smectite and kaolinite

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ABSTRACT

An extensive literature review has been conducted to observe the statistical correlation of the plasticity index, PI, with the liquid limit, LL, of smectite and kaolinite. Fifty-nine data for smectite and fifty-one for kaolinite have been plotted and compared to each other. The results show that PI is related to LL with equations PI = 0.97 x LL - 37.6 for smectite and PI = $5.94e^{0.023.LL}$ for kaolinite. An independent data set was used for the validation of the proposed relationships. Besides, it was possible to identify a confidence interval for PI, relative to a certain interval for LL values, to confirm the robustness of the relations given above. The findings of this research show that the relation between the Atterberg limits is clearly controlled by the clay mineralogy and that there is no unique way to get PI from LL if the clay mineralogy is not considered.

Keywords: Atterberg limits; smectite; kaolinite; probabilistic analysis; confidence interval.

NOTATION LIST

PI	Plasticity index (%)
LL	Liquid limit (%)
PL	Plastic limit (%)
R^2	Regression coefficient
k	Number of the available experimental points
$\overline{m}$	Mean values of m (linear regression equation)
$\overline{n}$	Mean value of n (linear regression equation)
$t_{\eta\%,n-2}$	Parameter t of Student, relative to a confidence level of η% and a degree of freedom of k-2
σ	Standard deviation

INTRODUCTION

Liquid (LL) and plastic limits (PL) are the basic geotechnical index parameters for the qualitative assessment of the physical properties of fine-grained soils. Atterberg (1911), who described first these parameters, stated that *"the liquid limit represents the state at which two small pieces of clay placed in a bowl no longer flow together when a bowl is struck violently and repeatedly on the hand*" (Haigh, 2012), and it physically describes the water content at the transition from the liquid to the pulpy state of a soil. Soils at LL have small shear strength, which is in the range between 0.5–5.6kPa (e.g. Wasti & Bezirci, 1986; Sridharan & Prakash, 1998). PL represents the water content at the transition from the rigid to the semi-solid state of a soil (DIN, 1997). Currently, there are two methods for obtaining LL: the Casagrande (1932) standardized cup method, which is a procedure currently defined in DIN 18122 part 1 (1997), AASHTO T89-07 (2007) and ASTM D4318-10 (2010), and the fall-cone-based method, standardized by the ISO/TS 17892-12 (2004) (Spagnoli, 2012). Regarding PL, the geotechnical standard procedure is given by the rolling test method.

Two different clay types have been investigated which represent the two extreme types of clay minerals: kaolinite (2-layered clay minerals) and smectite (3-layer clay minerals).

The latter has a 2:1 silica:alumina structure, with very weak van der Waals' forces (Sridharan, 2014), and repulsive forces between clay platelets which govern swelling (Taylor and Smith, 1986), mainly for Na-smectite (Olson and Mesri, 1970).

For kaolinite, positive cation exchange capacity was measured under low pH conditions when edges are positively charged indicates that some isomorphous substitution must exist (Mitchell and Soga, 2005) which was also shown by Brady et al. (1996) and Israelachvili (2011).

As stated by Sridharan and Venkatappa Rao (1975), as these clays represent the extreme types of clay minerals, any natural clay is likely to behave, from the geotechnical point of view, in between these two. It is important to observe how the Atterberg limits will change depending on the clay mineralogy. Several authors tried already to assess the behavior of clays regarding their basic geotechnical properties (e.g.: White, 1949; Seed et al., 1964; Dusseault and Scafe, 1979; Nagaraj and Jayadeva, 1983; Sivapullaiah and Sridharan, 1985; Sridharan et al., 1988; Panadian & Nagaraj, 1990; Mitchell and Soga, 2005; Polidori, 2007; Dolinar & Škrabl, 2013), however no unique correlation was given. White (1949) found that LL of kaolinite increased with decreasing particle size, whilst Seed et al. (1964) obtained a linear correlation between LL and percentage of clay size for washed sand with kaolinite. Nagaraj & Jayadeva (1983) found a relationship, where the plasticity index, PI, was 0.74 x (LL-8), based on statistical approaches, critical state concepts and on the Gouy-Chapman theory of double layer. However, as stated by Sridharan (2014), since kaolinite and smectite behave quite differently from each other, the mechanisms governing the Atterberg limits, and in turn LL, of kaolinite and smectite are different. The present study summarizes the results of forty-four published data, where the Atterberg limits for almost pure clays were given. From the statistical point of view the confidence interval for confidence levels of 95 and 99% for both smectite and kaolinite has been assessed.

METHODOLOGY

Data from literature about LL and PL for smectite and kaolinite (or well-known natural clays with a predominant clay mineralogy) were carefully analyzed. Only Atterberg limits obtained with the Casagrande cup and the rolling method were used. Regarding smectitic clays, where possible, the main cation was indicated. However, according to Bain (1971), it is possible to roughly distinguish between Na-smectite and Ca-smectite considering their PI values. Clay fraction less than 2µm were also indicated. Only Foreman and Daniel (1986), indicated the clay faction corresponding to 5µm.

Forty-four different published data were used to obtain the Atterberg limits for pure clays, i.e. smectitite and kaolinite mixed with water. Tab. 1 and 2 summarize the values used for the interpretation of the Atterberg limits for natural clays mixed with water. LL values are obtained with the Casagrande cup. Fifty-nine data were used for the interpretation for the smectite, and fifty-one for the kaolinite.

No.	Reference	< 2μm (%)	LL (%)	PL (%)	PI (%)	Cation
1	Seed et al. (1964)	95.5	521.5	48	473.5	not specified
2	Andrews et al. (1967)		506	55	451	Na
3	Mesri and Olson (1971)	97	675	39	636	not specified
4	Sridharan et al. (1973)	-	305	44	261	not specified
5	Sridharan and Venkatappa Rao (1975)	-	306	44	262	not specified
6	Samarasinghe et al. (1982)	-	118	46	72	Ca
7	Sridharan et al. (1986a)	100	495	49.2	445.8	Na
8	Sridharan et al. (1986a)	100	233	57.8	175.2	K
9	Sridharan et al. (1986a)	100	125	40.6	84.4	Ca
10	Sridharan et al. (1986a)	100	675	49.1	625.9	Li
11	Sridharan et al. (1986b)	37	84	42	42	Ca
12	Sridharan et al. (1986b)	31	100	45.2	54.8	Ca
13	Sridharan et al. (1986b)	42	106.4	44.1	62.3	Ca
14	Sridharan et al. (1986b)	39	124.2	23.2	101	Ca
15	Wasti and Bezirci (1986)	88	526	38	488	not specified
16	Sivapullaiah et al. (1987)	-	337	55.6	281.4	not specified
17	Rao et al. (1989)	100	348	43.9	304.1	not specified
18	Acar and Olivieri (1989)	12	88	54	34	Ca
19	Acar and Olivieri (1989)	80	425	58	367	Na
20	Abdul et al. (1990)	-	470	45	425	Na
21	Di Maio and Fenelli (1994)	100	330.6	55.2	275.4	not specified
22	Abu-Hassanein et al. (1996)	85	608	43	565	Na
23	Abu-Hassanein et al. (1996)	94	516	51	465	Na
24	Di Maio (1996)	80	400	80	320	Na
25	Gleason et al. (1997)	-	603	36	567	Na
26	Gleason et al. (1997)	-	590	37	553	Na
27	Gleason et al. (1997)	-	124	26	98	Ca
28	Gleason et al. (1997)	-	123	38	85	Ca

29	Sridharan et al. (1997)	38	74.7	20	54.7	not specified
30	Petrov et al. (1997)	-	530	50	480	Na
31	Robinson and Allam (1998)	98	321	58	263	Na
32	Hettiaratchi et al. (1999)	-	407	105	302	Na
33	Hettiaratchi et al. (1999)	-	98	61	37	Ca
34	Sridharan and Nagaraj (1999)	100	320	56.6	263.4	Na
35	Kayabal and Bulus (2000)	-	320	50	270	Na
36	Karunaratne et al. (2001)	78	465	41	424	not specified
37	Tuncan et al. (2003)	88	447	60	387	Na
38	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	100	205	35	170	not specified
39	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	90	184.5	31.5	153	not specified
40	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	80	164	28	136	not specified
41	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	70	143.5	24.5	119	not specified
42	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	60	123	21	102	not specified
43	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	50	102.5	17.5	85	not specified
44	Young Jo et al. (2004)	88	504	39	465	Na
45	Sivapullaiah and Lakshmikantha (2004)	-	310	49	261	Na
46	Kaya and Fang (2005)	-	440	70	370	Na
47	Mishra et al. (2005)	61.4	310.5	54.1	256.4	not specified
48	Spagnoli et al. (2010)	85	455	70	385	Na
49	Shariatmadari et al. (2011)	-	199.4	41.5	157.9	not specified
50	Younus and Sreedeep (2012)	49	224	31	193	not specified
51	Kumar Pal and Ghosh (2013)	64	159	37	122	not specified
52	Tiwari and Ajmera (2014)	-	148	49	99	not specified
53	Mir and Sridharan (2014)	63	84	25	59	Na
54	Ghazi (2015)	-	310	56	254	Na
55	Rageena and Rani (2015)	73	245	46	199	Ca
56	Ghadyani et al. (2016)	65	238	66	172	Na
58	Jang and Santamarina (2016)	-	276	44	232	not specified

57	Fan et al. (2017)	-	331.4	88.2	243.2	Ca
59	Deka and Sekharan (2017)	64	300	53	247	not specified

Table 1 Atterberg limits for smectitic clays with water as fluid

No.	Reference	< 2µm (%)	LL (%)	PL (%)	PI (%)
1	Andrews et al. (1967)	-	62	33	29
2	Mesri and Olson (1971)	47	45	29	16
3	Sridharan et al. (1973)	54	49	29	20
4	Sridharan and Venkatappa Rao (1975)	-	49	29	20
5	Littleton (1976)	-	83	30	53
6	Genevois (1977)	67	65	36	29
7	Genevois (1977)	67	73	44	29
8	Genevois (1977)	67	70	42	28
9	Genevois (1977)	67	69	39	30
10	Genevois (1977)	67	60	38	22
11	Genevois (1977)	67	61	39	22
12	Genevois (1977)	67	58	38	20
13	Genevois (1977)	67	64	44	20
14	Genevois (1977)	67	63	38	25
15	Genevois (1977)	67	58	37	21
16	Genevois (1977)	67	72	42	30
17	Genevois (1977)	67	122	63	59
18	Rao and Sridharan (1985)	54	49	29	20
19	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	100	45	29	16
20	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	90	40.5	26.1	14.4
21	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	80	36	23.2	12.8
22	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	70	31.5	20.3	11.2
23	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	60	27	17.4	9.6
24	Mesri and Cepeda-Diaz (1986)/Polidori (2003)	50	22.5	14.5	8
25	Foreman and Daniel (1986)	98	54	31	23
26	Bowders and Daniel (1987)	-	58	34	24

27	Sivapullaiah et al. (1987)	-	51	34	17
28	Sridharan et al. (1988)	25	25	13.8	11.2
29	Sridharan et al. (1988)	27	38	15.3	22.7
30	Acar and Olivieri (1989)	90	64	34	30
31	Abdul et al. (1990)	-	61	37	24
32	Meegoda and Ratnaweera (1994)	84	48	36	12
33	Di Maio and Fenelli (1994)	100	57.5	37.8	19.7
34	Robinson and Allam (1998)	-	53	32	21
35	Sridharan and Nagaraj (1999)	11.5	58.7	45.2	13.5
36	Sridharan and Nagaraj (1999)	32	55	31.4	23.6
37	Kumar and Muir Wood (1999)	95	80	39	41
38	Lemos and Vaughan (2000)	82	69	38	31
39	Karunaratne et al. (2001)	87	74	34	40
40	Kaya and Fang (2005)	-	42	29	13
41	Sentenac et al. (2006)	-	54	31	23
42	Polidori (2007)	97	62	36	26
43	Di Matteo et al. (2011)	34	56.51	34	22.51
44	Spagnoli et al. (2012)	48	57	35	22
45	Khosravi et al. (2013)	20	45	26	19
46	Estabragh et al. (2014)	25	47	20	27
47	Tiwari and Ajmera (2014)	-	65	36	29
48	Tiwari and Ajmera (2014)	-	70	30	40
49	Ghadyani et al. (2016)	30	32	22	10
50	Jang and Santamarina (2016)	-	67	31	36
51	Fan et al. (2016)	-	29.1	19.5	9.6

Table. 2 Atterberg limits for kaolinitic clays with water as fluid

RESULTS AND DISCUSSION

Correlations found

As LL value of clays depends on the type of clay mineral with associated cations (Mitchell and Soga, 2005), smectite and kaolinite have been analyzed separately. Fig. 1 shows the relation PI vs LL for smectitic clays. A linear correlation shows that PI = 0.97 x LL - 37.6, with a very good correlation coefficient of $R^2 = 0.99$. The correlation PI vs LL for smectite is statistical significant as p-value is <0.05 (i.e. $5.18 \cdot 10^{-61}$). The correlation matches very well with that found by Seed et al. (1964), where PI = 0.98 x LL - 27.5, who investigated artificial kaolinite-quartz mixtures in different amounts. The correlation found by Nagaraj & Jayadeva (1983) was PI = 0.74 x LL-8; however, this was based on natural clays coming from different depths with inhomogeneous mineralogy. Regarding the kaolinite data, an exponential correlation between LL and PI was found (Fig. 2). The equation was in this case PI = $5.94e^{0.023.LL}$ and was characterized by a correlation coefficient of $R^2 = 0.80$. With respect to the data for the smectite, the results are more scattered. However, the correlation PI vs LL for kaolinite is also statistical significant as p value <0.05 (i.e. $1.74 \cdot 10^{-19}$).

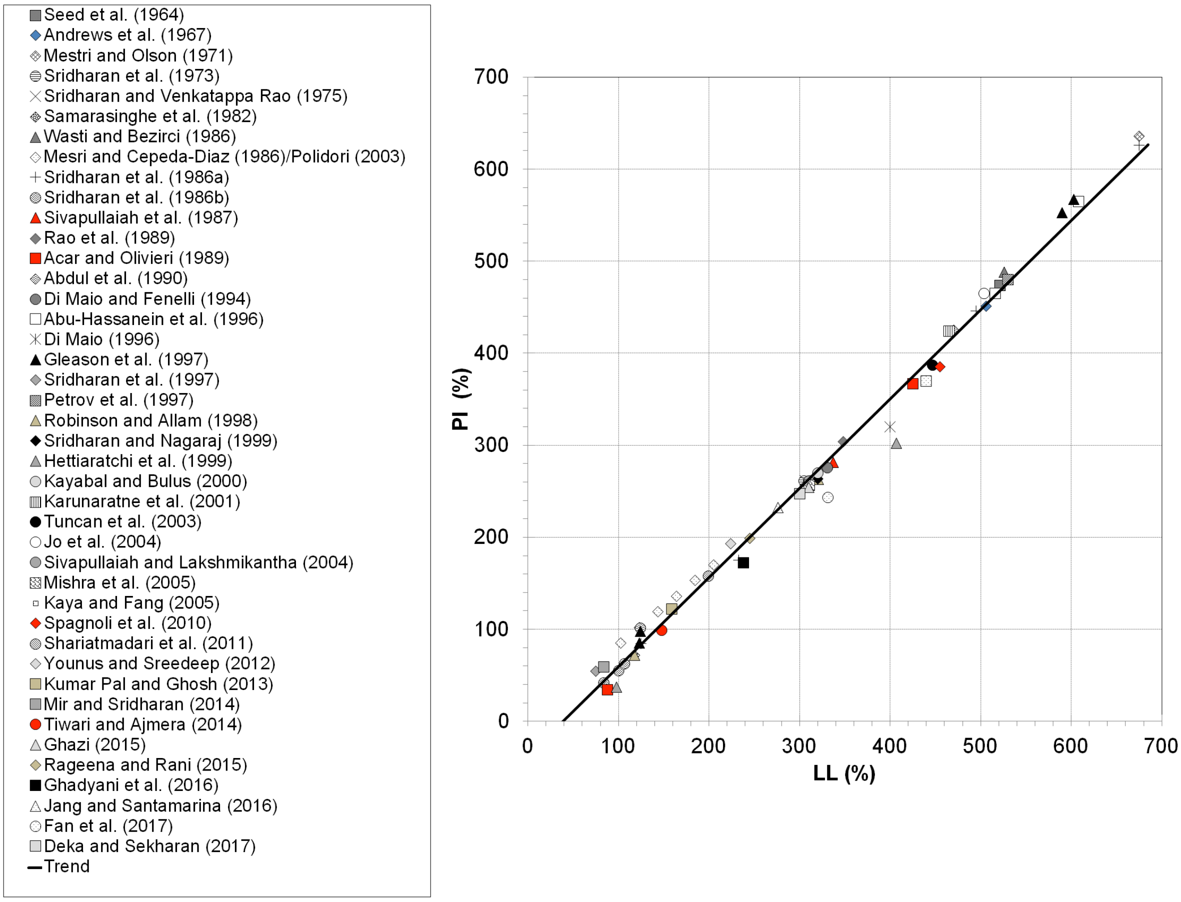


Fig. 1 PI vs LL for smectitic clays.

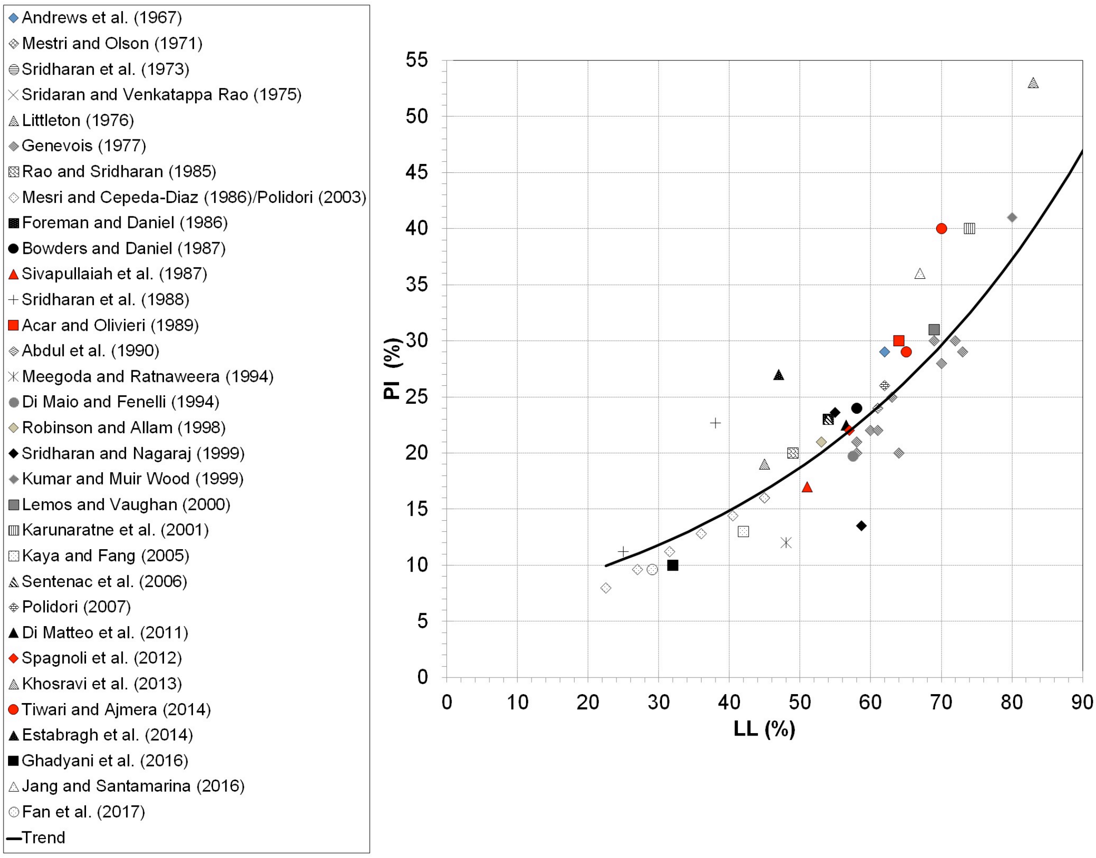


Fig. 2 PI vs LL for kaolinitic clays

Bearing in mind these findings, and considering the coefficient of correlations observed for smectite (R^2 = 0.99) and kaolinite (R^2 = 0.80), a comparison between experimental PI, which were derived from another published data shown in Tab. 3 and predicted PI values (obtained from the equations mentioned above) has been shown in Fig 3A & 3B. As for Tabs. 1 and 2, for the values showed in Tab. 3 only data where Atterberg limit for the Casagrande cup on pure kaolinite and smectite were selected. Smectitic soils show a linear relation with a very good R^2 value (0.99), where $PI_{predicted}=1.04xPI_{experimental}$.

For kaolinitic soils the predicted PI tends to overestimate the lab. PI up to 20%. From this point forward the predicted PI values underestimate the lab PI. The relation has the form of $PI_{predicted}=0.48xPI_{experimental}+10.26$.

This is likely due to the smaller R^2 values for the PI vs LL correlation with respect to the one obtained for smectite. However, the regression coefficient, R^2, gives a value of 0.94. Besides, both p-values for Figs 3A & 3B show also a statistical significance ($p<0.05$) between the predicted vs lab PI values, with $3.60 \cdot 10^{-10}$ and $3.46 \cdot 10^{-06}$ for smectite and kaolinite respectively.

No.	Source	LL (%)	PI (%)	Clay type
1	Stadtbäumer (1976)	269	222	Smectite
2	Egashira and Ohtsubo (1982)	114	63	Smectite
3	Bell (1994)	114	47	Smectite
4	Stavridakis (1999)	111	68	Smectite
5	Sivapullaiah et al. (2003)	310	261	Smectite
6	Eisazadeh et al. (2012)	301	260	Smectite
7	Fatahi et al. (2013)	340	290	Smectite
8	Prakash and Sridharan (2013)	100.8	19	Smectite
9	Kolay and Ramesh (2016)	603	508	Smectite
10	Basmenj et al. (2017)	470	395	Smectite
11	Stadtbäumer (1976)	63	36	Kaolinite
12	Stavridakis (1999)	34	29	Kaolinite
13	Sridharan and Prakash (2001)	48	35	Kaolinite
14	Sridharan and Prakash (2001)	44	25	Kaolinite
15	Sridharan and Nagaraj (2005)	48	35	Kaolinite

16	Sridharan and Nagaraj (2005)	55	31	Kaolinite
17	Park et al. (2006)	47	29	Kaolinite
18	Sachan et al. (2013)	65	30	Kaolinite
19	Pulat et al. (2014)	34	27	Kaolinite
20	Kolay and Ramesh (2016)	76	28	Kaolinite

Table. 3 Atterberg limits for smectitic and kaolinitic clays used for the validation of the predicted IP shown in Figs. 1 and 2

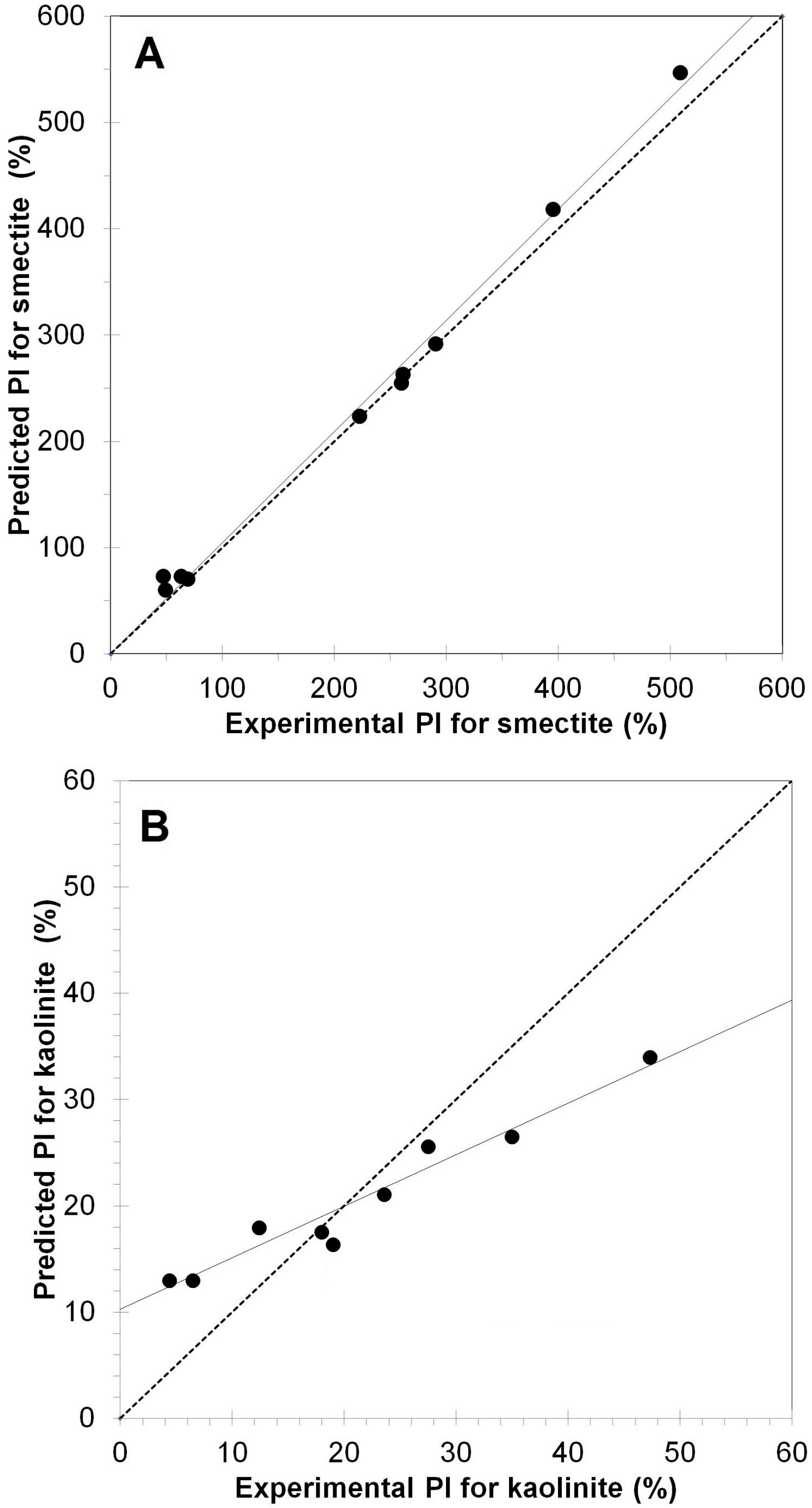


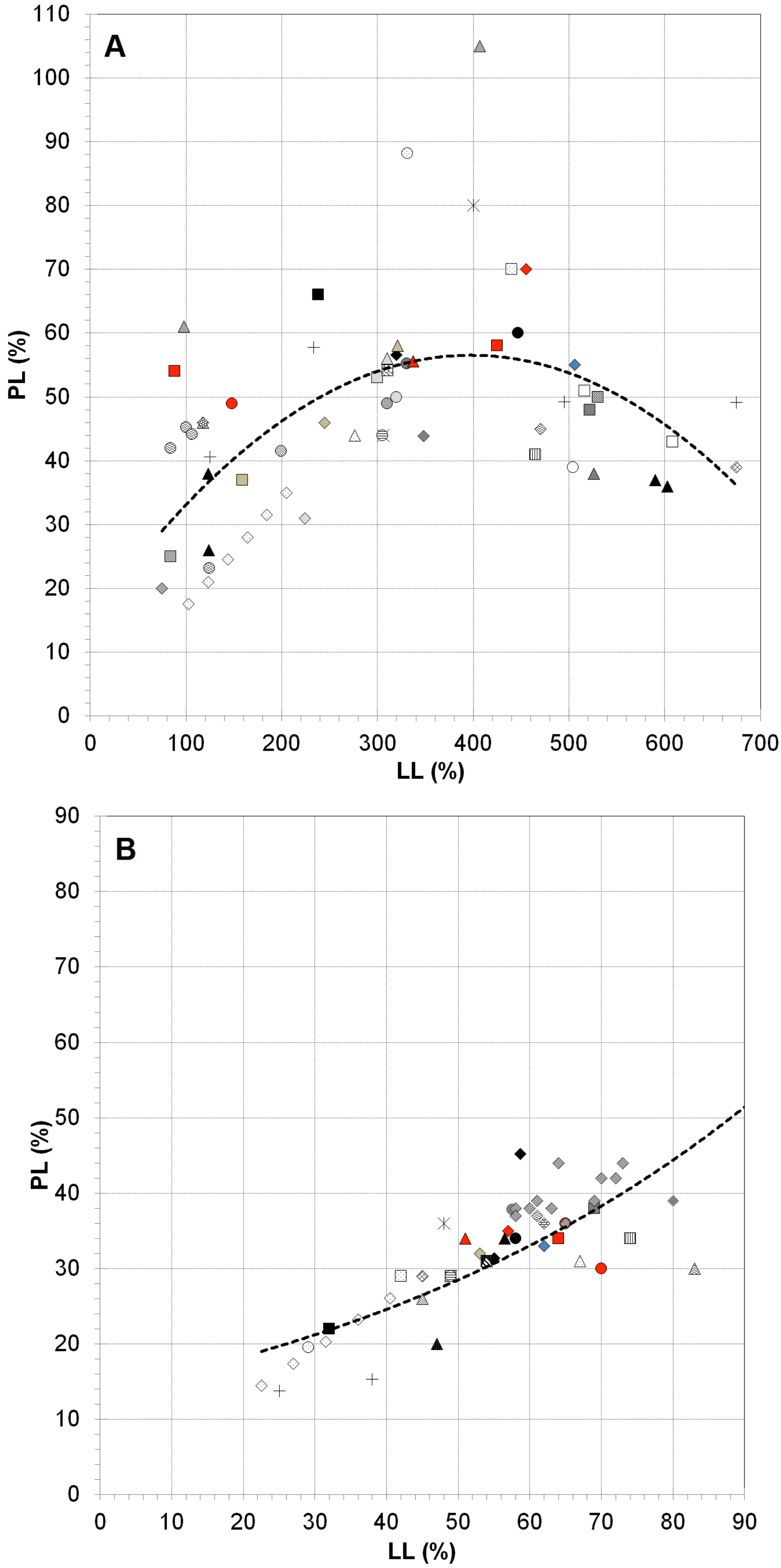
Fig. 3 Predicted PI vs experimental PI for smectitic clays (A); predicted PI vs experimental PI for kaolinitic clays (B). The experimental PI values refer to Tab. 3.

No significant correlation between LL and the percentage of clay size fraction was found for both smectite and kaolinite. This agrees with the findings of Sridharan et al. (1988), whereas disagrees with the statement of Seed et al. (1964) and Polidori (2007), who presented a linear variation of LL with the percentage of clay size fraction for quartz and pure clay mixtures. However, it is worth mentioning that the data of the literature reviewed used in this research refer to pure clays, which are normally characterized by a wider particle size distribution.

Fig. 4 (A and B) shows the relation PL vs LL as from Tab. 1 and 2 for both pure clays. The purpose of the diagram is not to find out a relation; it is rather to show how the parameters change with respect to each other. Smectitic clays (Fig. 4A) show a bell-shaped behavior, where the highest PL value does not correspond to the highest LL value. The increases in PL values follow increases in LL values up to a certain point, after which LL values increases but PL values decreases. While LL values are directly proportional to the water content and to the main cation involved, PL values show considerable variations (Bain, 1971). According to Haigh et al. (2013), PL relates to the capillary suction at which the water phase ceases to act as a continuum.

It is interesting to note that some Ca-smectites have PL values higher than the Na-smectite samples. PL variations might be due to the difficulties of the thread-rolling tests and also because due to the different drying (shrinkage) characteristics of the smectitic clays (Bain, 1971), where the shrinkage is directly proportional to the PI (Taylor and Smith, 1986). Recent work shows the electrochemical forces play role in shrinkage processes (Lu and Dong, 2017). In that case, PL is also dependent on the electrochemical forces similar to the LL.

Regarding the correlation PL vs LL for kaolinitic clays (Fig. 4B), the trend is similar as observed in Fig. 2, i.e. an exponential function links in an acceptable way the two parameters (R^2 = 0.70). However, the correlations shown in Fig. 4 are not meant to be statistically relevant.


A
PL (%)
LL (%)
110
100
90
80
70
60
50
40
30
20
10
0
0
100
200
300
400
500
600
700
B
PL (%)
LL (%)
90
80
70
60
50
40
30
20
10
0
0
10
20
30
40
50
60
70
80
90

161

Fig. 4 Correlation PL vs LL for smectitic (A) and kaolinitic clays (B). The correlating lines are dotted because they are not meant to give a statistical reference. Note that the legend is the same as per Figs. 1 and 2.

Estimation of the statistical variability of the PI *vs LL correlation*

In order to use the correlations obtained on the experimental measurements of Fig. 1 and 2 for smectite and kaolinite, an accurate probabilistic analysis is required. Since the collected data show some variability regarding the estimation of PI from LL, the estimation that can be made on PI leads to a probable range of variability rather than a simple deterministic value.

The confidence interval indicates the range that, with a certain probability (the confidence level), gives the true value of the parameter (Spagnoli et al. 2017).

A probabilistic analysis has been assessed for the validation of correlations PI vs LL shown in Fig. 1 & 2. A straight line, $y = m \cdot x + n$, better approximates the data (x_i and y_i) shown in Tab. 1 and 2 and Fig. 1 and 2. The coefficients m and n were determined with Cramer's method, which is useful for solving a system of linear equations using the determinant, in case the system has exactly one solution. It is assumed that the error in the determination of the parameter x is much smaller than one would have in the estimation of the parameter y, and that the error in the determination of each y_i is constant.

The uncertainty (standard deviation) σ_y on the parameters y_i is given by the following equation (Bacon, 1953):

$$\sigma_y = \sqrt{\cdot \frac{\sum_{i=1}^{k} \left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2}{(k-2)}} \qquad (1)$$

where k is the number of the available experimental points; $\overline{m}$ and $\overline{n}$ are the mean values of m and n and are obtained using the equations of the linear regression on the available data.

Applying the theory of propagation of errors, the uncertainties of the regression coefficients are obtained:

$$\sigma_m = \sqrt{\cdot \frac{\sum_{i=1}^{k} \left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2}{(k-2) \cdot \left(\sum_{i=1}^{k} x_i^2\right) - \frac{(k-2)}{k} \cdot \left(\sum_{i=1}^{k} x_i\right)^2}} \tag{2}$$

$$\sigma_n = \sqrt{\frac{\left(\sum_{i=1}^{k} x_i^2\right) \cdot \sum_{i=1}^{k} \left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2}{k \cdot (k-2) \cdot \left(\sum_{i=1}^{k} x_i^2\right) - (k-2) \cdot \left(\sum_{i=1}^{k} x_i\right)^2}} \tag{3}$$

The best estimation of the parameters *m* and *n* in relation to a certain level of confidence (expressed in percentage) is obtained through the confidence intervals:

$$m = \overline{m} \pm t_{\eta\%, k-2} \cdot \sigma_m \tag{4a}$$

$$n = \overline{n} \pm t_{\eta\%, k-2} \cdot \sigma_n \tag{4b}$$

where $t_{\eta\%, k-2}$ is the parameter *t* of Student, relative to a confidence level of η% and a degree of freedom of k-2. Student's t-distribution is a distribution of continuous probability governing the relationship between two random variables, the first with normal distribution, while, the second follows a squared distribution.

Based on the discussion above, it is possible to predict the value y_0 and its uncertainty, σ_{y0}, for a value of x_0. In fact:

$$\overline{y}_0 = \overline{m} \cdot x_0 + \overline{n} \tag{5}$$

Applying, then, the theory of propagation of errors to the equation 5, the standard deviation of y_0 is obtained by:

$$\sigma_{y0} = \sqrt{\cdot \frac{\sum_{i=1}^{k} \left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2}{(k-2)} \cdot \left[\frac{\left[\left(\sum_{i=1}^{k} x_i^2\right) - \frac{1}{k} \cdot \left(\sum_{i=1}^{k} x_i\right)^2\right] + \left[k \cdot x_0 - \left(\sum_{i=1}^{k} x_i\right)\right]^2}{k \cdot \left(\sum_{i=1}^{k} x_i^2\right) - \left(\sum_{i=1}^{k} x_i\right)^2} \right]} \tag{6}$$

The confidence interval for y_0 will be given, then, by the following expression, for a confidence level of η%:

$$y_0 = \bar{y}_0 \pm t_{\eta\%,k-2} \cdot \sigma_{y0} \qquad (7)$$

That means:

$$y_0 = \left(\overline{m} \cdot x_0 + \overline{n}\right) \pm t_{\eta\%,k-2} \cdot \sqrt{\frac{\sum_{i=1}^{k}\left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2}{(k-2)} \cdot \left[\frac{\left[\left(\sum_{i=1}^{k} x_i^2\right) - \frac{1}{k} \cdot \left(\sum_{i=1}^{k} x_i\right)^2\right] + \left[k \cdot x_0 - \left(\sum_{i=1}^{k} x_i\right)\right]^2}{k \cdot \left(\sum_{i=1}^{k} x_i^2\right) - \left(\sum_{i=1}^{k} x_i\right)^2}\right]} \qquad (8)$$

In this way, it will be possible to identify a range of variability of y, relative to a certain desired confidence level, corresponding to the parameter x_0 indicated.

Considering the data of this research for smectite (Fig. 1), the relation LL (x axis) vs PI (y axis) is shown. The linear regression analysis allowed to identify the line that best approximates the experimental points available (PI =0.97 x LL - 37.6). The standard deviation on the slope "m" is $\sigma_m = 0.012$; the standard deviation on constant term "n" is $\sigma_n = 4.157$. Moreover, we have: $k = 59$, $\sum_{i=1}^{k}\left(y_i - \overline{m} \cdot x_i - \overline{n}\right)^2 = 13417$, $\left(\sum_{i=1}^{k} x_i\right) = 18302$, $\left(\sum_{i=1}^{k} x_i^2\right) = 7381520$. If, for example, the confidence interval for PI with a confidence level of 95%, corresponding to a LL equal to 350% is requested, then PI = $301.38 \div 302.48$. The same PI confidence interval for a greater confidence level of 99%, would be PI = $301.20 \div 302.66$.

In the case of kaolinite, the relation PI -LL is exponential, as it can be seen from Fig. 2. Considering the same approach explained before, it is possible to analyze the relationship ln(PI)-LL for the available experimental data. In this case, it is possible to apply the linear regression and obtain, then, the line that best approximates the experimental data available. This line takes the form: ln(PI)= 0.026·LL + 1.61.

The standard deviation on the slope "m" is σ_m = 0.0012; the standard deviation on the constant term "n" is σ_n = 0.0998. Besides, we observe: $k = 51$, $\sum_{i=1}^{k}\left(y_i - \overline{m}\cdot x_i - \overline{n}\right)^2 = 1.52$, $\left(\sum_{i=1}^{k} x_i\right) = 2725.31$, $\left(\sum_{i=1}^{k} x_i^2\right) = 158641$. If, for example, the confidence interval for PI with a confidence level of 95%, corresponding to a LL equal to 50% is requested, then ln(PI)=2.91 $\div$ 2.93 and therefore PI=18.48 $\div$ 18.75. The same PI confidence interval for a greater confidence level of 99%, would be PI=18.44 $\div$ 18.80.

Both probabilistic assessments clearly confirm the robustness of the regression functions obtained for this research if the data shown in Fig. 1 and 2 are considered.

CONCLUSIONS

In this paper, results obtained from 44 technical papers for smectitic and kaolinitic clays respectively, published from different countries and authored by several authors over a period of 50 years have been analyzed with respect to Atterberg limits and some useful conclusions have been drawn. Good correlations have been obtained between liquid limit (LL) and plasticity index (PI) for both smectite and kaolinite. The range of LL varied up to 680% for smectitic and 85% for kaolinitic clays. It can be shown that the correlation of LL with PI for smectitic clays varies marginally with different ranges of LL. The predicted PI values haven been verified against another independent set of data, giving very good R^2 values, namely 0.99 and 0.94 for smectite and kaolinite respectively, although the predicted PI values compared with the lab PI values for kaolinite are not as precise as for smectite. The validity of the relation PI vs LL for both clay types has been also verified by a probabilistic analysis.

Besides, for smectitic clays it has been seen that PL increases along with LL up to a certain value and then it tends to decrease while LL still increases. Whereas, for kaolinitic clays, PL constantly increases with increasing LL values.

ACKNOWLEDGEMENT

The authors would like to thank the anonymous reviewer, who considerably increased the quality of the manuscript.

REFERENCES

AASHTO 2007 Standard method of test for determining the liquid limit of soils. AASHTO standard T89-07. American Association of State Highway and Transportation Officials (AASHTO). Washington. D.C.

Abdul, A.S., Gibson, T.L., Rai, D.N. 1990 Laboratory studies of the flow of some organic solvent and their aqueous solutions through bentonite and kaolin clays. *Groundwater*, **28**, 524-533.

Abu-Hassanein, Z., Benson, C., Wang, X., Blotz, L. 1996 Determining bentonite content in soil-bentonite mixtures using electrical conductivity. *Geotechnical Testing Journal*, **19**, 51-57, https://doi.org/10.1520/GTJ11407J.

Acar, Y.B., Olivieri, I. 1989 Pore fluid effects on the fabric and hydraulic conductivity of laboratory-compacted clay. *Transportation Research Record*, **1219**, 144-159.

Andrews, R. E., J. J. Gawarkiewicz, Winterkorn, H. F. 1967 Comparison of three clay minerals with water, dimethyl sulfoxide, and dimethyl formalmide. *Highway Research Record*, 209, 66-78.

ASTM 2010 Standard test methods for liquid limit. plastic limit. and plasticity index of soils. ASTM standard D4318-10. American Society for Testing and Materials. West Conshohocken. Pa.

Atterberg, A. 1911 Die Plastizität der Tone. *Internationale Mitteilungen der Bodenkunde*. **1**, 4‑37.

Bacon, R.H., 1953. The best straight line among the points. *American Journal of Physics*, 21, 428-446.

Bain, J.A. 1971 A plasticity chart as an aid to the identification and assessment of industrial clays. *Clay Minerals*, **9**, 1-17.

Basmenj, A.K., Mirjavan, A., Ghafoori, M., Cheshomi, A. 2017. Assessment of the adhesion potential of kaolinite and montmorillonite using a pull-out test device. *Bulletin of Engineering Geology and Hydrogeology*, **76**, 1507–1519, DOI 10.1007/s10064-016-0921-3.

Bell, F.G. 1994. An assessment of cement-PFA and lime-PFA used to stabilize clay-size materials. *Bulletin of Engineering Geology and Hydrogeology*, **49**, 25–32.

Bowders, J. J., Jr., Daniel, D. E. 1987 Hydraulic conductivity of compacted clay to dilute organic chemicals. *Journal of Geotechnical Engineering*, **113**, 1432–1448.

Brady, P.V., Cygan, R.T., Nagy, K.L. 1996. Molecular controls on kaolinite surface charge. *Journal of Colloid and Interface Science*, **183**,356-364.

CEN ISO/TS 17892-12 2004 Geotechnical investigation and testing-Laboratory testing of soil-Part 12: Determination of Atterberg limits. European Committee for Standardization.

Deka, A., Sekharan, S. 2017 Contaminant retention characteristics of fly ash-bentonite mixes. *Waste Management & Research*, **35**, 40-46, doi: 10.1177/0734242X16670002.

Di Maio, C. 1996 Exposure of bentonite to salt solution: osmotic and mechanical effects. *Geotechnique*, **46**, 695-707.

Di Maio, C., Fenelli, G. B. 1994 Residual strength of kaolin and bentonite: the influence of their constituent pore fluid. *Geotechnique,* **44**, 217–226.

Di Matteo, L., Bigotti, F., Ricco, R. 2011 Compressibility of Kaolinitic Clay contaminated by ethanol-gasoline blends. *Journal of Geotechnical and Geoenvironmental Engineering*, **137**, 846-849.

Di Matteo, L., Ricco, R., Filippini, L., Vinti, G. 2016 Permeability of remoulded low-plasticity clay contaminated by bioethanol-based fluids. *Bulletin of Engineering Geology and Hydrogeology*. **75**. 293–300. DOI 10.1007/s10064-015-0739-4.

DIN 1997 Baugrund Untersuchung von Bodenproben- Zustandsgrenzen (Konsistenzgrenzen) DIN 18122 Teil 1: Bestimmung der Fließ- und Ausrollgrenze. Beuth Verlag, Berlin.

Dolinar, B., Škrabl, S. 2013 Atterberg limits in relation to other properties of fine-grained soils. *Acta Geotechnica Slovenica*, **10**, 5-13.

Dusseault, M.B., Scafe, D., 1979. Mineralogical and engineering index properties of the Basalt McMurray formation of clay shales. *Canadian Geotechnical Journal*, 16, 285-294.

Egashira, K., Ohtsubo, M. 1982 Smectite in marine quick-clays of Japan. *Clays and Clay Minerals*, **30**, 275-28.

Estabragh, A.R., Beytolahpour, I., Moradi, M., Javadi, A.A. 2014 Consolidation behavior of two fine-grained soils contaminated by glycerol and ethanol. *Engineering Geology*, **178**, 102–108.

Eisazadeh, A., Kassim, K.A., Nur, H. 2012. Cation exchange capacity of phosphoric acid and lime stabilized montmorillonitic and kaolinitic soils. *Geotechnical and Geological Engineering*, **30**, 1435–1440.

Fan, R.D., Liu, S.Y., Du, Y.J., Reddy, K.R., Yang, Y.L. 201 Impacts of presence of lead contamination on settling behavior and microstructure of clayey soil - calcium bentonite blends. *Applied Clay Science*, **142**, 109-119, http://dx.doi.org/10.1016/j.clay.2016.10.042.

Fatahi, B., Le, T.M., Fatahi B., Khabbaz, H. 2013. Shrinkage Properties of Soft Clay Treated with Cement and Geofibers. *Geotechnical and Geological Engineering*, **31**, 1421–1435.

Foreman, D.E., Daniel. D.E. 1986 Permeation of compacted clay with organic chemicals. *Journal of Geotechnical Engineering*, **112**. 669-681.

Genevois, R. 1977 Chemical Interactions on the Compressibility of Remoulded Kaolin. Proc. 9th International Conference on Soil Mechanics and Foundation Engineering, 109-116.

Ghadyani, M., Hamidi, A., Hatambeigi, M. 2016 Triaxial shear behaviour of oil contaminated clays. *European Journal of Environmental and Civil Engineering*, http://dx.doi.org/10.1080/19648189.2016.1271359.

Ghazi, A. F. 2015 *Engineering characteristics of compacted sand-bentonite mixtures*. MSc thesis. Edith Cowan University. Australia.

Gleason, M.H. Daniel, D.E., Eykholt, G.R. 1997 Calcium and sodium bentonite for hydraulic containment applications. *Journal of Geotechnical and Geoenvironmental Engineering,* **123**, 438–445

Haigh, S.K. 2012 Mechanics of the Casagrande liquid limit test. *Canadian Geotechnical Journal*, **49**, 1015‑1023.

Haigh, S.K., Vardanega, P.J., Bolton, M.D. 2013 The plastic limit of clays. *Geotechnique*, **63**, 435–440, http://dx.doi.org/10.1680/geot.11.P.123.

Hettiaratchi, J.P.A., Achari, G., Joshi, R.C., Okoli, R.E. 1999 Feasibility of us-ing fly ash admixtures in landfill bottom liners or vertical barriers at contaminated sites. *Journal of Environmental Science and Health, Part A*, **34**, 1897-1917.

Israelachvili, J.N. 2011. *Intermolecular and Surface Forces*, 3rd Edition. Elsevier.

Jang, J. & Santamarina, J.C. 2016 Fines classification based on sensitivity to pore-fluid chemistry. *Journal of Geotechnical and Geoenvironmental Engineering*, **142**, 1-8.

Jo, H.Y., Benson, C.H., Edil, T.B. 2004 Hydraulic conductivity and cation exchange in non-prehydrated and prehydrated bentonite permeated with weak inorganic salt solutions. *Clays and Clay Minerals*, **52**, 661-679.

Karunaratne, G.P., Chew, S.H., Lee, S.L., Sinha, A.N. 2001 Bentonite:kaolinite clay liner. *Geosynthetics International*, **8**, 113-133.

Kaya, A., Fang, H.Y. 2005 Experimental evidence of reduction in attractive and repulsive forces between clay particles permeated with organic liquids. *Canadian Geotechnical Journal*, **42**, 632-640, DOI: 10.1139/t04-099.

Kayabal, K., Bulus, G. 2000 The usability of bottom ash as an engineering material when amended with different matrices. *Engineering Geology*, **56**, 293–303.

Khosravi, E., Ghasemzadeh, H., Sabour, M.R., Yazdani, H. 2013 Geotechnical properties of gas oil-contaminated kaolinite. *Engineering Geology*, **166**, 11-16.

Kolay, P.L., Ramesh, K. C. 2016. Reduction of Expansive Index, Swelling and Compression Behavior of Kaolinite and Bentonite Clay with Sand and Class C Fly Ash. *Geotechnical and Geological Engineering*, **34**, 87–101, DOI 10.1007/s10706-015-9930-4.

Kumar, G. V., Muir Wood, D. 1999. Fall cone and compression tests on clay–gravel mixtures. *Geotechnique*, **49**, 727–739.

Kumar Pal, S., Ghosh, A. 2013 Hydraulic conductivity of Fly Ash–montmorillonite clay mixtures. *Indian Geotechnical Journal*, 2013, 43, 47–61.

Lemos, L. J. L., Vaughan, P. R. 2000. Clay-interface shear resistance. *Geotechnique,* **20**, 55–64.

Littlelton, I. 1976 An experimental study of the adhesion between clay and steel. *Journal of Terramechanics*, **13**, 141-152.

Lu, N., Dong, Y. 2017. Correlation between soil-shrinkage curve and water-retention characteristics. *Journal of Geotechnical and Geoenvironmental Engineering*, 143, https://doi.org/10.1061/(ASCE)GT.1943-5606.0001741.

Meegoda, N.J., Ratnaweera, P. 1994 Compressibility of fine-grained soils. *Geotechnical Testing Journal*, **17**, 101-112.

Mesri, G., Olson, R.E. 1971 Mechanics controlling the permeability of clays. *Clays and Clay Minerals*, **19**, 151-158.

Mesri, G., Cepeda-Diaz, A. F. 1986 Residual shear strength of clays and shales. *Geotechnique*, **36**, 269–274

Mir, B.A., Sridharan, A. 2014 Volume change behavior of clayey soil–fly ash mixtures. *International Journal of Geotechnical Engineering*, **8**, 72-83.

Mishra, A.K., Ohtsubo, M., Li, L., Higashi, T. 2005 Effect of salt concentrations on the permeability and compressibility of soil-bentonite mixtures. *Journal Faculty of Agriculture Kyushu University*, 50, 837-849.

Mitchell, J. K., Soga, K. 2005 *Fundamentals of Soil Behavior*. 3rd ed., John Wiley. New York.

Nagaraj, T. S., Jayadeva, M. S. 1983. Critical reappraisal of plasticity index of soils. *Journal of Geotechnical Engineering ASCE*, **109**, 994–1000.

Olson, R.E., Mesri, G., 1970, "Mechanism controlling compressibility of clays," J. Soil. Mech. Found. Div. ASCE, Vol. 96, pp. 1863-1878.

Panadian, N. S., Nagaraj, T. S. 1990 Critical reappraisal of colloidal activity of clays. *Journal of Geotechnical Engineering ASCE*, **116**. 285–296.

Park, J., Vipulanandan, C., Kim, J.W., Oh, M.H. 2006. Effects of surfactants and electrolyte solutions on the properties of soil. *Environmental Geology*, **49**, 977–989.

Petrov, R.J., Rowe, R.K., Quigley, R.M. 1997 Selected factors influencing GCL hydraulic conductivity. *Journal of Geotechnical and Geoenvironmental Engineering*, **123**, 683-695.

Polidori, E. 2003. A new plasticity chart. *Geotechnique*, **53**, 397-406.

Polidori, E. 2007 Relationship between the atterberg limits and clay content. *Soils and Foundations*, **47**, 887-896. DOI: 10.3208/sandf.47.887.

Prakash, K., Sridharan, A. 2013. Permeability of layered soils: an extended study. *Geotechnical and Geological Engineering*, **31**, 1639–1644.

Pulat, H.F., Yukselen-Aksoy, Y., Egeli, I. 2014 The effect of soil mineralogy and pore fluid chemistry on the suction and swelling behavior of soils. *Bulletin of Engineering Geology and Hydrogeology*, **73**, 37–42, DOI 10.1007/s10064-013-0499-y.

Rageena S.S., Rani V. 2015 Effect of various pore fluids on free swell and shrinkage cracking of clays. *International Journal of Innovative Research in Science, Engineering and Technology*, **4**, 5277-5282.

Rao, S. M., Sridharan, A. 1985 Mechanism controlling the volume change behavior of kaolinite. *Clays and Clay Minerals*, **33**, 323-328

Rao, S. M., Sridharan, A., Chandrakaran, S. 1989 Influence of drying on the liquid limit behaviour of a marine clay. *Geotechnique*, **39**, 715–719.

Robinson, R.G., Allam, M.M. 1998 Effect of clay mineralogy on coefficient of consolidation. *Clays and Clay Minerals*, **46**, 596-600.

Samarasinghe, A.M., Huang, Y.H., Drnevich, V.R. 1982 Permeability and consolidation of normally consolidated soils. *Journal of the Geotechnical Engineering, ASCE*, **108**, 835-850.

Sachan, A., Vikash, G., Prashant, A. 2013. Development of intermediate microfabric in kaolin clay and its consolidation behavior. *Geotechnical and Geological Engineering*, **31**, 23–34.

Seed, H.B., Woodward, R.J., Lundgren R. 1964 Clay mineralogical aspects of the Atterberg Limits. *Journal of the Soil Mechanics and Foundations Division*, ASCE, **90**, 107-131.

Sentenac, P., Taylor, R.N., Lynch, R.J., Bolton, M.D. 2006 Butanol effect on consolidated clay. *International Journal of Physical Modelling in Geotechnics*, **6**, 19 –27, DOI: 10.1680/ijpmg.2006.060402.

Shariatmadari, N., Salami, M., Fard, M.K. 2011 Effect of inorganic salt solutions on some geotechnical properties of soil-bentonite mixtures as barriers. *International Journal of Civil Engineering*, **9**, 103-110.

Sivapullaiah, P. V., Sridharan, A. 1985 Liquid limit of soil mixtures. *Geotechnical Testing Journal*, **8**, 111–116.

Sivapullaiah, P. V., Sitharam, T.G., Subbarao, K.S. 1987 Clay-organic molecule interaction: consolidation behaviour. *Clay Research*, **6**, 76-80. 1987.

Sivapullaiah, P.V., Lakshmi Kantha, H., Madhu Kiran, K. 2003. Geotechnical properties of stabilised Indian red earth. *Geotechnical and Geological Engineering*, **21**, 399–413.

Sivapullaiah, P.V. & Lakshmikantha, H. 2004 Properties of Fly Ash as hydraulic barrier. *Soil and Sediment Contamination*, **13**, 489-504.

Spagnoli, G., Fernandez-Steeger, Feinendegen, M., Stanjek, H., Azzam R. 2010 The influence of the dielectric constant and electrolyte concentration of the pore fluids on the undrained shear strength of smectite. *Soils and Foundations*, **50**, 757-763. http://doi.org/10.3208/sandf.50.757.

Spagnoli. G. Stanjek. H., Sridharan. A. 2012 Influence of ethanol/water mixture on the undrained shear strength of pure clays. *Bulletin of Engineering Geology and the Environment*, **71**, 389-398. DOI: 10.1007/s10064-011-0393-4.

Spagnoli G. 2012 Comparison between Casagrande and drop-cone methods to calculate liquid limit for pure clay. *Canadian Journal of Soil Science*, **92**, 859-864, 10.4141/cjss2012-011.

Spagnoli G, Sridharan A, Oreste P, Di Matteo L. 2017. A probabilistic approach for the assessment of the influence of the dielectric constant of pore fluids on the liquid limit of smectite and kaolinite. Applied Clay Science, 145, 37-43, https://doi.org/10.1016/j.clay.2017.05.009.

Sridharan. A. 2014 Fourth IGS-Ferroco Terzaghi Oration: 2014: Soil clay mineralogy and physico-chemical mechanisms governing the fine-grained soil behavior. *Indian Geotechnical Journal*, **44**, 371-399. DOI: 10.1007/s40098-014-0136-0.

Sridharan, A., Venkatappa Rao. G. 1975 Mechanism controlling the liquid limit of clays. Proceedings of Istanbul Conference on Soil Mechanics and Foundation Engineering, 1. 75-84.

Sridharan, A., Prakash. K. 1998 Characteristic water contents of fine grained soil–water system. *Geotechnique*, **48**, 337–346

Sridharan, A., Venkatappa Rao, G., Samudra Pandian, R. 1973 Volume change behaviour of partly saturated clays during soaking and the role of effective stress concept. *Soils and Foundations*, **13**, 1-15.

Sridaran, A., Rao, S.M., Murthy, N.S. 1986a Compressibility behaviour of homoionized bentonites. *Géotechnique*, **36**, 551-564.

Sridharan, A., Sudhakar. M.R., Murthy. N.S. 1986b Liquid limit of montmorillonite soils. *Geotechnical Testing Journal*, **9**, 156-159.

Sridharan, A., Rao. S.M. & Murthy, N.S. 1988 Liquid limit of kaolinitic soils. *Geotechnique,* **38**, 191-198.

Sridharan, A., Prashanth, J.P., Sivapullaiah, P.V. 1997 Effect of fly ash on the unconfined compressive strength of black cotton soil. *Proceedings of the Institution of Civil Engineers-Ground Improvement*, **1**, 169-175.

Sridharan, A., Nagaraj, H. B. 1999 Absorption water content and liquid limit of soils. *Geotechnical Testing Journal*, **22**, 121-127.

Sridharan, A., Nagaraj, H. B. 2005. Hydraulic conductivity of remolded fine-grained soils versus index properties. *Geotechnical and Geological Engineering*, **23**, 43–60.

Sridharan, A., Prakash, K. 2001 Consolidation and permeability behavior of segregated and homogeneous sediments. *Geotechnical Testing Journal*, **24**, 109–120

Stavridakis, E.I. 1999. Influence of Liquid Limit and Slaking on Cement Stabilized Clayey Admixtures. *Geotechnical and Geological Engineering*, **17**, 145–154.

Stadtbäumer, F.J. 1976. The influence of inorganic salts on some soil mechanical parameters of clays. *Bulletin of Engineering Geology and the Environment*, **13**, 65-69.

Taylor, R.K., Smith, T.J., 1986, The engineering geology of clay minerals: swelling, shrinking and mudrock breakdown. *Clay Minerals*, **21**, 235-260.

Tiwari, B., Ajmera, B. 2014, "Effects of saline fluid on compressibility of clay minerals," *Environmental Geotechnics*, Vol. **1**, No. 2, pp. 108-120.

Tiwari, B., Ajmera, B. 2014 Effects of saline fluid on compressibility of clay minerals. *Environmental Geotechnics*, **1**, 108-120.

Tuncan, A., Tuncan, M., Koyuncu, H., Guney Y. 2003. Use of natural zeolites as a landfill liner. *Waste Management & Research*, **21**, 54–61

Wasti, Y., Bezirci, M.H. 1986 Determination of the consistency limits of soils by fall cone test. *Canadian Geotechnical Journal*, **23**, 241–246.

White, W.A. 1949 Atterberg plastic limits of clay minerals. *American Mineralogist*, **34**, 508-512.

Yong, R.N., Warkentin, B.P. 1975 Soil Properties and Behaviour. Elsevier Scientific Publishing Company.

Younus, M. M., Sreedeep, S. 2012 Evaluation of bentonite-Fly Ash mix for its application in landfill liners. *Journal of Testing and Evaluation*, **40**, 357-362, https://doi.org/10.1520/JTE104161.

FIGURE CAPTIONS

Fig. 1 PI vs LL for smectitic clays.

Fig. 2 PI vs LL for kaolinitic clays

Fig. 3 Predicted PI vs experimental PI for smectitic clays (A); predicted PI vs experimental PI for kaolinitic clays (B). The experimental PI values refer to Tab. 3.

Fig. 4 Correlation PL vs LL for smectitic (A) and kaolinitic clays (B). The correlating lines are dotted because they are not meant to give a statistical reference. Note that the legend is the same as per Figs. 1 and 2.