

Micro to macro investigation of clays advising their constitutive modelling – part II

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Article

Keywords

Microstructure
Constitutive modelling
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Abstract

This paper reports the second part of the keynote lecture, whose part I has been already published in this journal, presenting extensive experimental research on the investigation of clay microstructure and its evolution upon loading. Whether the first part focused on the micro to macro behaviour of different reconstituted clays, this part instead concerns the microscale features of the corresponding natural clays, their changes under different loading paths and the ensuing constitutive modelling implications. The experimental investigation is carried out according to the methodology outlined in the part I-paper, hence micro-scale analyses are presented on natural clays subjected to macro-scale mechanical testing, with the purpose to provide experimental evidence of the processes at the micro-scale which underlie the clay response at the macro-scale. As for the reconstituted clays in the part I-paper, original results on stiff Pappadai and Lucera clay, this time in their natural state, are compared to literature results on clays of different classes, either soft or stiff. The results presented in this paper, together with those discussed in the part I, allow for a conceptual modelling of the microstructure evolution under compression of natural versus reconstituted multi-mineral clays, providing microstructural insights into the macro-behaviour described by constitutive laws and advising their mathematical formalization in the framework of either continuum mechanics or micro-mechanics.

1. Introduction

This paper reports the second part, II, of the keynote lecture whose part I has been published in this journal by Cotecchia et al. (2024) focused on the micro to macro behaviour of reconstituted clays. This paper, instead, is concerned with the microscale features of different natural clays, their changes under compression and the corresponding constitutive modelling implications, all investigated through the same methodology outlined in the part I-paper. Hence, the reader is referred to the part I-paper for the research objectives and methods of the whole research work, presented in both the part I-paper and this part II-paper.

Among natural clays, the microstructural features vary not only due to differences in composition and stress history, as outlined for reconstituted clays in the part I-paper. Natural clays' microstructural features depend also on their original natural deposition conditions and on many possible chemo-hydro-mechanical processes taking place over the geological history (e.g. creep, thixotropy, calcite deposition from water supersaturated with calcium carbonate, diagenesis,

weathering, drying-wetting cycles, etc.; e.g. O'Brien & Slatt (1990), Mitchell & Soga (2005), Cotecchia & Chandler (1995, 2000), Cafaro & Cotecchia (2001)).

According to the methodological approach described in the part I-paper (see Figure 1 in the part I-paper), the microstructures of different natural clays and their variations under compression are discussed in the following, in order to shed light on the micro-scale processes that are the source of variable macro-scale responses among natural clays. The natural clays of reference are those whose reconstituted version has been the subject of the part I-paper.

2. Clays under study

2.1 Composition, deposition conditions and processes impacting the microstructure

Table 1 reports the index properties (discussed in the companion paper, see Figure 4 of the part I-paper) and in situ states of the natural clays under study. For some of these, the

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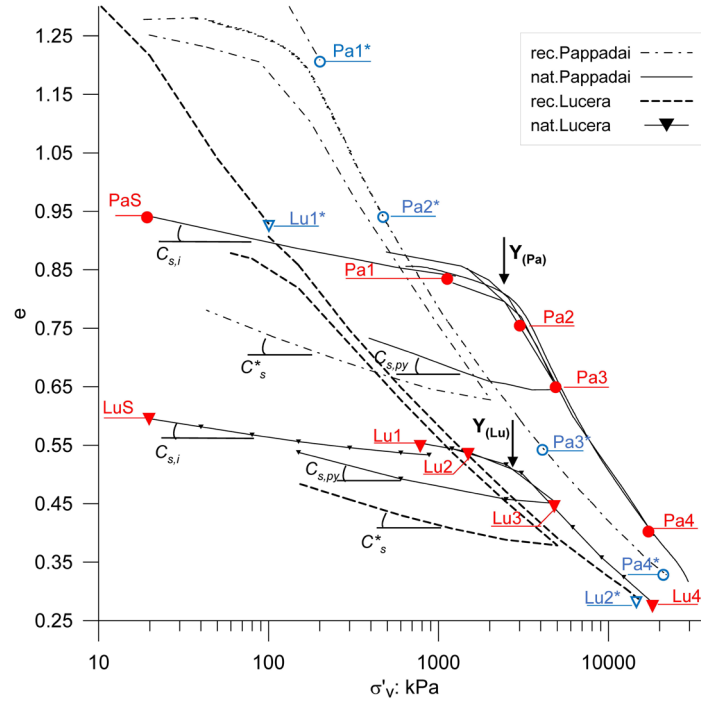


Figure 1. 1D behaviour of Lucera and Pappadai clay in the $[e; \sigma'_v]$ plane. Y = gross yield. States of microstructural analysis discussed in the paper are highlighted for both the natural and reconstituted clay [modified after Guglielmi et al. (2022)].

yield stress ratio, YSR (Burland, 1990), values exceed the geological overconsolidation ratio, OCR , due to geological processes which occurred under burial and affected the clay microstructure (change from sedimentation to post-sedimentation structure according to Cotecchia & Chandler (2000)). The table reports also the clay stress sensitivity¹, S_σ (Cotecchia & Chandler, 2000), and the ‘strength sensitivity’ S_t (Terzaghi, 1944; Skempton & Northey, 1952; Cotecchia & Chandler, 2000; here computed as ratio of the clay undrained strength for $YSR = 1$, c_{uNC} to that of the reconstituted clay² at the same void ratio, c_{uNC}^* , $S_t = c_{uNC} / c_{uNC}^*$).

The deposition environment of the clays and the processes experienced after deposition are synthesized in the following.

2.1.1 Soft clays

St Marcel, Ballina, Bothkennar and Gulf of Guinea clays are soft clays; in particular, both St Marcel and Ballina clay are slightly to medium quick clays [S_t in the range 10-20; Mitchell & Soga (2005)].

St Marcel clay is a Champlain Sea clay from Quebec, whose sensitivity was induced by the leaching action generated by brackish to fresh water, which replaced the original saline

pore water (Quigley, 1980; Delage & Lefebvre, 1984). As typical for Champlain clays, a non-negligible proportion of its clay fraction is not made of phyllosilicates, being composed of primary minerals, such as quartz, feldspar, amphibole, mica and chlorite (Quigley, 1980). These minerals are ground down to very small sizes by glaciers to form rock flour (Quigley, 1980; Locat et al., 1984; Delage, 2010). The clay mineralogy (Table 1) confers a low activity index (part I-paper). Scanning Electron Microscopy, SEM, and Mercury Intrusion Porosimetry, MIP, investigations were carried out by Delage & Lefebvre (1984) on St Marcel clay block samples from 5.2 m depth, soon after sampling and 1D compression.

Ballina clay originated, in the Holocene, in the Pimplico clay deposit (east Australian coastline), in a low-energy estuarine environment (Bishop & Fityus, 2006; Pineda et al., 2016b). Compared to St Marcel clay, Ballina clay is of far higher plasticity and activity index (see Table 1) due to a higher content of illite and mixed illite/smectite layers. Pineda et al. (2016b) carried out microstructural analyses on a natural undisturbed sample (Sherbrooke sampler) from 7m depth (Table 1).

Bothkennar clay (Scotland) is a mainly illitic Holocene medium sensitive clay (Table 1), of high plasticity and activity (Table 1; Cotecchia et al. (2024) – part I). It deposited in a sheltered environment of brackish water, allowing the setting of a flocculated fabric (Hight et al., 1992). In addition, post-

¹i.e. the ratio of the natural clay gross yield pressure in compression to the equivalent pressure on the NCL of the reconstituted clay.

²Reconstituted clay states will be denoted with * in the following.

Table 1. Properties of the clays discussed in the paper [modified after Guglielmi et al. (2022)].

Clay	z (m)	w ₀ (%)	e ₀ (-)	σ'_{v0} (kPa)	CF (%)	LL (%)	PI (%)	A (-)	mineralogy*	YSR	S _p , S _σ
<i>soft clays</i>											
BOTHKENNAR (Bot) Hight et al. (1992), Smith et al. (1992), Tanaka et al. (2003)	17	70	1.61	≈100	28-40	55-86	40-50	1.3	I; K; Qz; Fd †	1.4-1.6	4-7
GULF OF GUINEA (GoG) Hattab et al. (2013)	10	130-170	3.28	36.5	40-60	110-160	70-80	1.5	K: 50; S: 15-25 †	1.3-1.4	6-8
ST MARCEL (StM) Delage & Lefebvre (1984)	5.2	80	2.3	<10	80	60	35	0.4	Ch: 4-16; I: 2-10; V: 2-9 ‡	-	19
BALLINA (Ba) Pineda et al. (2016a, b)	7.4-7.9	110-120	2.95-2.97	<50	78-82	115-130	70-80	1	K: 2,3-24,5; I: 7,8-23,3; Qz: 8,8-55,9; I/S: 9,5-20,1; amorphous minerals: 1,7-22,4; Pl: 2,9-15,2; Py: 0-6,4; K-Fd: 1,2-5,5; Mca: 0,9-2,1; Ca: 0-1 ‡	<2	10-20
<i>stiff clays</i>											
PAPPADAI (Pa) Cotecchia & Chandler (1997, 2000), Cotecchia et al. (2019), Guglielmi (2018)	25	30	0.88	414	58	65	35	0.6	I: 20; Ch: 14; S: 12; K: 12; I/S: 10 ‡	6	3.5
LUCERA (Lu) Mitaritonna (2006), Guglielmi (2018)	40	18	0.55	≈800	48	48	24	0.5	I/S: 77 (40-45); K: 9; I: 8; Ch: 6 †	3.7	2
BOOM (Bo) Belanteur et al. (1997), Delage et al. (2007), Lima et al. (2008), Della Vecchia et al. (2011)	223	22.6-24	0.58-0.61	2250	50-60	67	38	0.7	I: 50; S: 30; I/S+K: 10 †	2.3-2.4	2(?)
LONDON Gasparre et al. (2007)	1-45	21-29	0.6-0.8		41-63	59-78	33-62	0.7-0.8	I: 21-38; K: 11-15; Ch; S; M ‡	3-20	1.3-2.9
BROWN WEATHERED LONDON (BWL) Skempton & Jones (1944), Monroy et al. (2010)	4-6	-	1.175		58	83	54	0.93	-	-	-

Keys: z, sample depth; w₀, initial water content; e₀, initial void ratio; σ'_{v0} , in situ vertical effective stress; CF, clay fraction; LL, liquid limit; PI, plasticity index; A, activity index; YSR, Yield Stress Ratio; S_p, strength sensitivity, or S_σ, stress sensitivity (in italics). *Percentages are shown, when available, otherwise the minerals are listed in descending order of dominance: I, illite; K, kaolinite; Ch, chlorite; S, smectite; I/S, mixed-layers illite/smectite (illite % within parentheses); V, vermiculite; M, montmorillonite; Qz, quartz; Fd, feldspar; Ca, carbonate; Pl, plagioclase; Py, pyrite; Mca, mica. † % of the fraction ≤ 2 μm. ‡ % of the bulk sample.

depositional bonding, ageing and leaching have impacted its microstructure (Paul et al., 1992; Hight et al., 1992; Tanaka, 2000; Tanaka et al., 2003). Tanaka et al. (2003) carried out MIP tests on the undisturbed clay sampled at 17 m depth (Japanese fixed piston sampler).

Gulf of Guinea clay (GoG) is a deep-water marine clay, sampled by means of a 10 m long sampler below the sea floor of the Gulf of Guinea, under 700 m sea water column, in an oil-producing area (Hattab et al., 2013). The clay is of very

high plasticity and has the highest activity index among the soft clays of reference (part I-paper), due to its considerable smectite content. At the microscale, it embodies a physico-chemical bonding generated during the sedimentation and consolidation process in the seabed (De Gennaro et al., 2005; Hattab & Favre, 2010; Hattab et al., 2013). Hattab et al. (2013) analysed the microstructure of the undisturbed clay and after both 1D and isotropic compression to $\sigma'_v = 1000$ kPa and $p' = 900$ kPa, respectively.

2.1.2 Stiff clays

Both Pappadai and Lucera clay are stiff clays part of the Pleistocene Subapennine Blue clays (Del Prete & Valentini, 1971; Cotecchia & Chandler, 1995), illitic and of medium-high plasticity and low activity (see Table 1). The deposition environment of Pappadai clay was a protected still water sea, where the clay overlaid a calcarenite bedrock (Ciaranfi et al., 1971) in a chemically reducing environment, allowing for flocculation and lamination (O'Brien & Slatt, 1990; Cotecchia & Chandler, 1995). The deposition of Lucera clay, instead, occurred in a marine environment of higher energy than for Pappadai clay, in a peripheral margin of the Apennine foredeep (Gallicchio et al., 2003; Santaloia et al., 2004).

Both clays were buried under more than one hundred metres of sediments, overconsolidated due to erosion and subjected to diagenesis, which strengthened the clay bonding, as confirmed by mineralogical evidence (Cotecchia & Chandler, 1995) and chemical micro-probing in the SEM (Cotecchia & Chandler, 1997; Guglielmi et al., 2018). Microstructural analyses have been performed on both clays, at different levels of compression (Table 1).

For comparison, the microstructural features of stiff Boom clay (Lima et al., 2008), a mainly illitic marine Tertiary clay, of high plasticity and medium-low activity (Table 1), block sampled at more than 200 m depth (HADES underground laboratory, Mol - Belgium), will be also discussed. For all these stiff clays, S_g is 2-3 (Table 1).

2.2 Clay states and compression macro-behaviour

Figure 1 shows the results of 1D compression and swelling tests on natural and reconstituted Pappadai and Lucera clay specimens.

Both stiff clays exhibit a stiff response to swelling, with high swell sensitivity C_s^*/C_{si} (2.5 for Pappadai clay and 2 for Lucera clay), as already observed by Cotecchia & Chandler (1997) and Cafaro & Cotecchia (2001), which is indicative of a significant strength of the natural bonding developed through diagenesis during ageing under burial (Cotecchia & Chandler, 1997). Such bonding provides both clays with $YSR > OCR$ (e.g. for Pappadai clay, $YSR = 6$ twice the OCR) and $S_g > 1$, although much lower than for soft clays. The compression post-gross yield (Figure 1) is seen to cause a rather immediate C_s^*/C_s decrease, indicative of a rather fast weakening of bonding upon gross yielding, and a progressive reduction in S_g . The micro-analyses for states Pa1*, Pa4*, Pa1 and Pa4 in Figure 1 have been already referred to in the companion paper (see §3 in the part I-paper) and in Cotecchia et al. (2019) and will be recalled later.

Figure 2 illustrates compression data already plotted in Figure 1 along with data resulting from laboratory 1D compression tests on either natural, or reconstituted samples of several of the other clays in Table 1. The compression data are normalized for composition using the void index $I_v = (e -$

$e_{100}^*)/(e_{100}^* - e_{1000}^*)$ (Burland, 1990), where e_{100}^* and e_{1000}^* are the void ratios on the intrinsic compression line, ICL, for σ_v' equal to 100 kPa and 1000 kPa, respectively. The states corresponding to the microstructural analyses (SEM and/or MIP) are indicated with large symbols (full symbols correspond to natural samples and empty symbols to the reconstituted ones).

The natural samples of soft St Marcel, Ballina, Bothkennar and GoG clay, normally consolidated to lightly overconsolidated in situ, exhibit $YSR < 2$ (Table 1; Delage & Lefebvre, 1984; Pineda et al., 2016a; Hight et al., 1992; Smith et al., 1992; Hattab et al., 2013) and S_g higher than for stiff overconsolidated Pappadai, Lucera and Boom clay. Evidently, the bonding they achieved in the geological history, although much weaker than for the stiff diagenized clays, has allowed them to keep far more open fabrics than the reconstituted clay since early stages of compression. However, the large compressibility of these clays post-gross yield [e.g. Locat & Lefebvre (1985), Leroueil & Vaughan (1990), Delage (2010)] indicates a fragile weakening of such bonding upon gross yielding. It is worth noting that for Bothkennar clay, the 1D compression curve (Bot b) in Figure 2 refers to a sample (Smith et al., 1992) different from that subjected to microstructural analysis [Bot a, Tanaka et al. (2003)].

While the post-mortem micro-analyses on the specimens indicated in Figures 1 and 2 allow for insight into the evolution of microstructure solely due to 1D compression, the microstructural changes activated under different constant $q/p' = \eta$ compressions have been investigated for both the natural Lucera and Pappadai clays. Figure 3 reports the comparison, in the $[v; \log p']$ plane, between the 1D and the isotropic compression curve of the two clays.

Pre-gross yield, the straight-line gradient κ ($\kappa = 0.015$ for Lucera clay and $\kappa = 0.02$ for Pappadai clay) is lower than the corresponding κ^* . For both stiff clays, κ^*/κ , as for C_s^*/C_s , reduces to unity post-gross-yield, confirming that bonding weakens also with gross yield in isotropic compression (Cotecchia et al., 2016, 2019, 2020). In isotropic compression, Lucera clay (Figure 3a) and Pappadai clay (Figure 3b) exhibit λ values of 0.141 (Guglielmi, 2018) and 0.254 (Cotecchia & Chandler, 1997) respectively, both close to the λ value in 1D compression (as expected according to critical state soil mechanics) and exceeding λ^* (0.139 for reconstituted Lucera clay and 0.204 for reconstituted Pappadai clay). Figure 3 indicates the states for which post-mortem micro-analyses have been conducted.

3. Natural clay microstructure under compression

3.1 Soft clays under 1D compression

Figure 4 shows the SEM micrographs for states StM1 and Ba (Figure 2) of the highly sensitive St Marcel and Ballina clay (Table 1), which were reported by Delage

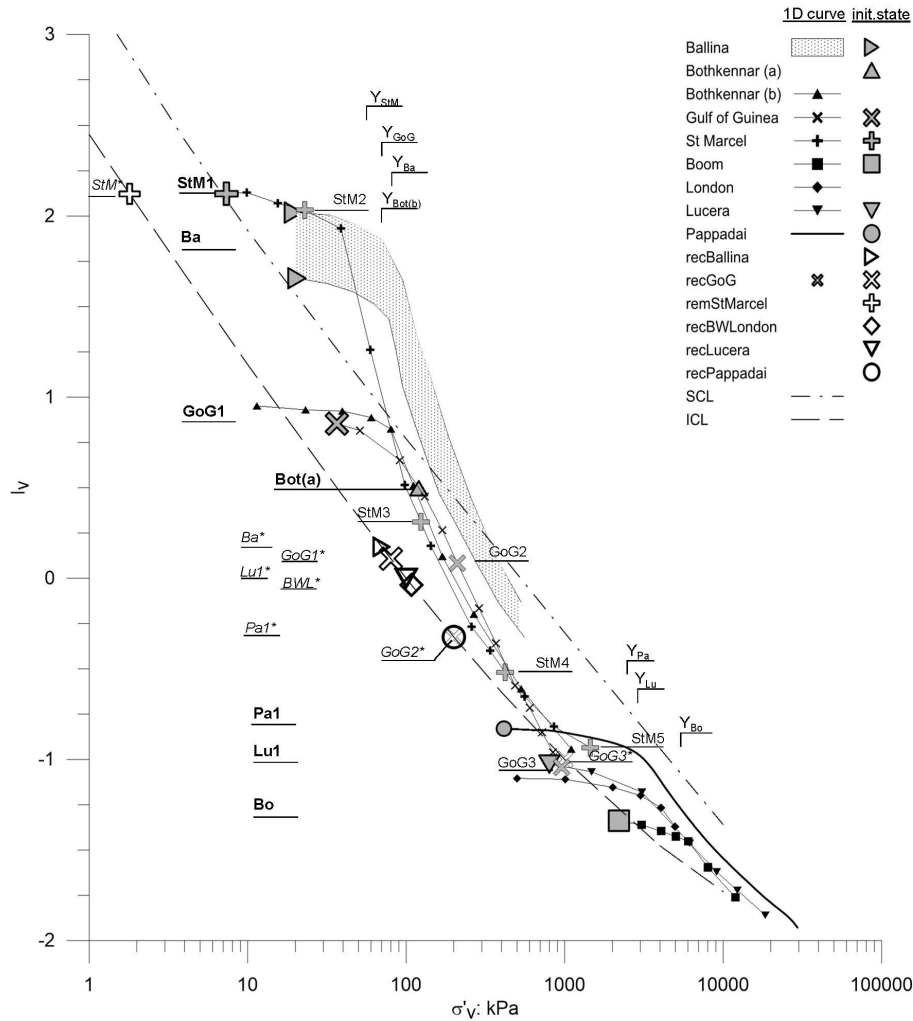


Figure 2. One-dimensional compression of natural and reconstituted clays normalized for composition using the void index, I_v [data after Bishop et al. (1965), Burland (1990), Cotecchia & Chandler (2000), Delage & Lefebvre (1984), Hattab et al. (2013), Smith (1992), Pineda et al. (2016a, b)]. Y = gross-yield; ICL = Intrinsic Compression Line; SCL = Sedimentation Compression Line. Larger symbols (empty for reconstituted, full grey for natural) represent states of samples on which microstructural analyses were carried out; the largest symbols correspond to the sample initial states [modified after Guglielmi et al. (2022)].

& Lefebvre (1984) and Pineda et al. (2016a), respectively. The micrographs in Figure 4a and 4c correspond to the micro-REV scale (Cotecchia et al., 2019), and those in Figure 4b and 4d to larger scales. At the micro-REV scale, aggregates of random orientation particles appear to be connected by bridges formed of domains and to confine inter-aggregate pores for both the less plastic St Marcel clay and the more plastic Ballina clay. A similar fabric was observed for the reconstituted more plastic clays, Ballina and GoG (ss sketched in Figure 7f in the part I-paper).

Despite the higher void ratio of Ballina clay with respect to St Marcel clay, both clays appear to contain a significant inter-aggregate porosity (Figure 4a, 4c), but Ballina clay includes also an important intra-aggregate porosity (Figure 4d), formed of pores smaller than for the intra-aggregate porosity of St Marcel clay (Figure 4b). Such evidence is consistent

with the more clastic nature and lower activity of the clay fraction of St Marcel clay. The more active CF of Ballina clay is largely made up of small active clay particles which can form aggregates that include small pores. Similar fabric features are recognizable in the micrographs of the slightly less sensitive natural GoG clay samples (state GoG1 in Figure 2) described by Hattab et al. (2013). Overall, the fabric features observed through SEM in these natural soft clays are qualitatively similar to those observed for the lightly consolidated reconstituted clays, discussed in part I-paper (see Figure 7 in the part I-paper).

Figure 5 compares the MIP curves of the clay samples StM1, Ba, GoG1 and Bot(a) (Figure 2), which are all either normally consolidated clays, or slightly overconsolidated. A bimodal porosity is observed for Ba, GoG and Bot(a), with inter-aggregate dominant pore size, DPS, about 1000

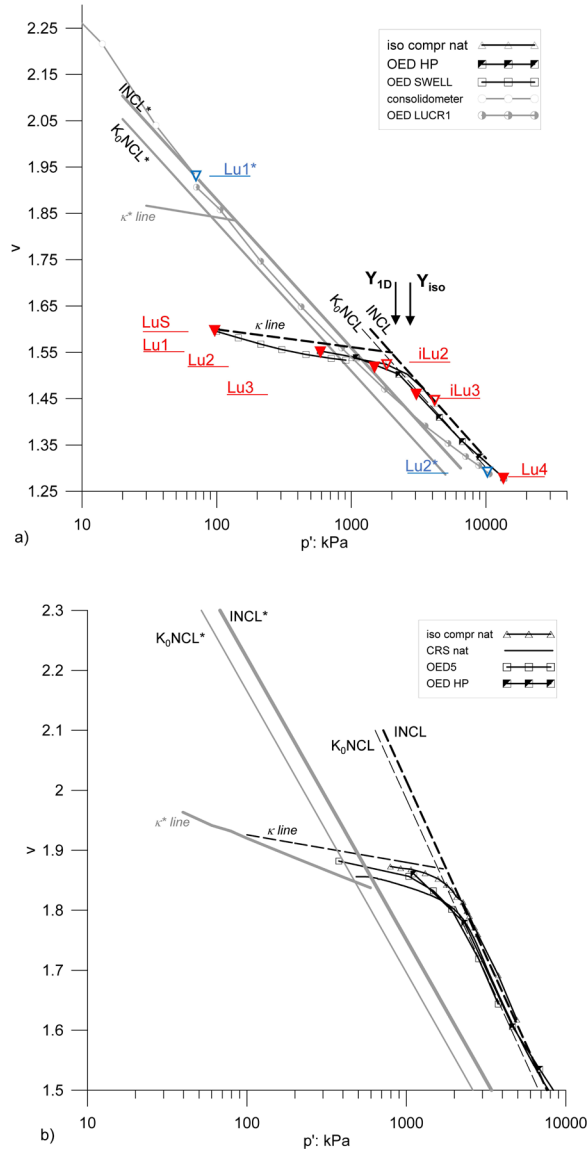


Figure 3. Isotropic and 1D behaviour of Lucera (a) and Pappadai (b) clay. Y = gross yield; $INCL$ = Isotropic Normal Compression Line; $K_0NCL = K_0$ Normal Compression Line. States of microstructural analysis discussed in the paper are highlighted for both the natural (in red: Lu1 for the initial state; Lu2, Lu3 and Lu4 for the clay compressed 1D; LuS for the clay swelled 1D; iLu2, iLu3 and iLu4 for the clay compressed isotropically) and the reconstituted (in blue: Lu1* after consolidometer and Lu2* compressed 1D) clay.

nm for both Ba and GoG1, and of 1700 nm for Bot(a). Also, a smaller frequency DPS is found to correspond to intra-aggregate pore volume, about 63 nm for GoG1, 100 nm for Bot(a) and 50-70 nm for Ba.

Hence, the MIP data are consistent with the SEM pore observations [sizes of inter-aggregate pores indicated in Figure 4d for Ba; Pineda et al. (2016a)] and confirm that, in addition to the large inter-aggregate pores, significant porosity is made of small intra-aggregate pores within the

higher plasticity clays. This intra-aggregate porosity is much less for lower plasticity clays (e.g. St Marcel).

It follows that both SEM and MIP investigations show that the fabric features of soft natural clays, characterized by an activity from medium-low to high (Table 1) and originally deposited in a marine environment, are similar to those of the corresponding reconstituted clays (part I-paper), consolidated in the laboratory from slurries³. Such fabric features comply with the fabric sketch by Griffiths & Joshi (1990) (as sketched in Figure 7f in the part I-paper), which appears therefore suited to represent the fabric of such clays when at $I_v > 0$, whether natural or reconstituted (Figure 2). The fabric is flocculated and inter-aggregate macro-porosity is dominant, but also micro-porosity can occur within the aggregates. The latter corresponds to a secondary DPS whose size decreases with increasing activity index.

These findings then suggest that the difference in response to compression of natural and reconstituted clays of $I_v > 0$ (Figure 2; Burland, 1990; Cotecchia & Chandler, 2000) is more the result of a difference in their bonding features, than in their fabric features. Such difference in bonding allows the difference between the normal consolidation line of the reconstituted clay (ICL, Figure 2) and that of the natural clay, which lies to the right of the ICL, consistent with $S_\sigma > 1$.

Natural St Marcel clay was subjected to several 1D compression tests, stopping at σ'_v of 23, 124, 421 and 1452 kPa, i.e. states StM2, StM3, StM4, StM5 in Figure 2, respectively. Both SEM and MIP tests were carried out on each specimen after unloading to 4 kPa (Delage & Lefebvre, 1984). As the St Marcel clay gross-yield pressure is equal to 54 kPa (Y_{StM} , Figure 2), the MIP data for the first specimen, StM2 ($\sigma'_v = 23$ kPa), allow to check whether microstructural changes take place during compression pre-gross yield, when compared with the MIP data of the undisturbed specimen StM1 (Figure 6). As commented by Delage & Lefebvre (1984), neither the micrographs, nor the PSD curves in Figure 6 give evidence of any significant change in clay microstructure with respect to the initial one, suggesting that pre-gross yield compression does not cause significant modifications in microstructure, even for highly sensitive clays.

When compressed beyond gross-yield to state StM3 ($\sigma'_v = 124$ kPa; Figure 2), St Marcel clay experiences a reduction of the inter-aggregate porosity and an onset of fabric anisotropy [large pores and aggregates are stretched along the horizontal plane; Figure 7a, 7b; Delage & Lefebvre (1984)]. The initial undisturbed fabric arrangement (Figure 4a, b) is no longer recognizable at states StM4 ($\sigma'_v = 421$ kPa; Figure 7c, 7d) and StM5 ($\sigma'_v = 1452$ kPa; Figure 2), since the particles are densely packed and horizontally oriented (Figure 7e, 7f). The corresponding PSD curves are plotted in Figure 8a and show how post-gross yield compression determines a mono-modal PSD, with a single DPS, whose value and frequency

³Made using either tap water, or distilled water.

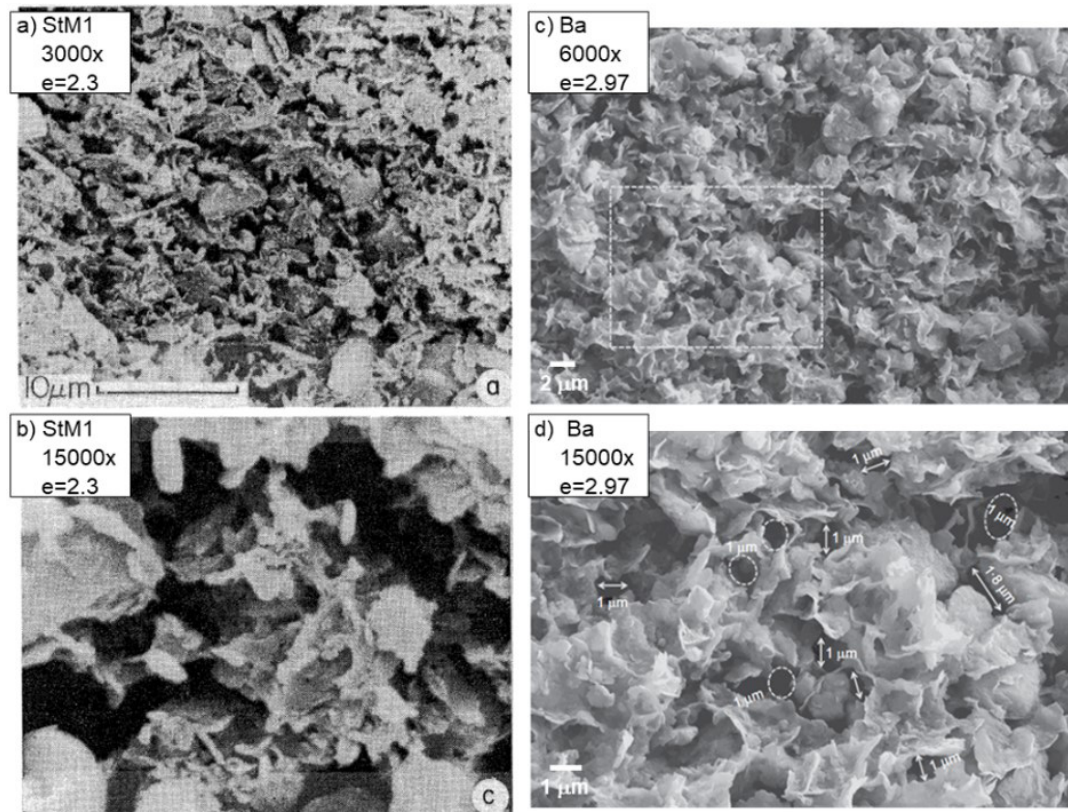


Figure 4. SEM micrographs of natural St Marcel clay (a, b; state StM1 in Figure 2) and natural Ballina clay (c, d; state Ba in Figure 2). **Sources:** Delage & Lefebvre (1984), Pineda et al. (2016b); modified after Guglielmi et al. (2022).

reduces due to the collapse of the inter-aggregate porosity. However, the intra-aggregate pore sizes are not significantly modified by compression up to $\sigma'_v = 1452$ kPa for this clay (Delage & Lefebvre, 1984).

Similar microstructural changes are observed for natural GoG clay, compressed from GoG1 state to GoG2, $\sigma'_v = 200$ kPa, and GoG3, $\sigma'_v = 1000$ kPa (Figure 2). Figure 8b shows a transition from a prevailing inter-aggregate porosity to a prevailing intra-aggregate porosity, with a complete loss of bimodality post-gross yield [at GoG3; Hattab et al. (2013)], even by a σ'_v increase smaller than that required for a similar process with reconstituted clay GoG* (see Figure 11a in the part I-paper).

Overall, the data suggest that 1D compression of clays, starting at $I_v > 0$, causes a transition from a large inter-aggregate porosity to a prevailing small intra-aggregate porosity, accompanied by an increase in fabric orientation. Both processes start only beyond gross-yield for natural soft clays, i.e. when bonding weakens. For the reconstituted clays, the fabric degree of orientation has been shown to reach a maximum constant value by I_v values little below 0 (part I-paper). Data are not available to check the reach, through 1D compression, of a constant fabric orientation degree, for the soft natural clays here of reference.

3.2 Stiff clays under 1D compression

According to the mineralogy (Table 1) and deposition conditions of natural Lucera clay (Lu1 in Figures 1 and 2) and Pappadai clay (Pa1), the original fabric of both these clays was most probably either honeycomb, or bookhouse (part I-paper), the same as that recognized for the soft clays discussed above. In their geological history, such fabric has been 1D compressed to significant pressures and unloaded (due to erosion – current $e_0 - \sigma'_{v0}$ states reported in Table 1); furthermore, diagenesis has impacted the clay microstructure under burial. For Pappadai clay, Cotecchia & Chandler (1995, 1997) report that diagenesis determined the growth of an amorphous calcite film binding the clay particles. Medium magnification micrographs of the current micro-REV fabric of both Lu1 and Pa1 (Figures 1 and 2) are shown in Figure 9.

Both Pa1 and Lu1 are found to embody a fabric (Figure 9a and 9c) formed of thick stacks of particles/domains, which are mostly sub-horizontal and densely packed. These bury local aggregates of randomly oriented domains. Further evidence of the fabric features of natural Pappadai and Lucera clay, detected through micrographs of magnification varying from medium to high, are provided by Cotecchia et al. (2019) [see Figure 5 in Cotecchia et al. (2019)] and Guglielmi (2018), respectively. According to such studies, the local

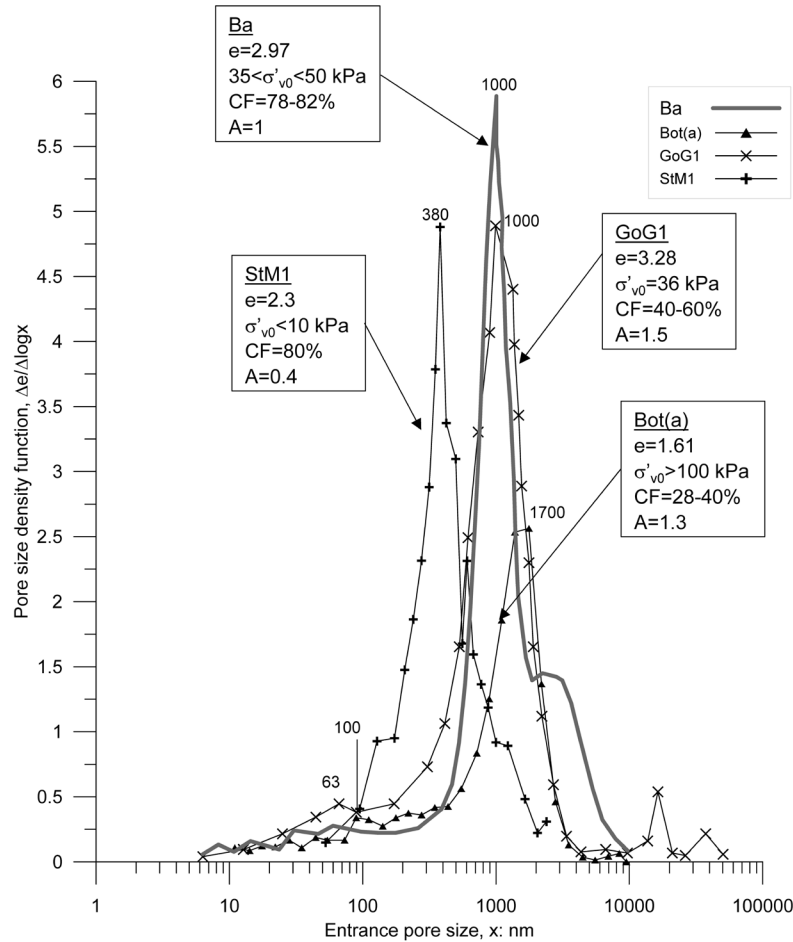


Figure 5. Comparison of pore size density, PSD, curves for soft St Marcel, Gulf of Guinea, Bothkennar and Ballina clay at undisturbed state (states StM1, GoG1, Bot(a) and Ba in Figure 2). Source: Delage & Lefebvre (1984), Hattab et al. (2013), Tanaka et al. (2003), Pineda et al. (2016b); modified after Guglielmi et al. (2022).

fabric of these stiff clays is highly variable, since the degree of local fabric orientation varies from very high to very low, the latter applying to relic portions of primary bookhouse fabric which did not undergo any significant increase in orientation during the geological pre-compression (Sfondrini, 1975). Nonetheless, the average degree of orientation index, L (see part I-paper), characterizing the micro-REV fabric observed through medium magnification micrographs, is high, corresponding to $L = 0.24-0.37$ for Pappadai clay and $0.22-0.29$ for Lucera clay (Figure 9b and 9d). Thus, both these stiff clays are found to embody a fabric qualitatively similar to that of reconstituted Pappadai clay 1D compressed to $I_v < 0$ (part I-paper).

The PSD curves of both stiff clays, Pa1 and Lu1, are shown in Figure 10, compared with that of Boom clay (Bo in Figure 2), reported by Lima et al. (2008). As for the PSD curves of reconstituted clay samples at $I_v < 0$ (Figure 2), i.e. BWL* and Pa1*, all the PSD curves in Figure 10 are monomodal and indicative of a pore volume mostly distributed in the micro-porosity range (pore size $< 1 \mu\text{m}$). For both

the high plasticity Boom and Pappadai clay (Table 1), the DPS (respectively 90 nm and 220 nm) is lower than for the lower plasticity and more silty Lucera clay (around 300 nm). The DPS of Boom clay is the lowest due to the far larger σ'_p and lower I_v (Figure 2).

Integrating the previous findings about the in-situ fabric of soft clays with those concerning the fabric of stiff natural clays, it appears that 1D compression of medium to high plasticity clays in the field, caused by the increase in burial depth, at early stages of compression ($I_v > 0$) allows for an either honeycomb or bookhouse fabric of bi-modal PSD. Further compression due to increase in burial depth brings about an increase in micro-REV fabric orientation, a loss of inter-aggregate porosity and a reduction in size of the monomodal DPS. The DPS of the natural stiff clays here of reference, of $I_v < 0$, is up to an order of magnitude smaller than that of the micro-porosity DPS of the soft clays of $I_v > 0$, and is of much lower frequency too.

Examples of pores of different size in stiff clays are shown in Figures 9 and 11. Where a relic macro-porosity,

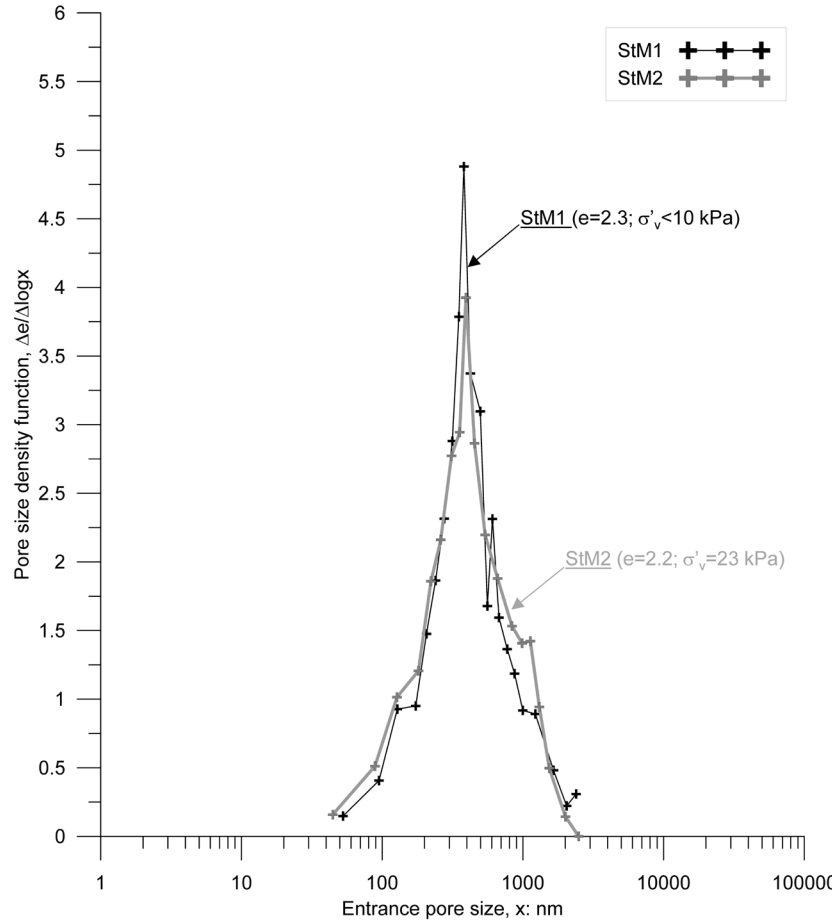


Figure 6. Comparison of PSD curves for pre-gross-yield states of St Marcel clay [states StM1 and StM2 in Figure 2, replotted and adapted after Delage & Lefebvre (1984)].

covering a wide range of entrance pore sizes, occurs here and there locally, most of the micro-pores of size about the DPS occur inside the densely packed stacks of particles.

The microstructure changes of both Pappadai and Lucera clay, induced by swelling from the undisturbed state to very low pressures ($\sigma'_v = 20$ kPa), have been investigated, along with their microstructure changes under compression, either before or beyond gross-yield (Figures 1 and 3). For the swelled natural Pappadai clay (PaS, Figure 1), SEM micrographs of medium magnification (e.g. Figure 12) show a very similar fabric to that of Pa1 (Figure 9a). Although long thick stacks of oriented particles are recognizable (Figure 12c), preferred orientation is not ubiquitous. The corresponding L values (Figure 12b, 12d) demonstrate that monotonic swelling does not change the original fabric orientation degree ($L = 0.24$ - 0.37). The PSD curve of the swelled specimen (Figure 13a) is still monomodal, with only a slight shift to the right of the DPS, for a slight increase of the intra-aggregate pores. Local widening of inter-aggregate pores is also observed, probably due to the opening of fissures.

Similarly, the PSD curve of natural Lucera clay, when either swelled to state LuS, or compressed pre-gross-yield, to $\sigma'_v = 1.4$ MPa (Lu2; Figure 1), are very similar to that of the undisturbed specimen (state Lu1), as shown in Figure 13b. Hence, the fabric and pore size distribution of either soft or stiff clays undergo no significant changes under monotonic swelling and compression pre-gross yield.

Figure 14a-14d shows examples of micrographs of natural Pappadai clay compressed beyond gross-yield, to states Pa3 and Pa4 ($\sigma'_v = 5$ MPa and $\sigma'_v = 18.5$ MPa; Figure 1), with the corresponding direction histograms and indices of fabric orientation. Even when the clay is compressed to very high pressures (state Pa4), the micro-REV fabric (Figure 14c) appears formed by thick layers of stacks interbedding randomly oriented fabric portions, which do not acquire an oriented fabric. Indices of average fabric orientation in the range $0.24 < L < 0.345$ are obtained, confirming that compression to high pressures does not determine any significant increase in the degree of micro-REV fabric orientation, with respect to the undisturbed fabric of the stiff natural clay occurring in the field, Pa1 ($L = 0.24$ - 0.37).

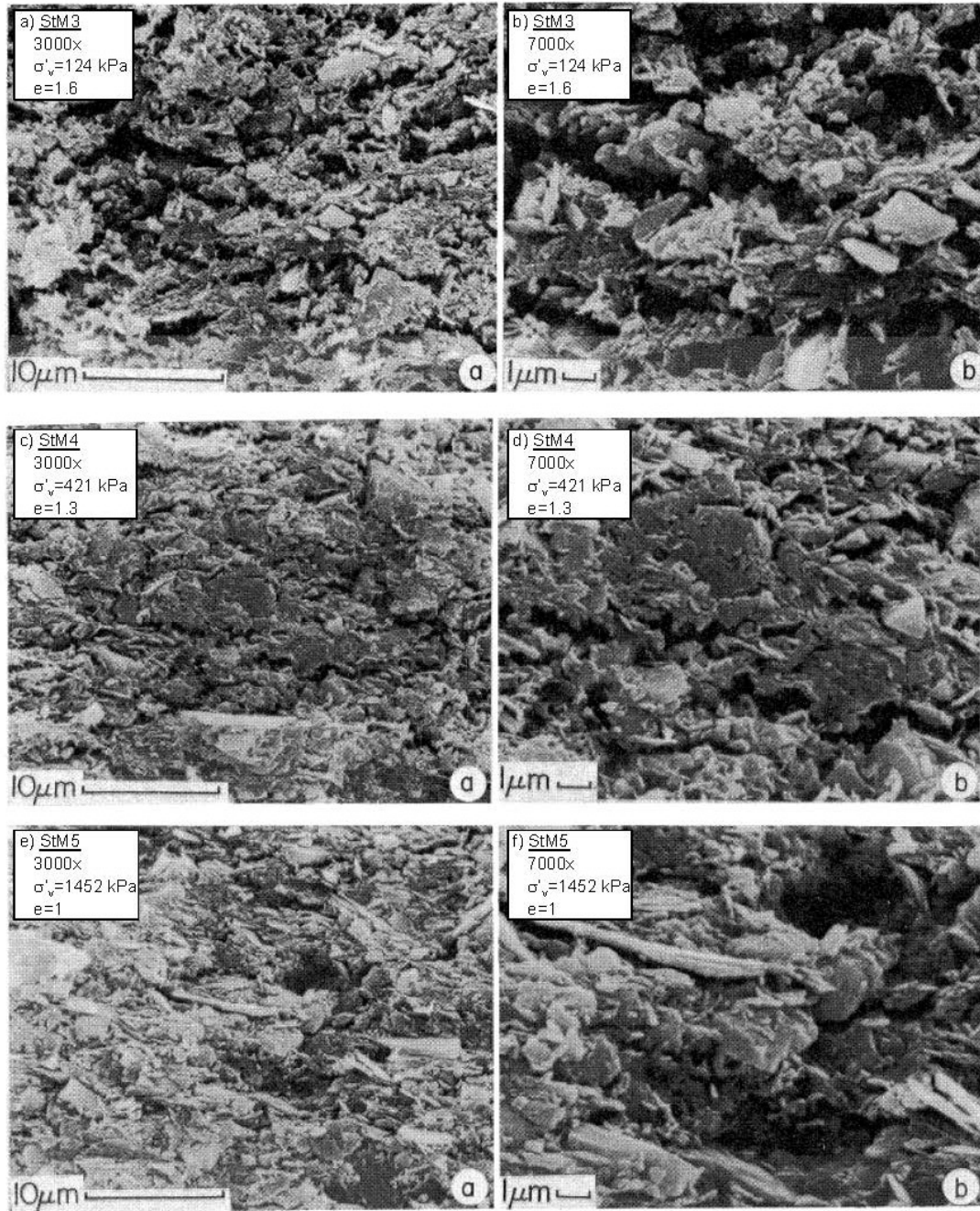


Figure 7. Evolution in 1D compression of the fabric of natural soft St Marcel clay, analyzed by means of SEM at different magnification along vertical planes at states: StM3 (a, b); StM4 (c, d); StM5 (e, f) in Figure 2 [source: Delage & Lefebvre (1984); modified after Guglielmi et al. (2022)].

For natural Lucera clay compressed to state Lu3 ($\sigma'_v = 5$ MPa; Figure 1), indices of micro-REV fabric orientation in the range 0.289-0.335 are obtained from image processing (an example is given in Figure 14e, 14f), indicating, again, that 1D compression to high pressure may determine only a minor increase in orientation, with respect to in situ clay fabric Lu1 ($L = 0.22$ -0.24).

The evolution in PSD of the stiff clays during 1D loading beyond gross-yield is shown in Figure 15. The PSD curves of Pappadai clay (Figure 15a) are indicative of a translation of the DPS towards smaller pore sizes and a reduction of the DPS frequency, both starting soon after gross-yield (Pa2 , $\sigma'_v = 3$ MPa; Figure 1) and continuing all way through compression to high pressures. The same evolution is observed

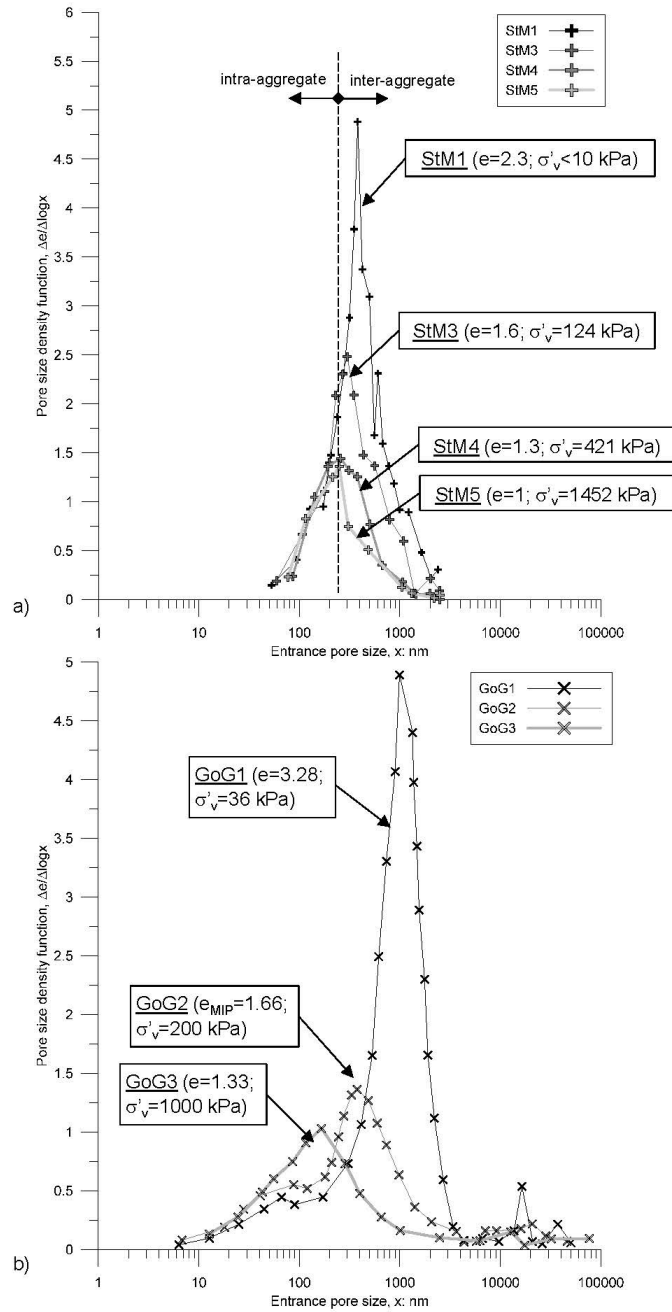


Figure 8. Evolution in 1D compression of PSD curves of a) natural St Marcel clay [states in Figure 2; replotted and adapted after Delage & Lefebvre (1984)] and b) natural GoG clay [states in Figure 2; after Hattab et al. (2013)]. Note that e_{MIP} is the maximum void ratio measured in the MIP test, reported when the specimen void ratio is not available [modified after Guglielmi et al. (2022)].

for Lucera clay (Figure 15b) and for the PSD of stiff Kyoto clay [1D compressed to ≈ 10 MPa; Tanaka et al. (2003)]. Such reduction in size of the intra-aggregate pores suggests that 1D compression determines the vertical straining of either the stacks, or the relic random fabric portions of the stiff clays. Such straining is such to keep constant the index of average fabric orientation, L , all way through 1D compression from the in-situ state of the stiff clay to high pressure, as

also observed for reconstituted clays 1D compressed to high pressures (part I-paper).

3.3 Different η compressions

For natural GoG clay isotropically compressed from the undisturbed state GoG1 (Figure 2) to $p' = 1000$ kPa, Hattab et al. (2013) observed a loss of inter-aggregate pores

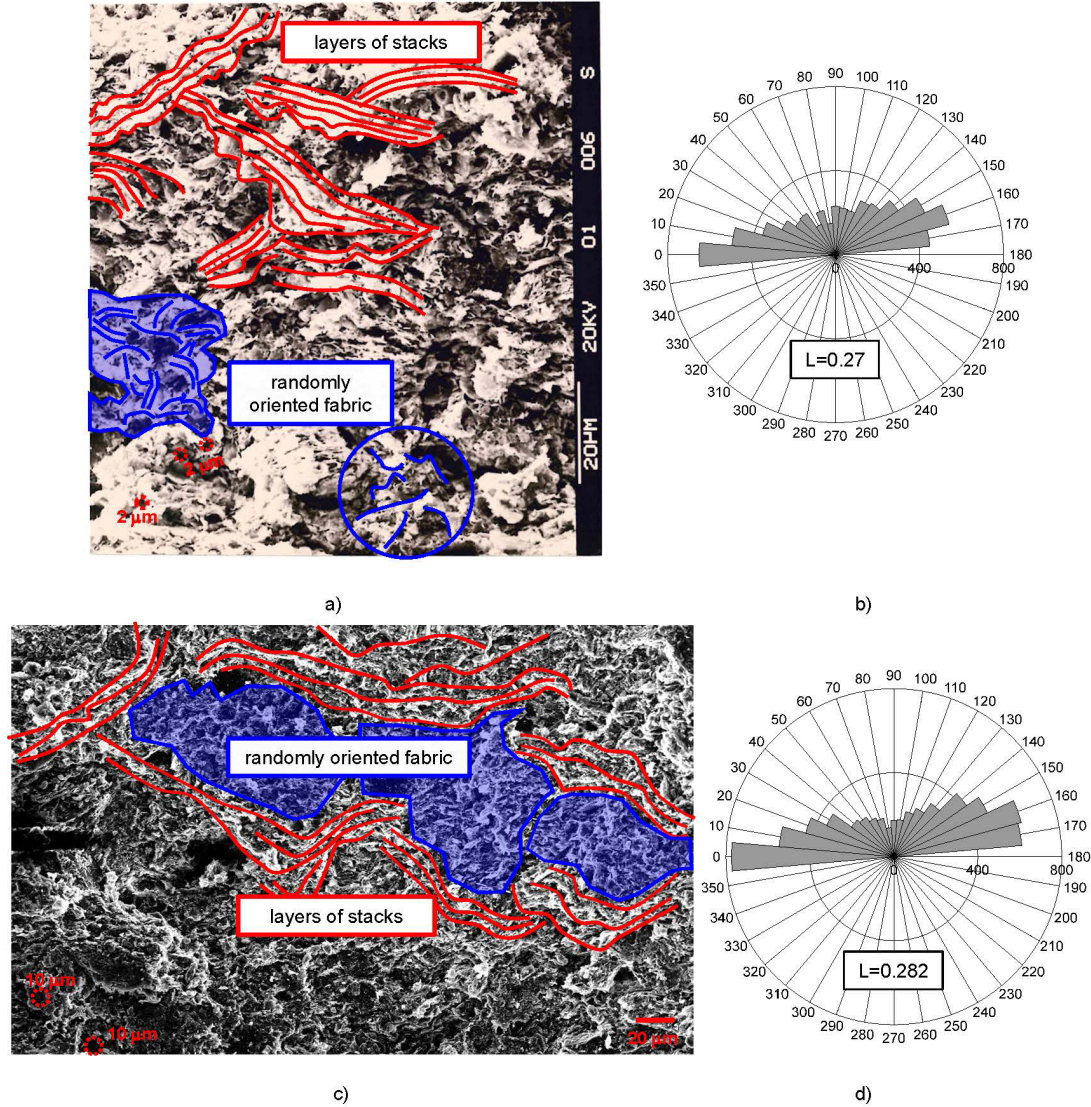


Figure 9. Micro-REV fabric and corresponding direction histograms of: natural Pappadai clay (a, b; state Pa1 in Figures 1, 2) [modified after Cotecchia et al. (2019)]; natural Lucera clay (c, d; state Lu1 in Figures 1, 2).

and bimodality [see Figure 19b in Guglielmi et al. (2022)] and a random orientation fabric. As for reconstituted clays (part I-paper), the change in PSD appears faster in isotropic than in 1D compression.

Pre-gross-yield, isotropic compression of natural Lucera clay ($p'_{y, is} \approx 2700$ kPa; Y_{iso} in Figure 1), from Lu1 to iLu2 ($p' = 1640$ kPa; Figure 3), is found to induce a very limited change in PSD, the same as pre-gross yield 1D compression (Figure 16). In contrast, isotropic compression beyond gross yield to iLu3 ($p' = 4200$ kPa $\approx 1.5 \cdot p'_{y, is}$; Figure 3) determines a reduction in DPS of natural Lucera clay to 220 nm (Figure 16), again at a faster rate than in 1D compression (state Lu3, $\sigma'_v = 5$ MPa; Figure 3: both iLu3 and Lu3 underwent the same volumetric strain of 7% since Lu1), as found already for both soft or reconstituted clays.

The micro-REV fabric orientation for state iLu3 (Figure 17) suggests that isotropic compression to $p' \approx 1.5 \cdot p'_{y, is}$ is not enough to induce a considerable change in L , which is found still high (range 0.25-0.32; Figure 17). Small particle aggregations defining concentric layouts are observed in both iLu2 and iLu3 (Figure 17b), which might represent just the start of a reduction in fabric orientation degree.

4. Modeling implications of the micro-rev changes: preliminary conclusions

The data discussed so far, in both this part II-paper and previous part I-paper, allow for a conceptual modelling of the microstructure evolution under compression of natural versus reconstituted multi-mineral clays, of low-medium to high

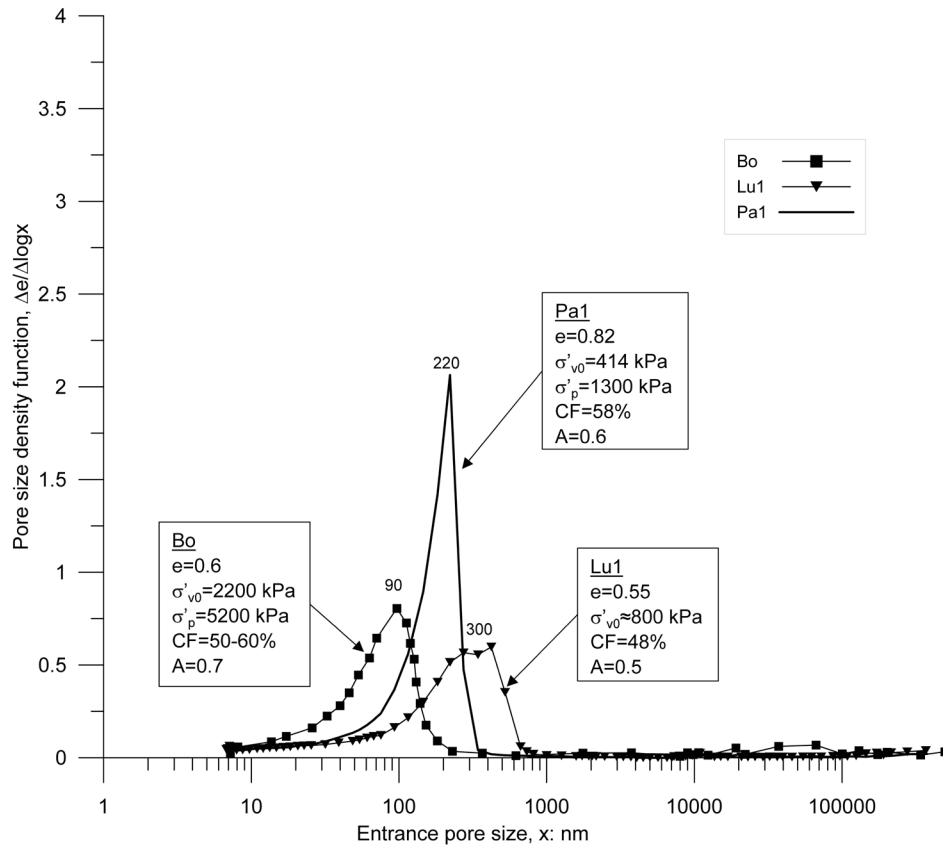


Figure 10. Comparison of PSD curves of stiff Pappadai, Lucera and Boom clay at undisturbed state [Pa1, Lu1, Bo in Figure 2; data for Boom clay after Lima et al. (2008); modified after Guglielmi et al. (2022)].

activity (Table 1). Figure 18 shows such conceptual model, in the $I_v - \sigma'_v$ plane, with reference to 1D compression. Based on the post-mortem micro-analyses discussed above and in the part I-paper, the model portrays a fabric of low orientation degree, $L \ll 0.21$, when the clays are consolidated to $I_v > 0$, whether in the field, or in the laboratory (from slurry). Such fabric is classifiable as either honeycomb or bookhouse [see Figure 7f in the part I-paper; Sides & Barden (1971); Griffiths & Joshi (1990)]. Inter-aggregate macropores and intra-aggregate micro-pores characterize the honeycomb fabric PSD, which is markedly bimodal. In contrast, macropores are more evenly distributed in the bookhouse fabric (e.g. St Marcel clay), whose PSD is already closer to mono-modal at the early stages of compression. Therefore, the micro-scale data provide evidence that natural and reconstituted clays of low-medium to high activity achieve fabrics of low degree of orientation whether they are consolidated from slurry in the laboratory⁴, or in either marine or periglacial environments over geological time.

Upon 1D compression of these initially high I_v clays, either natural or reconstituted, the model shows a major loss of macropores (§3.1; Figures 5 and 8) and the onset

of a monomodal PSD for any clay reaching I_v about 0. At the same time, as observed for reconstituted clays (see Figure 13 in the part I-paper), a diffuse increase in fabric orientation degree starts, since particles and domains tend to coalesce into stacks, in the same way for 1D compression in the laboratory of reconstituted clays, or in the field, for natural clays under burial. The single DPS, recognizable for all clays by $I_v < 0$, occur mostly within the stacks and evolves towards smaller values, reaching values typical of microporosity (Figure 18).

As shown by the model, the development of stacks that takes place through 1D compression towards $I_v < 0$ occurs for: i) the reconstituted clay 1D compressed in the laboratory from a slurry of initial $I_v \gg 0$; ii) the soft natural clay of high I_v in the field, 1D compressed in the laboratory post-gross yield; ii) the natural clay 1D compressed under burial in the field, during the geological history. The enrichment in stacks with 1D compression brings about an increase in micro-REV fabric orientation degree, such that L becomes higher than 0.21 for all the cited classes of clays. However, the maximum micro-REV fabric orientation degree is reached already by I_v values a little below 0 (depending on clay composition). During further 1D compression to high pressures, the clay micro-REV fabric keeps L_{const} , relics of bookhouse fabric

⁴Using either distilled or tap water.

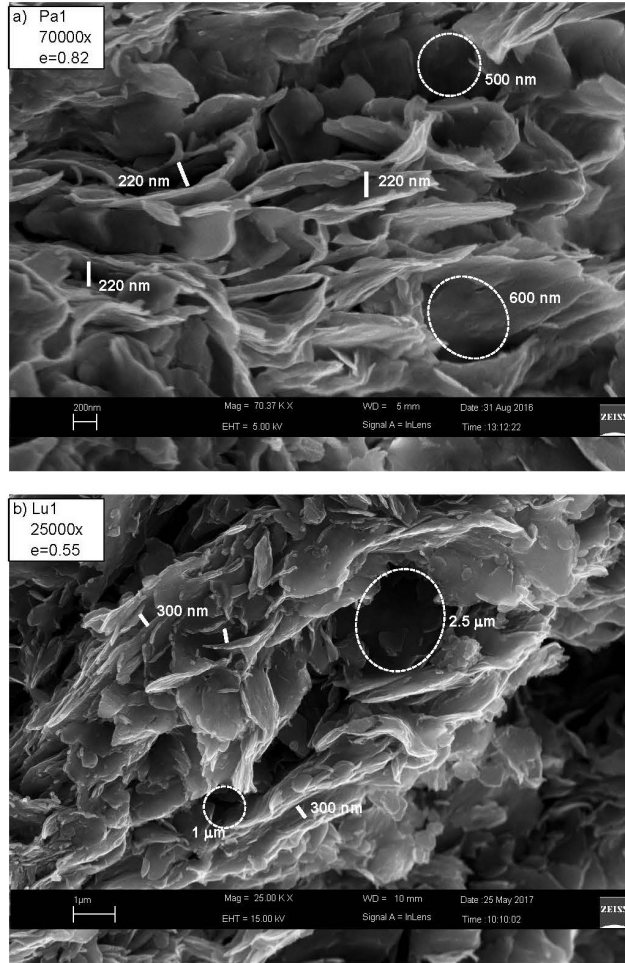


Figure 11. High magnification SEM micrographs of natural Pappadai clay (a; Pa1 in Figure 1) and natural Lucera clay (b; Lu1 in Figure 1); modified after Guglielmi et al. (2022).

are preserved between the stacks (Figure 18) and particle orientation does not become ubiquitous (complete preferred orientation, CPO, is achieved within the stacks and low orientation fabric is interbedded between the stacks).

As anticipated in part I-paper, the reduction in clay void ratio while micro-REV fabric orientation has become steady, L_{const} , results from a combination of micro-scale processes taking place with 1D compression at $I_v < 0$. These are: a) the rotation and coalescence of particles and domains with the sub-horizontal stacks, contributing to their thickening; b) further reduction of the DPS due to vertical straining of both the stacks and the relic bookhouse fabric portions. The stacks, though, are more compressible than the preserved random fabric portions (part I-paper) and contribute most to the overall clay straining. At the same time, though, they are thickened thanks to further orientation of particles, so that, on the whole, the ratio between the CPO fabric portions and the flocculated ones does not increase, and L remains constant. Such finding represents a major target of micro-

scale modelling of clays (see Figure 1, line b in the part I-paper), either reconstituted or natural. At the same time, this finding provides indications for the hardening function of constitutive laws, formalized in the framework of elastoplasticity and accounting for fabric as internal variable within a microstructure parameter of the constitutive law (see Figure 1, line a-ii in the part I-paper). It is proposed that, for the hardening in 1D compression, $L(I_v)$ (Figure 18) might be used as function representing the evolving micro-REV fabric orientation (e.g. Figure 17b in the part I-paper). Both the hardening function and the elastic stiffness anisotropy of the clay under 1D compression could be related to $L(I_v)$.

The micro to macro-scale investigation results discussed in the previous sections show also that, despite the similarities in fabric features between the natural and the reconstituted clay at the same void ratio (Figure 18), the corresponding microstructures are of different strength. Hence, such difference is evidently due to a difference in bonding between the two types of clays, and bonding can be considered the main source of the differences between the gross yield states in 1D compression of the natural and reconstituted clays in Figure 18.

It is worth recalling that, in the literature, gross yield corresponds to the effective stress state, $Y(\sigma'_{ij})$, after which a major immediate drop in stiffness occurs along a given stress path, and for clays, this has been assumed to correspond to the onset of major microstructural change (Hight et al., 1992; Smith et al., 1992; Cotecchia & Chandler, 1997, 2000). Direct evidence of such change has been seldom provided (Cotecchia et al., 2016, 2019; Guglielmi et al., 2022) and both parts, I and II, of this keynote lecture contribute to the provision of such evidence. Furthermore, for both natural and reconstituted clays, gross yield has been generally found to occur near the state boundary surface in the $v-q-p'$ space, SBS [e.g. Leroueil et al. (1979), Graham et al. (1988), Cotecchia & Chandler (2000), Smith et al. (1992)], as shown for some clay examples in Figure 19. The model in Figure 18 suggests that the difference in bonding between the natural and the reconstituted clay is the main source of the differences between the gross yield surface of the natural clay, $F(\sigma'_{ij})$, and that of the reconstituted clay, $F^*(\sigma'_{ij})$, for the same hardening stage. In the same way, bonding is the source of the difference between the natural clay state boundary surface, SBS, and the reconstituted clay SBS* (e.g. Figure 19), in the $v-q-p'$ space. It follows that the microstructure constitutive function controlling the clay gross yielding and hardening law (see Figure 1, line a-ii, in the part I-paper) should include the clay micro-REV fabric orientation degree, the porosimetry, and the clay bonding, all evolving with compression.

The data presented in the previous sections provide also direct evidence of the insignificant changes in clay microstructure that occur during monotonic loading paths pre-gross yield, at least for either reconstituted, or intact unfissured clays of the type in Table 1. For example, the data show that along monotonic 1D swelling, or 1D compression,

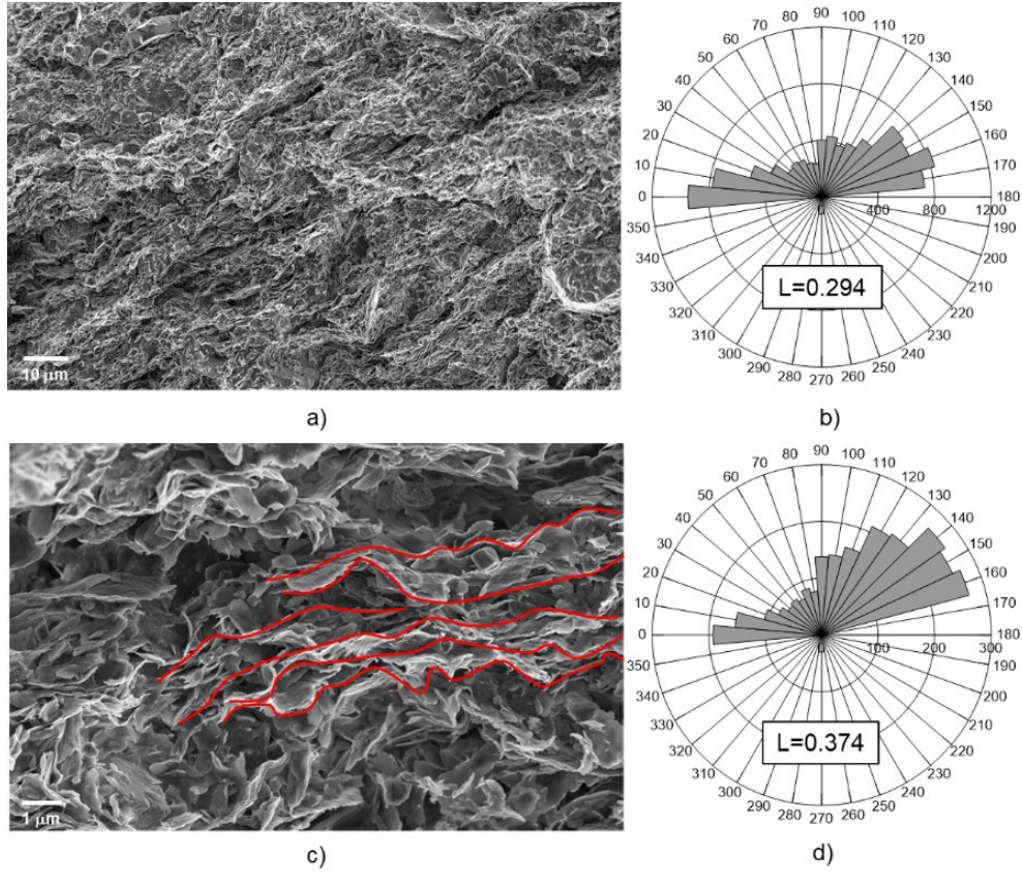


Figure 12. SEM micrographs along vertical fractures and corresponding direction histograms of natural Pappadai clay 1D swelled to $\sigma'_v = 20$ kPa (state PaS in Figure 1) at the micro-REV scale (a, b) and at higher magnification (c, d), with example of stack [modified after Guglielmi et al. (2022)].

or isotropic compression pre-gross yield, no changes of either the bonding strength (reflected by C_s^*/C_s), or the PSD, or the orientation degree of the micro-REV fabric (L), are observed for either the natural or the reconstituted clay. Such observations confirm previous speculations about the source of gross yielding and should inform the modeling of the hardening plasticity inside the SBS [see Figure 1, line b in the part I-paper; e.g. Kavvas & Amorosi (2000), Rouainia & Muir Wood (2000), Baudet & Stallebrass (2004)], suggesting that this should not be related to microstructural changes in monotonic loading paths. This is not the case, however, for fissured clays, as shown by Vitone & Cotecchia (2011) and Vitone et al. (2019). Also, cyclic loading pre-gross yield may be capable of weakening the clay bonding [e.g. Massaro (2004)] and affecting the clay fabric and PSD below the SBS, also for un-fissured natural clays.

Given the similarity in the type of fabric for the natural and the reconstituted clay of given void ratio and the negligible change in microstructure taking place with monotonic loading pre-gross yield, if the natural bonding is isotropic and amplifies the gross yield stresses through a scalar factor common to all the loading paths (i.e. the onset

of the natural bonding degradation, determining gross yield, is not influenced by the stress ratio and occurs when it is reached a given ratio $\sigma'_{ijY} / \sigma_{ije}^*$ common to all the loading paths), the SBS and the SBS* are homotetic. This is the case of natural Pappadai clay, whose gross yield curve is compared with that of the reconstituted clay in Figure 19a, and of several other firm to stiff clays [e.g. Coop & Cotecchia (1995), Cotecchia & Chandler (2000), Cafaro & Cotecchia (2001), Cotecchia et al. (2007), Guglielmi (2018)]. In this case, the scalar stress sensitivity, $S_\sigma = p'_{K0Y} / (p_{K0Y}^*)$, may be considered the effect on gross yielding of the difference in bonding between the natural and the reconstituted clay.

If the bonding is isotropic, but the onset of bonding degradation occurs at ratios $\sigma'_{ijY} / \sigma_{ije}^*$ which vary depending on the loading paths, i.e. bonding degradation is sensitive to the stress ratio η , the SBS and SBS* are not homotetic. This is the case for several highly sensitive soft clays, e.g. Saint Alban and Bothkennar clay, whose SBS and SBS* are shown in Figures 19b and 19c. For such a class of clays, S_σ is found to represent the gross yield amplification ratio only for loading paths characterized by $\eta \geq \eta_{K0^*}$, as will be discussed later.

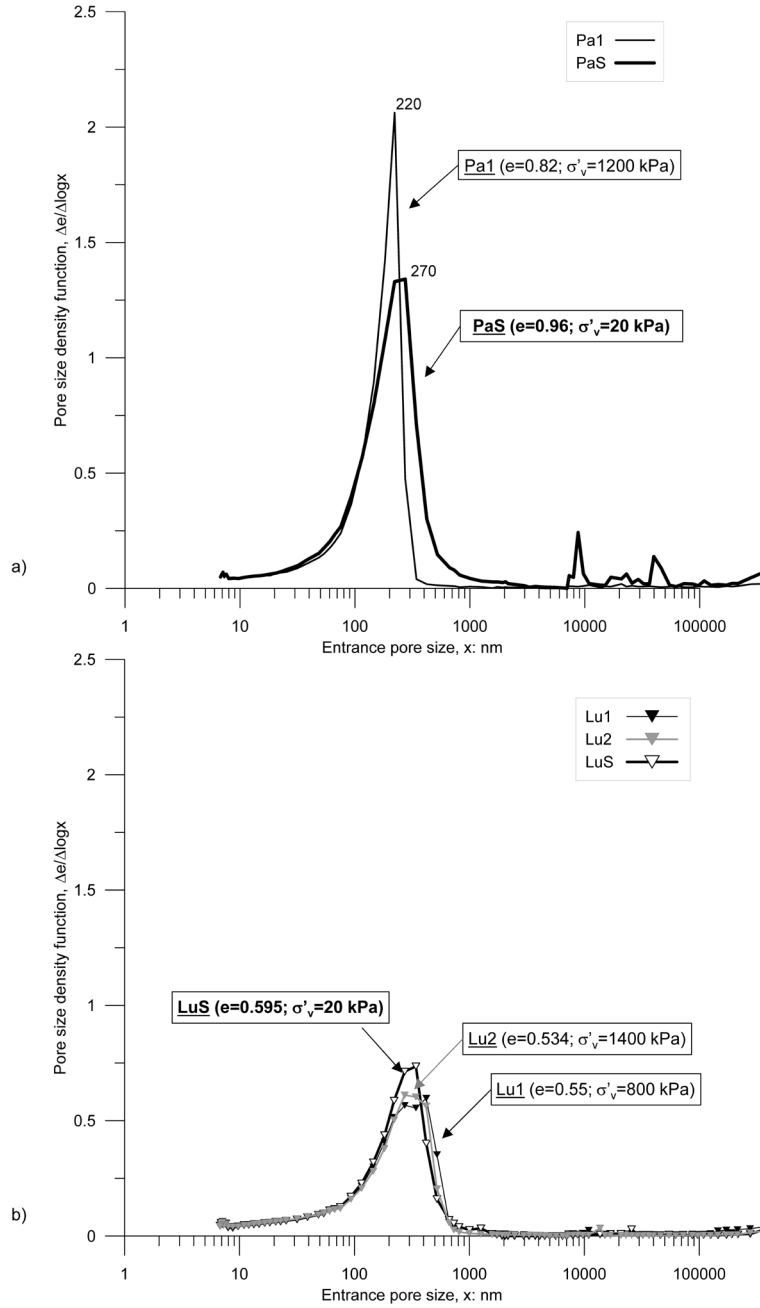


Figure 13. Comparison of PSD curves for pre-gross-yield states of a) natural Pappadai clay and b) natural Lucera clay [modified after Guglielmi et al. (2022)].

Regarding the rate of bonding decay, the data discussed in the previous sections provide evidence of a rather fast decay of the bonding strength over gross yielding, since the swell sensitivity of the natural clay, C_s^*/C_s , drops to 1 (Figure 1) soon after gross yield. Correspondingly, S_σ starts decreasing, and the fabric starts changing too (Figure 18). However, despite the fast drop of C_s^*/C_s , S_σ decreases slowly and the function $S_\sigma(\epsilon_{vol}^p)$ represents the effect of current bonding on the clay 1D compressibility. $S_\sigma(\epsilon_{vol}^p)$ keeps being higher than 1 up to high pressures. This observation highlights that the

combination of fabric and bonding acquired by the natural clay differs from that of the reconstituted clay at any stage of compression, despite the qualitative similarity of the natural and reconstituted clay fabric for any given void ratio. Hence, natural and reconstituted clays must be considered different materials at the micro to the macro-scale, at any stage of loading, despite their equal composition and fabric features.

Since the data discussed so far in this part II-paper have shown that, irrespective of the bonding features, the fabric changes occurring in natural clays under compression, either

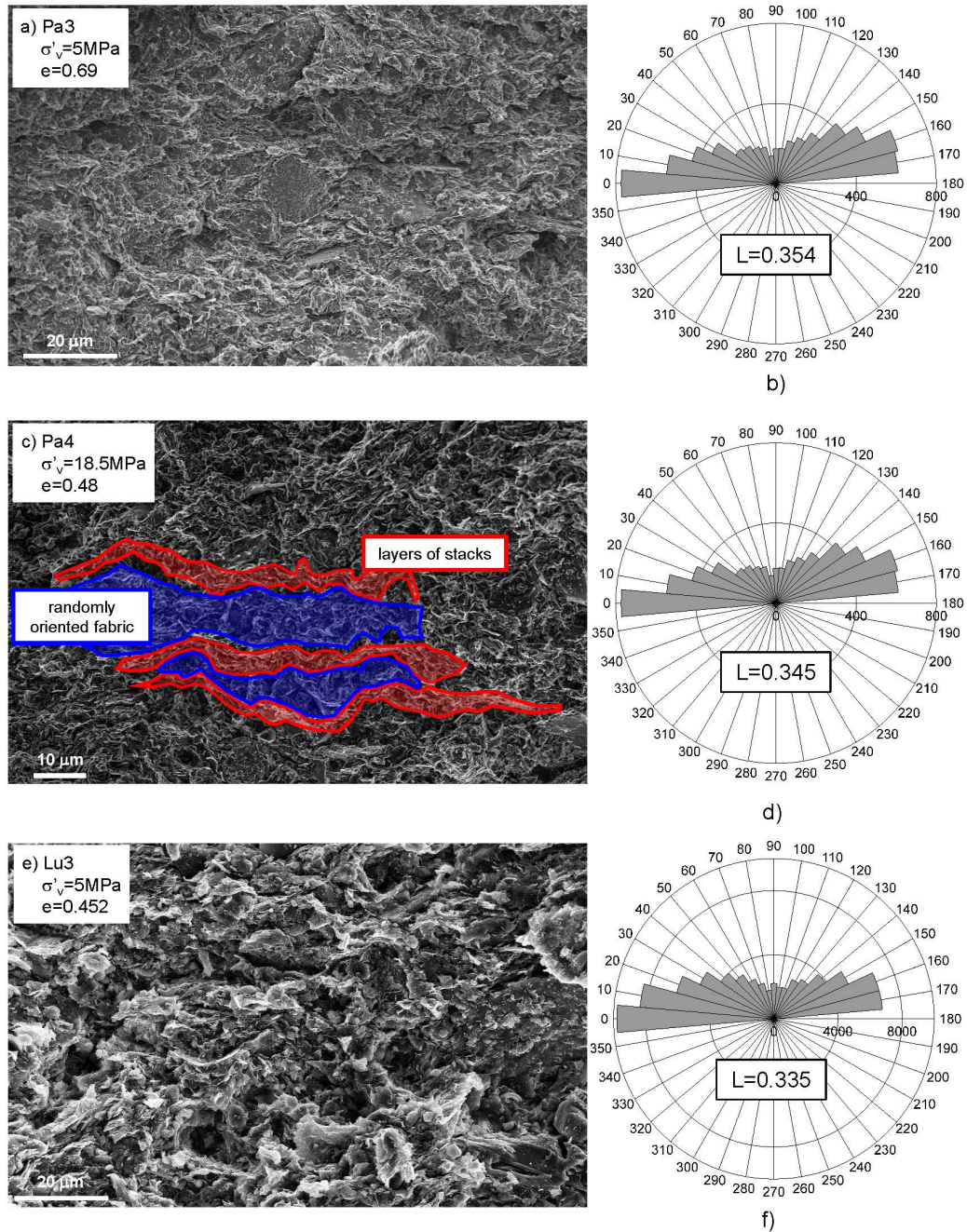


Figure 14. Micro-REV fabric and corresponding direction histograms of: natural Pappadai clay compressed to state Pa3 (Figure 1) (a, b) and to state Pa4 (Figure 1) (c, d); natural Lucera clay compressed to state Lu3 (Figure 1) (e, f); modified after Guglielmi et al. (2022).

1D or isotropic, are similar to those observed for reconstituted clays, in Figure 20a the same framework used in part I-paper to schematize such changes for reconstituted clays is used to represent the micro-REV fabric changes in natural clays, under either 1D or isotropic compression (in this case along the K_0 NCL and the INCL, whereas in Figure 13 of part I-paper these changes occur along K_0 NCL* and INCL*, respectively). Accounting for such framework and for the changes in bonding strength discussed above for the natural clays, indications about

the reasons for the differences in yield surface and plastic strain hardening of natural stiff ($I_v < 0$) versus soft clays ($I_v > 0$) can be commented. It is convenient to start with comparing the gross yield hardening of the natural stiff clay with that of the corresponding reconstituted clay when both are at $I_v < 0$. The analysis will again refer first to the strain hardening in 1D compression and, secondly, to that in isotropic compression, concluding with more general considerations referring also to the other possible loading paths.

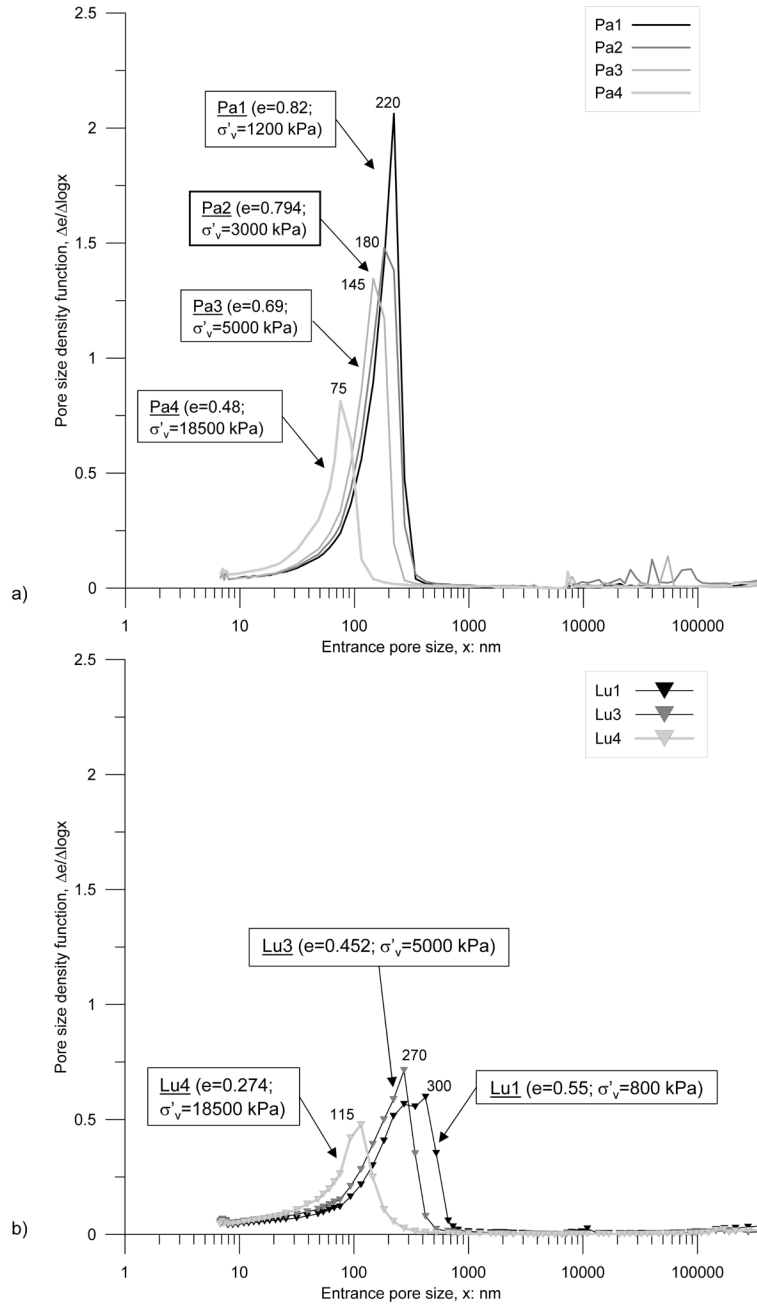


Figure 15. Evolution in 1D compression of PSD curves of natural Pappadai (a) and natural Lucera clay (b); modified after Guglielmi et al. (2022).

The data have shown that, for $I_v < 0$, the orientation degree acquired by both the natural and the reconstituted micro-REV fabric is already high, $L > 0.21$. Furthermore, since the degree of fabric orientation does not increase with 1D compression post-gross yield of the natural clay, the only evolving fabric feature which influences the hardening in post-gross yield 1D compression is the DPS reduction and the corresponding reducing porosity (Figure 18). It follows that the natural fabric changes in 1D compression are a source of an isotropic gross yield hardening, relating to plastic volumetric straining, as already observed for reconstituted

clays. This is made evident by macro-scale test data such as those acquired through shear testing of specimens 1D compressed to states either about gross yield, or post-gross yield, like those shown in Figure 20b and 20c, respectively for stiff Montemesola clay (Cotecchia et al., 2007) and Vallericca clay (Amorosi, 1996; Amorosi & Rampello, 1998). For these clays the undrained shear paths of specimens compressed to different post-gross yield pressures are similar. However, the hardening function holding for reconstituted clays, $p^*(\epsilon_{vol}^p)$, does not normalize these stress paths, due to the concurrent weakening of bonding, reflected in the scalar function

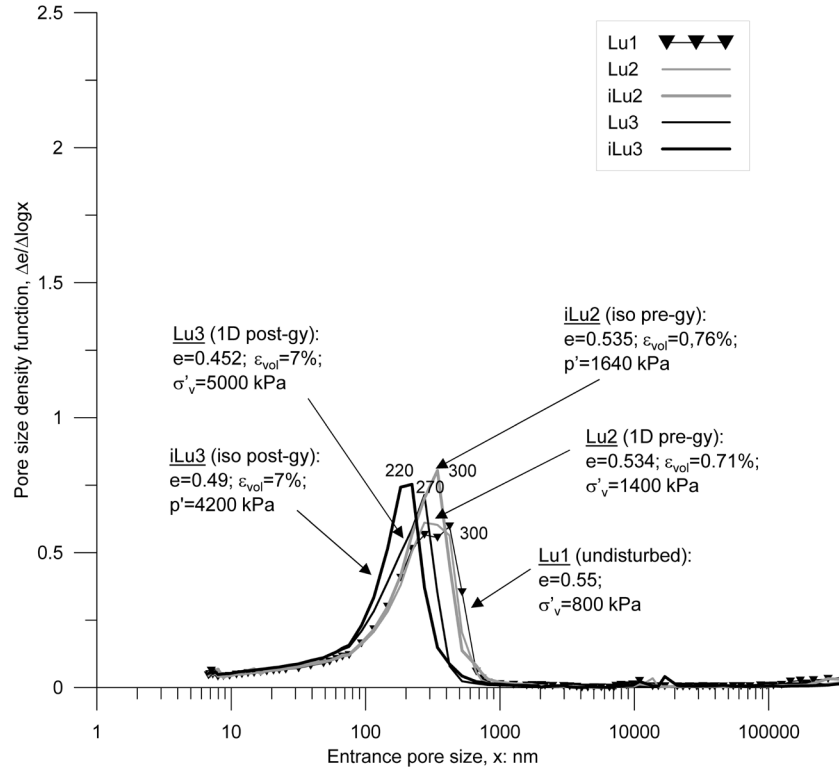


Figure 16. Comparison of PSD curves for specimens of natural Lucera clay 1D and isotropically compressed to states pre- and post-gross-yield to approximately the same void ratios (states in Figure 1); modified after Guglielmi et al. (2022).

$S_{\sigma}(\varepsilon_{vol}^p)$. Such weakening determines a negative hardening contribution, which combines with the positive hardening determined by the volumetric plastic straining for natural clays, but not for reconstituted clays.

As suggested by Cotecchia & Chandler (2000), for this class of clays, the scalar function $S_{\sigma}(\varepsilon_{vol}^p)p_e^*(\varepsilon_{vol}^p)$ may be suitable as isotropic volumetric hardening function to predict the response of the 1D compressed natural stiff clay. Such suitability is confirmed by the normalization of the results of shear tests on stiff clay specimens 1D compressed to different p'_{K0y} , shown in Figure 21. Both $S_{\sigma}(\varepsilon_{vol}^p)$ and $L(\eta_{K0})$ may become components of the microstructure constitutive parameter of the clay constitutive law, the first to represent the clay bonding, and the second for the clay fabric under 1D compression.

For such class of clays, which are only mildly sensitive ($S_{\sigma} \leq 3$), the natural bonding is rather stable and the onset of bonding degradation has been found not to be sensitive to η and, as such, to be the source of an amplification of the gross yield surface (scalar ratio $\sigma'_{ijy}/\sigma'_{ije} = S_{\sigma}$, or $p'_{K0y}/p^*_{K0y} = S_{\sigma}$). Therefore, the SBS and the SBS* are homothetic, as shown in Figure 19a for Pappadai clay. Both state boundary surfaces are seen to have an arch shape, probably partly due to the high degree of micro-REV fabric anisotropy. Nonetheless, $p'_{yi} > p'_{YK0}$ and $N-N_{K0} > 0$, as for reconstituted clays [see Cotecchia et al. (2007); also, Lucera clay data in Figure 3

and Figure 20a]. Evidently, the reduction of η , does not cause a weakening of the clay bonding faster than for $\eta \geq \eta_{K0}$, so that $S_{\sigma} = p'_{K0y}/p^*_{K0y} = p'_{yi}/p^*_{yi}$.

For this type of clays, isotropic compression post-gross yield determines a weakening of bonding equivalent to that occurring in 1D compression, as shown by the drop in swell sensitivity, κ^*/κ , recorded post isotropic gross yielding, as shown in Figure 3. With regard to fabric, the isotropic compression post-gross yield seems to cause a reduction of the macro-porosity faster than for 1D compression (Figure 16) and the onset of a less oriented fabric, but the fabric transition seems as slow as for reconstituted clays (see part I-paper). Therefore, the micro-scale data suggest that, for either 1D or isotropic compression (varying η), the microstructure of the stiff clays undergoes similar evolutions to those observed for reconstituted clays, except for the weakening of bonding which is experienced by the natural clay only and appears to be the same in isotropic and 1D compression (i.e. not dependent on η), being represented by the scalar function $S_{\sigma}(\varepsilon_{vol}^p)$. Such similarity is consistent with the parallelism of the INCL and the K_0 NCL of the stiff natural clays, making $S_{\sigma} = p'_{K0y}/p^*_{K0y} = p'_{yi}/p^*_{yi}$ at any void ratio.

It follows that the fabric changes, similar for the natural and the reconstituted stiff clays undergoing the same constant η compression, result in constant L values which reduce with reducing η (see Figure 17 in the part I-paper). Consequently,

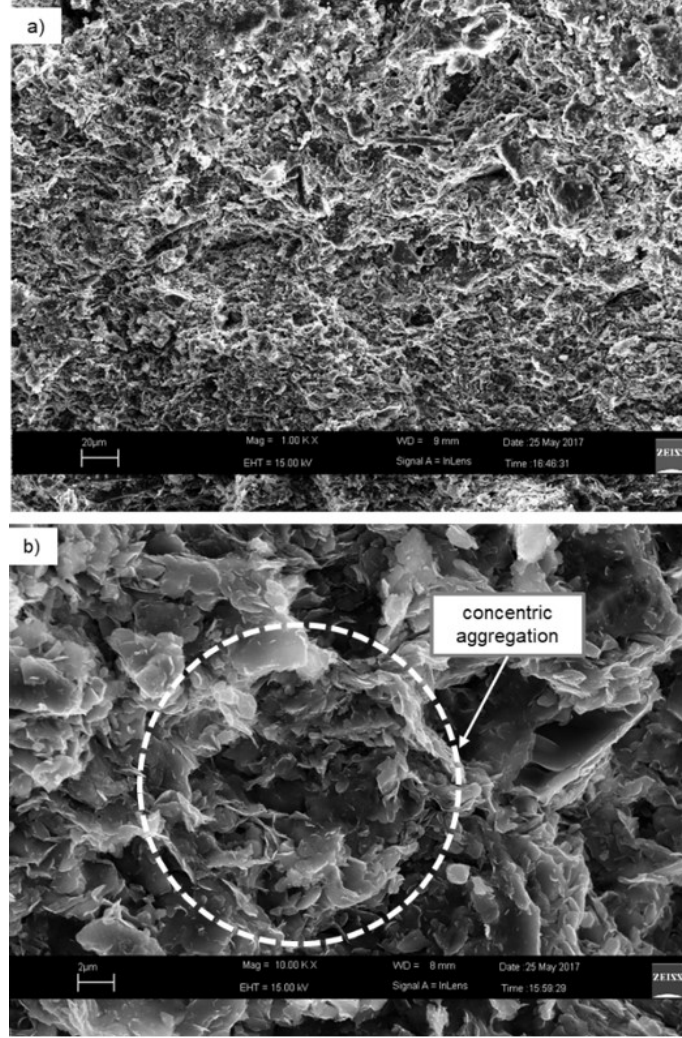


Figure 17. Fabric of natural Lucera clay isotropically compressed to post-gross-yield state iLu3 (Figure 3) at the micro-REV scale (a); example of concentric aggregation of particles (b); modified after Guglielmi et al. (2022).

the function controlling the isotropic hardening of the clay depends on η , as sketched in Figure 20a (as well as in Figure 13 of part I-paper), for both natural and reconstituted clays. However, the response of the natural clay depends also on bonding weakening, which determines a negative hardening contribution. This is exemplified in Figure 19a, where, along with the gross yield curve of the undisturbed Pappadai clay, the shear stress paths of natural clay samples compressed isotropically post-gross yield are also shown; it can be seen that these are not normalized by the sole hardening function $p_e^*(\epsilon_{vol}^p) (= e^{[(N^*-1)/2] \eta})$. Cotecchia & Chandler (2000) show that the isotropic volumetric hardening function $S_\sigma(\epsilon_{vol}^p) p_e^*(\epsilon_{vol}^p)$ suits, in first approximation, the normalization of the gross yield states of the undisturbed clay and those of the samples compressed post-gross yield, either isotropically, or 1D, as shown in Figures 21 and 22. However, the figures show that while the function $S_\sigma(\epsilon_{vol}^p) p_e^*(\epsilon_{vol}^p)$ predicts the influence on the clay response of the evolution in bonding strength and

PSD, it is not sufficient to predict the effects of the variation in fabric anisotropy determined by the variation in η applied during compression. The same limitation applies to the use of isotropic volumetric hardening with reconstituted clays (see Figure 18 in the part I-paper). For both the reconstituted and the natural clay, the positive hardening function $p_e^*(\epsilon_{vol}^p)$ does not represent successfully the effects of fabric on the hardening law, since it does not take into account the effects on clay stiffness of the different fabric orientation degrees, acquired by the clay for different η compressions. The different L values achieved through different η compressions cause, for example for η_{K0} and η_0 , the normalized undrained shear paths of the 1D consolidated clay to be much steeper than those of the isotropically consolidated specimens.

Differently from what is observed for stiff clays, gross yielding of soft sensitive clays [e.g. Bothkennar clay, Pisa clay, Saint Alban clay, Gulf of Guinea clay, etc.; e.g. Smith et al. (1992), Leroueil et al. (1979), Le (2008), Callisto

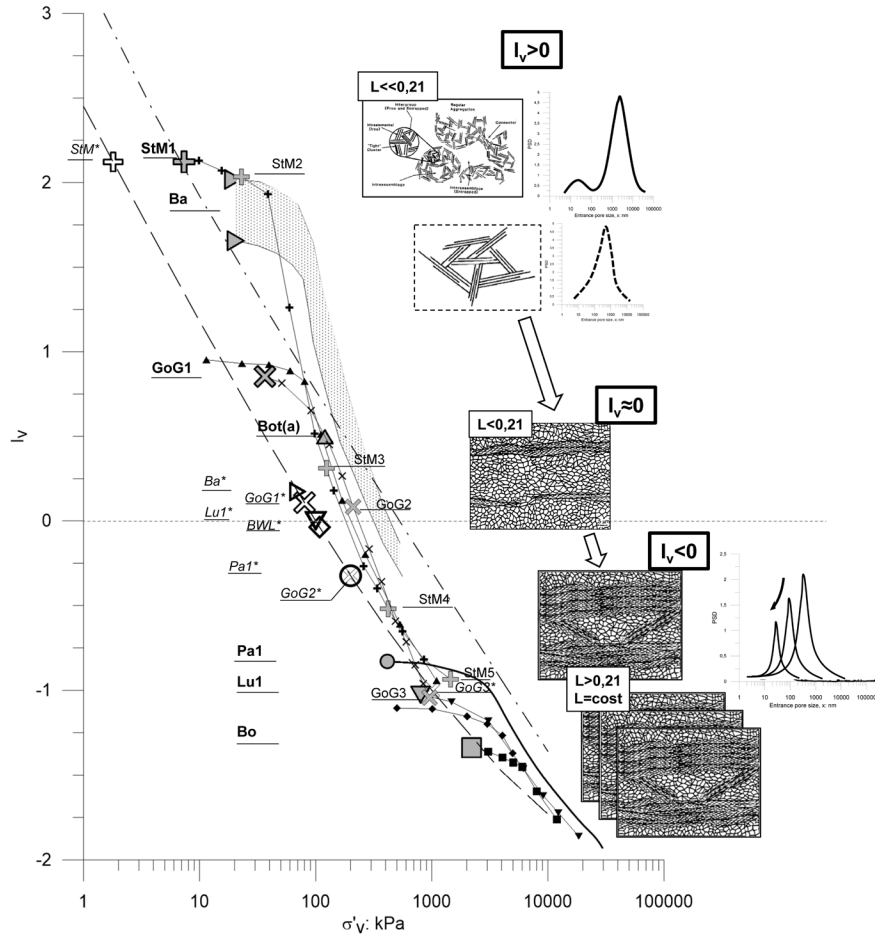


Figure 18. Conceptual model of the fabric features of multi-mineral clays under 1D compression [modified after Guglielmi et al. (2022)].

& Calabresi (1998)] is found to be dominated by a fragile weakening of the clay bonding, which is also sensitive to η . Hence, the ratio $\sigma'_{ijY} / \sigma'^*_{ije}$ is found to reduce severely with reducing η . In other words, the weakening of the clay bonding occurs much earlier in isotropic compression than in anisotropic compression. Consequently, the clay undergoes important volumetric straining soon after the η reduction to 0, as shown in Figure 23 for Saint Alban and Bothkennar clays. In this case, the drop in stiffness typically characterizing gross yielding, is much smoother in isotropic yielding than in 1D yielding and $p'_{YK0} > p'_{yi}$, $N - N_{K0} < 0$ and $S_\sigma = p'_{YK0} / p'^*_{eK0} \neq p'_{yi} / p'^*_{ei}$. Consequently, the shape of the lower portion of the SBS differs from that of the SBS*, as shown for Saint Alban and for Bothkennar clay in Figure 19b and 19c. Nonetheless, while bonding degrades, the clay compressibility in isotropic compression is lower than that exhibited by the clay in a more fragile gross yielding in 1D compression. Consequently, the state path of the clay undergoing isotropic compression crosses that of the clay in 1D compression post-gross yield in $e-p'$ plane (Figure 23) and, once bonding is largely weakened, the INCL becomes

parallel to the K_0 NCL and shifts to the right, acquiring the pattern typical of reconstituted clays. Such behaviour may be envisaged to be due to the increase in orientation that the original random fabric acquires with 1D compression post-gross yield, with respect to the lower orientation fabric of the isotropically compressed clay, in accordance with the framework shown in Figure 20a. Therefore, at relatively high pressure $N_{K0} < N$ also for this type of clay.

Once bonding has been weakened and the degree of fabric orientation has evolved through the different η compressions, the role and limitations of the hardening function $S_\sigma(\epsilon_{vol}^p) p_e^*(\epsilon_{vol}^p)$, discussed above for stiff clays, applies also to soft clays. Nevertheless, the processes taking place at the micro-scale and their macro effects at gross yielding for the natural soft clay differ from those occurring in stiff clays. On the whole, the data suggest that the soft clay response at gross yielding is dominated by the fragile bonding decay, which masks the effects of the L variations between the fabric pre- and post-gross yield. Thereafter, the clay response heads towards that of the reconstituted clay, with a corresponding variation in L controlled by η , although

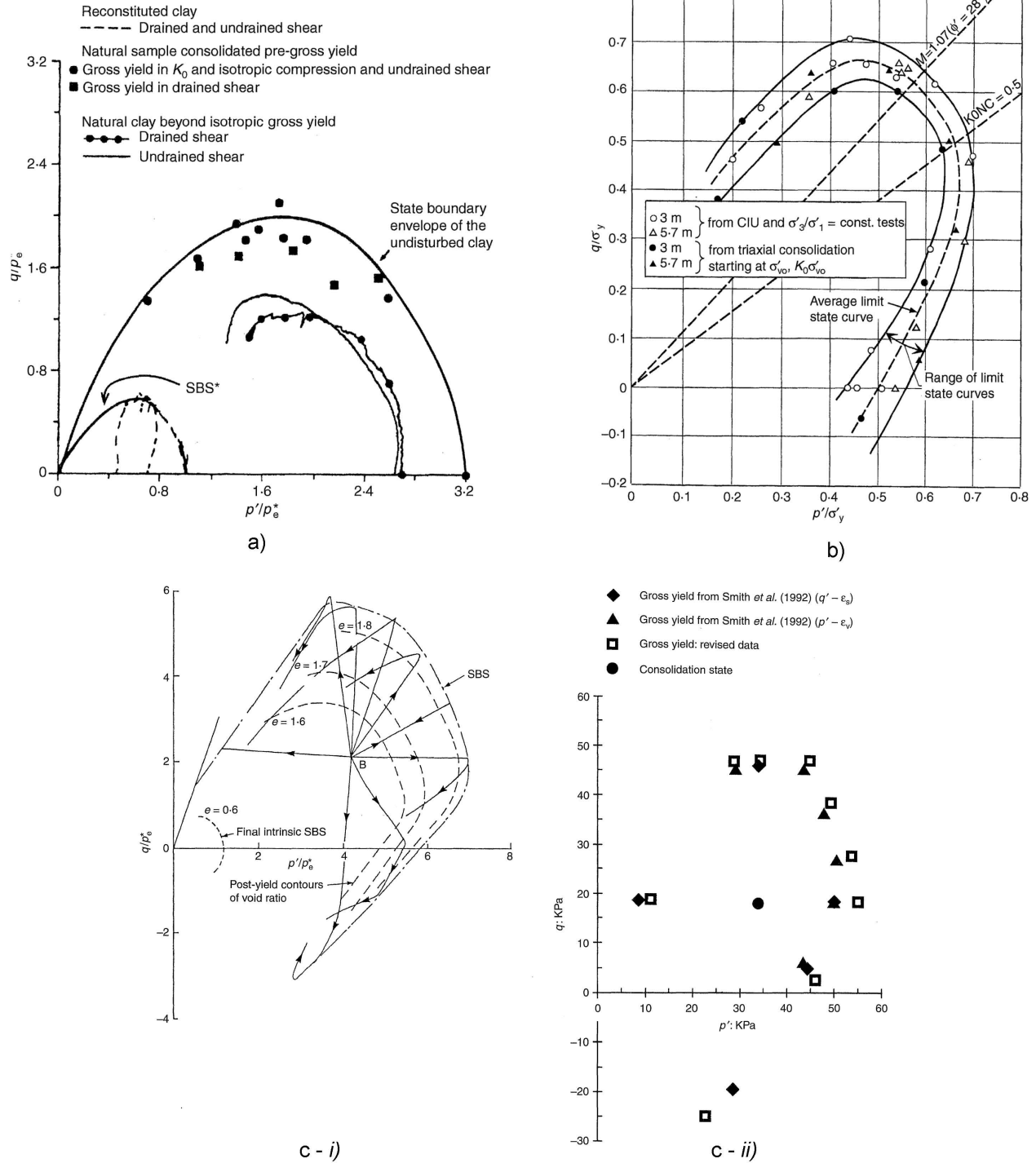


Figure 19. a) Pappadai clay data in the plane of the effective stresses normalized with respect to specific volume by means of p_e^* : stress paths and state boundary surface of the reconstituted clay, SBS*, gross yield states and state boundary envelope of the natural clay consolidated pre-gross-yield and shear paths of the natural clay consolidated beyond isotropic gross-yield [after Cotecchia & Chandler (1997)]; b) Saint Alban clay data in the plane of the effective stresses normalized with respect to specific volume by means of p_y : gross yield data for samples from two depths [from Leroueil *et al.* (1979)]; c) Bothkennar clay data: i) SBS* and results of stress path tests on the natural clay in the plane of the effective stresses normalized by p_e^* [from Smith *et al.* (1992)]; ii) gross yield states along the stress paths in (i) plotted in the effective stress plane [modified after Cotecchia & Chandler (2000)].

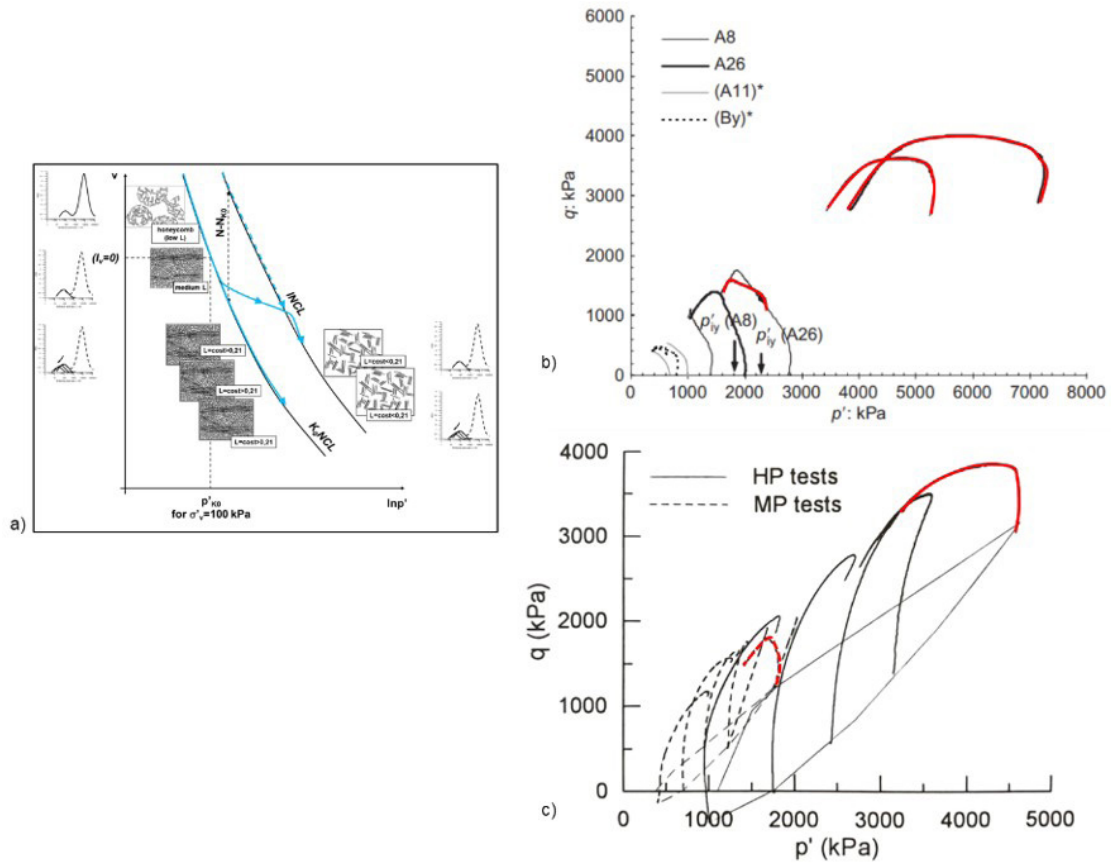


Figure 20. a) Schematic framework of the evolution in micro-fabric applying to either reconstituted (see Figure 13 of part I-paper), or natural clays subjected to compression; b) Montemesola clay: undrained shear paths of natural and reconstituted specimens consolidated before and beyond gross-yield, either isotropically, or anisotropically (these latter are highlighted in red for specimens compressed either to or beyond gross yield); c) Vallericca clay: undrained shear paths of 1D consolidated specimens; the shear paths of specimens sheared after 1D consolidation either to or beyond gross yield are highlighted in red; modified after Amorosi & Rampello (1998).

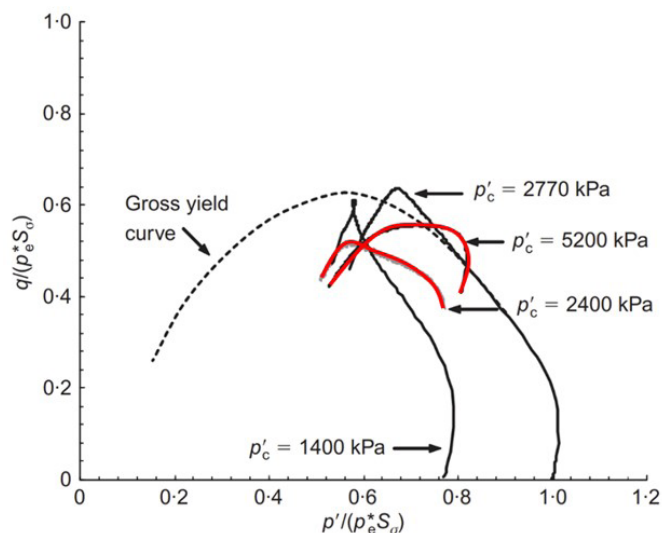


Figure 21. Undrained shear paths of specimens from sample A8 of natural stiff Montemesola clay normalised for volume and structure [modified after Cotecchia et al. (2007)].

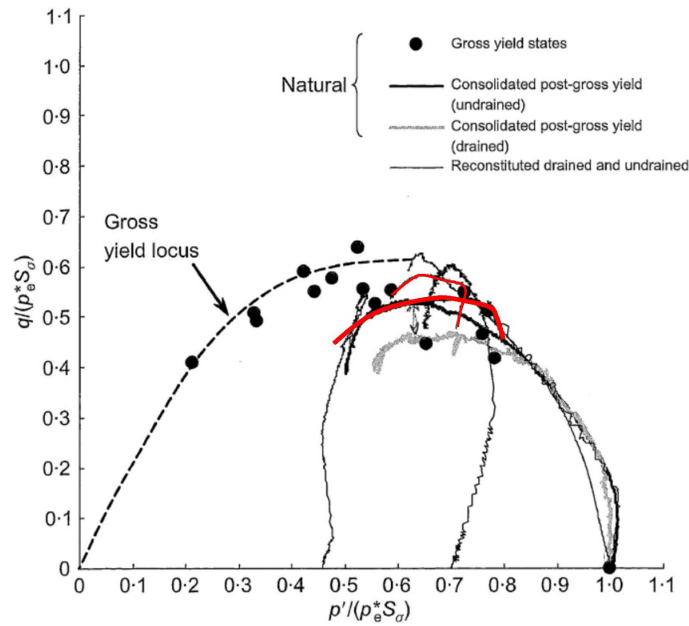
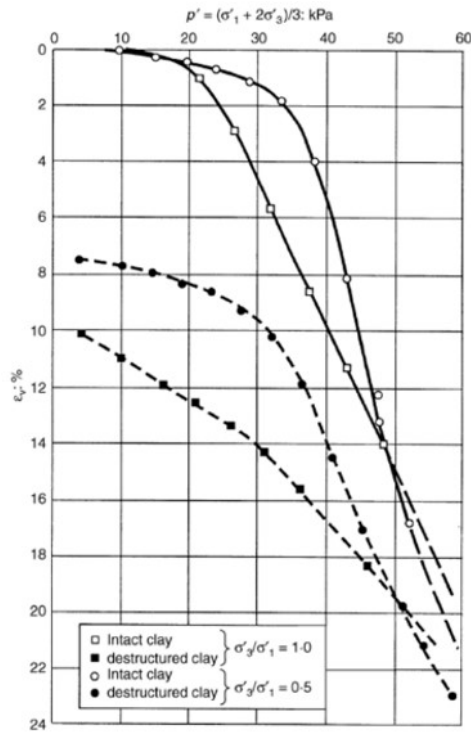
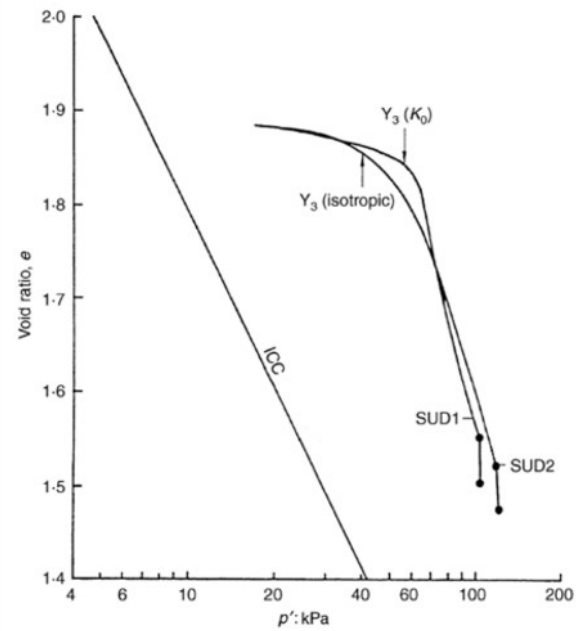


Figure 22. Pappadai clay behaviour normalized for both volume and structure: gross yield data of the natural clay, post-gross yield stress paths of the natural clay (undrained and drained), and stress paths of the reconstituted clay [modified after Cotecchia et al. (2007)]. In red, the shear paths of 1D compressed specimens.



a)



b)

Figure 23. 1D and isotropic compression curves: a) Saint Alban clay (Leroueil et al., 1979); b) Bothkennar clay [ICC = Intrinsic Compression Line; Smith (1992)].

bonding is still different from that of the reconstituted and is reflected by $S_{\sigma}(\varepsilon_{vol}^p)$.

When the undisturbed sensitive not oriented fabric of the soft clay is subjected to isotropic compression and gross yielding, again its response is dominated by the bonding decay, although it differs from that taking place in 1D compression, being less abrupt. It is evident, then, how this stage of behaviour of soft clays requires a function representative of the η - dependent bonding decay, different from $S_{\sigma}(\varepsilon_{vol}^p)$.

Declaration of interest

The authors have no conflicts of interest to declare. All co-authors have observed and affirmed the contents of the paper and there is no financial interest to report.

Authors' contributions

Federica Cotecchia: conceptualization, methodology, validation, writing – reviewing and editing. Simona Guglielmi: conceptualization, investigation, data curation, validation, writing – reviewing & editing. Francesco Cafaro: investigation, validation, reviewing. Antonio Gens: supervision, validation, reviewing & editing.

Data availability

The datasets generated analysed in the course of the current study are available from the corresponding author upon request.

List of symbols and abbreviations

e_0	initial void ratio
w_0	initial water content
A	activity index
CF	clay fraction
I_v	void index
LL	liquid limit
OCR	overconsolidation ratio
PI	plasticity index
S_t	strength sensitivity
S_{σ}	stress sensitivity
YSR	yield stress ratio
σ'_{v0}	in situ vertical effective stress

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