THE ROLE OF CLAY MINERALS IN THE UNCONFINED COMPRESSIVE STRENGTH LOSS OF ROCKS AFTER SATURATION

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ABSTRACT

The mechanical characteristics of rocks, such as their strength, elastic modulus, porosity and permeability, have been found to be reduced in the presence of water. Researches have been contacted in a range of rock types, mostly in sedimentary rocks; igneous and metamorphic rocks have been lesser investigated. From all of the available literature, it has been found that indeed water has a negative effect on such properties However, the magnitude of it is not well defined, since the range of the reduction is great through different rock types, or even into the same rock type.

In the present thesis, the role of clay minerals in the unconfined compressive strength reduction of rocks is investigated. The survey starts with providing the basic background of clay mineral structure and behavior, after hydration.

The basic aspects investigated, are the mechanisms related to clay minerals, the unconfined compressive strength loss magnitude at different saturation degrees, as well as at wetting and drying cycles, the saturation time influence, and how the strength loss behavior changes when saturated with cationic solutions. For that, two rock groups investigated, with the first group being rocks with high clay content (mudstones, siltstones and marls), and the second one being sandstones alone, in order to compare the different behavior presented by the two groups, concerning their mineral content.

After that, it is suggested that for rocks with high clay content, the wet behavior of clay is the basic reason for the strength loss. Such statements cannot be made for sandstones; however, clay seems to have an impact in sandstone strength as well.

After statistical analysis, no clear relation was found between clay content and strength loss, for both groups. After discussion, author suggested that the clay mineral itself is not sufficient enough to indicate a strong correlation, instead, the clay content should be studied in addition to the pore space, for both groups.

Other mechanisms affecting the strength reduction are presented in Appendixes. These mechanisms concern the presence of quartz and calcite, as well as the pore water itself.

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Chapter 1 Introduction

In most civil engineering cases, rock formations are considered to be a solid and stable foundation background; and for the most common applications, this consideration seems to be satisfying. However, for bigger structures, like a tunnel or a dam, or for applications of great importance and environmental influence, such as a CO2 storage aquifer, this assumption is not safe enough. Thus, the ability of rocks to support such structures should not be taken for granted, and their mechanical properties should be further investigated.

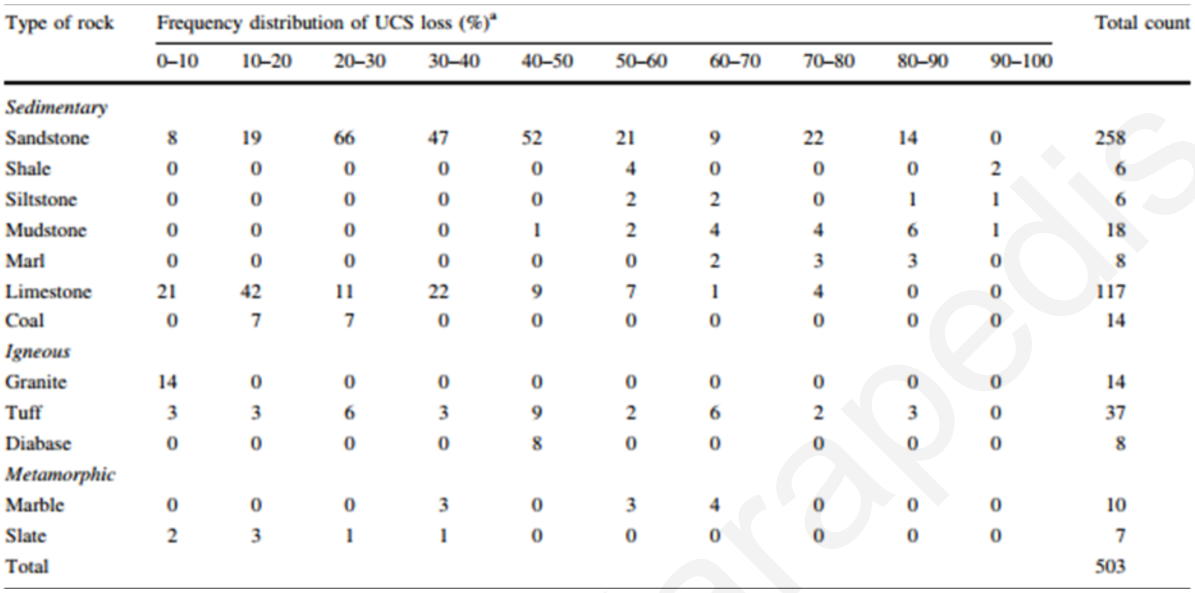
One of the most studied rock property, if not the most studied one, is its compressive strength, especially the Unconfined Compressive Strength (UCS), and in a lesser degree, the Triaxial Compressive Strength (TCS). Compared to soil materials, most rocks present greater strength. But, as in soils, the presence of water in rock body can greatly affect its strength. In 1946, Obert et al., (it is available online-given in literature) performed tests on 6 different rocks (marble, limestone, granite, slate and two sandstones) in three wetting conditions: air dried, oven dried and saturated. They found that the oven-dried samples showed the largest compressive strength, followed by the air-dried ones, while the saturated samples showed the lowest compressive strength. At that time, the authors did not consider the water-weakening effect important, so they proceeded with their tests using only the air-dried samples *"as the value obtained is intermediate to that for the extreme moisture conditions"*.

Throughout the following years, numerous researches worked on the study of water affect in rock strength (Burshtein, 1969; Erguler & Ulusay, 2009; Hadizadeht & Lawi, 1991; Hawkins & Mcconnell, 1992; Van Eeckhout, 1976; Wasantha & Ranjith, 2014; Zhao et al., 2021). Researchers performed tests on many rock types, using different test methods and procedures. In hardly every relative experiment, the trend for rock strength to reduce after the presence of water, was confirmed.

Wong et al. (2016) issued a review in which, among other, they summarized the results of previous tests on different rock types. In Table 1, their findings on the UCS loss from dry-state to saturation are summarized. From Table 1 and Figure 1, one can see that the fluctuation of UCS loss is great, even between rocks of the same type (e.g. sandstones and tuffs). This fact, proves that trying to generalize regarding the UCS loss in rocks due to the presence of moisture is risky, because this seems to depend on many factors (like mineralogy, structure, geometry etc.) (Atkinson & Meredith, 1981; Cai et al., 2019; Hadizadeht & Law, 1991; Lin et al., 2005; Van

Eeckhout, 1976), which show a great difference between different rocks. Thus, more focused investigations of the mechanisms affecting the water weakening effect, would be enlightening.

Table 1 - Summarized results on UCS loss for different rock types (from Wong et al. (2016))



Type of rock	Frequency distribution of UCS loss (%)ª										Total count
	0–10	10–20	20–30	30–40	40–50	50–60	60–70	70–80	80–90	90–100	
Sedimentary											
Sandstone	8	19	66	47	52	21	9	22	14	0	258
Shale	0	0	0	0	0	4	0	0	0	2	6
Siltstone	0	0	0	0	0	2	2	0	1	1	6
Mudstone	0	0	0	0	1	2	4	4	6	1	18
Marl	0	0	0	0	0	0	2	3	3	0	8
Limestone	21	42	11	22	9	7	1	4	0	0	117
Coal	0	7	7	0	0	0	0	0	0	0	14
Igneous											
Granite	14	0	0	0	0	0	0	0	0	0	14
Tuff	3	3	6	3	9	2	6	2	3	0	37
Diabase	0	0	0	0	8	0	0	0	0	0	8
Metamorphic											
Marble	0	0	0	3	0	3	4	0	0	0	10
Slate	2	3	1	1	0	0	0	0	0	0	7
Total											503

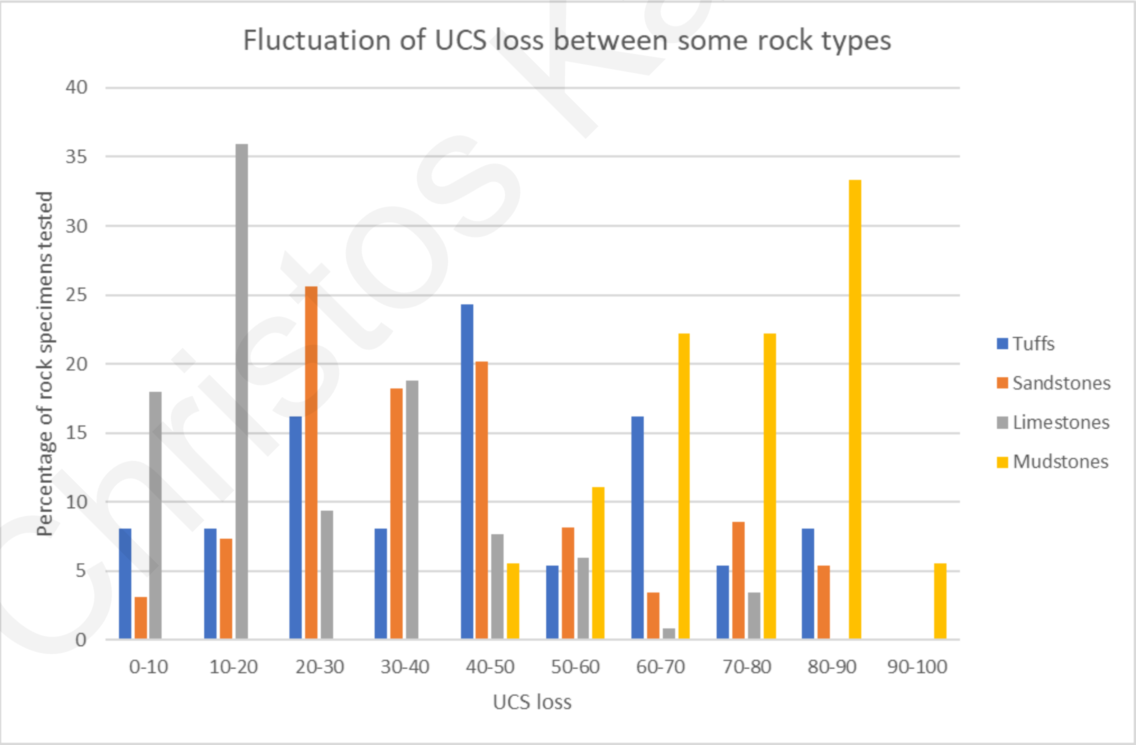


Figure 1 - Fluctuation of UCS loss for some rock types. Data obtained from Wong et al. (2016)

The present essay focuses on the influence that clay minerals have in UCS loss in rocks. Clay minerals are very reactive with water, and thus, their wet behavior is constantly under investigation and concern many engineering applications, such as foundation and underground structures design, underground hydraulics, historical monuments preservation, etc. Clay reactivity also concerns the field of rock mechanics, since clay minerals are present in almost every rock type, and in many cases, have the role of cementing material, i.e., the material that keeps rock grains attached. Thus, the study of their behavior into rocks, especially at wet state, has attracted the interest of researchers, in order to explain wet rock behavior (Aksu et al., 2015; Jiménez-González et al., 2008; B. Li et al., 2019; Lu et al., 2017).

In Chapter 2, the basics of clay minerals will be presented, such as its structure, water sorptivity, the behavior after hydration and its following effects on their properties. The point of this chapter is to provide readers with no relative background, the basic knowledge of clay mineralogy, in order to understand the different ways clay affect rock strength.

In Chapter 3 and Chapter 4, two rock groups are studied. The first group is mudstones, siltstones and marls, where clay minerals are dominant, and the second one refers to sandstones, where clay minerals can be found in proportions from 0% to 20-25%. The purpose of this separation, is to investigate the different behavior that these two groups exhibit after saturation, to discuss about the role clay has in each case and to extract some basic conclusion about the impact of clay minerals in wet UCS. However, this chapter could also be useful as a general review of these two rock groups behavior after saturation, not only from clay mineral concern.

In Chapter 5, the basic conclusion extracted from the aforementioned research are presented.

In addition, in the Appendixes, a brief introduction in other mechanism affecting the wet UCS of rocks, is given. Specifically, Appendix 1 presents how quartz reacts with water and its impact on crack propagation, the reactivity of calcite with water is summarized in Appendix 2, while the behavior of pore water itself is concerned in Appendixes 3, 4 and 5, e.g. water distribution in rock samples, pore water pressure under loading and capillary phenomena.

Please note that the present thesis deals with common environmental conditions, like water and air temperature, or the overburden pressure. Thus, any reference or data used refer to such conditions; the behavior of clay or rocks under high temperatures or under high confining pressures, as these exhibited in geological applications, are not part of the present thesis.

Chapter 2 Basics of clay minerals

There is no global definition of what is called a “clay mineral”. According to Guggenheim & Martin (1995), *the term “clay mineral” refers to phyllosilicate minerals and to minerals which impart plasticity to clay and which harden upon drying or firing”.* The term *“phyllosilicate”* (or *“layer silicate”* or *“sheet silicate”*) refers to the nature of the structure of the clay minerals, which consists of different sheets of cations -tetrahedra and octahedra- more commonly of Silica (Si), Aluminium (Al) and Magnesium (Mg), that form layers. Because of their structure, clay minerals exhibit a platy or flaky shape (Barton & Karathanasis, 2002; Guggenheim, n.d.). Figure 2 shows a SEM picture of the clay mineral kaolinite, where the alternate flaky layers can be seen.

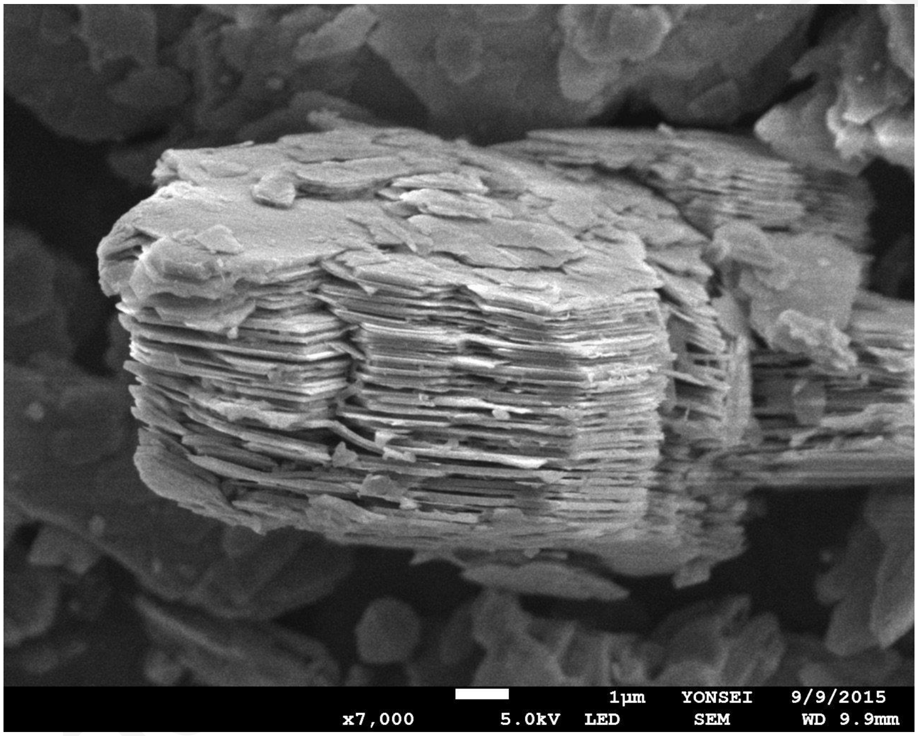


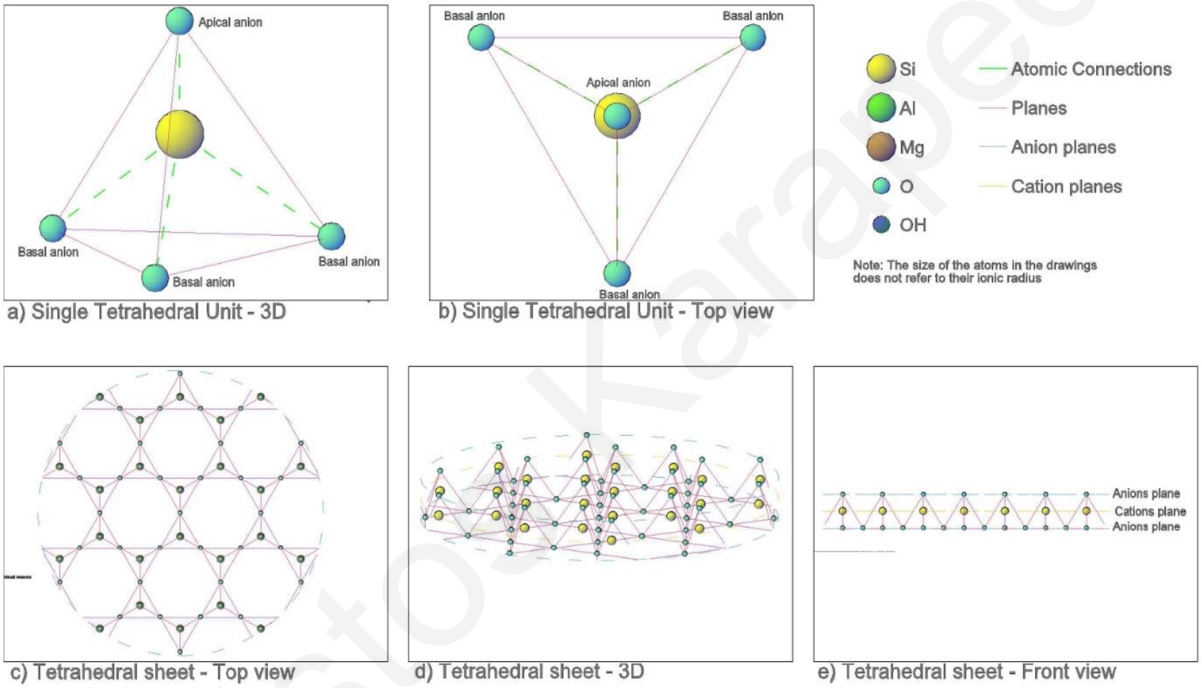
Figure 2 – Ordinary kaolinite under an electron microscope. Credit: Yonsei University, Yongjae Lee, Accessed 05 July 2022 <https://phys.org/news/2017-11-clay-mineral-earth-mantle.html>

2.1 Structure

2.1.1 Tetrahedral units and sheets

As shown in Figure 3a-b, a single tetrahedral unit consists of one Silicon cation (Si^{4+}) surrounded by four Oxygen anions (O^{2-}). Oxygen atoms can be considered as the edges of a tetrahedron, whilst the planes defined by them as the sides of it. From this group of atoms, three out of four oxygen anions are also shared with other tetrahedral units, with each of those oxygen atoms shared among two tetrahedra, while the remaining one is not. The shared oxygen atoms are known as “basal”, whilst the non-shared one is known as “apical” (Kumari & Mohan, n.d.). The sharing of the oxygen atoms among different tetrahedra leads to the formation of a tetrahedral sheet (Figure 3c-e). Note how a hexagonal network is formed (Figure 3c), because every basal oxygen atom belongs to two tetrahedra.

In terms of electric charge, a single tetrahedral unit in a tetrahedral sheet is negatively charged. This is due to the fact that basal anions are shared with other tetrahedra, while the apical anion is not; so, the negative electric charge is 3 X 1^- + 1 X 2^- = 5^- while the positive is 4^+, leading to an electric charge of 1^-.

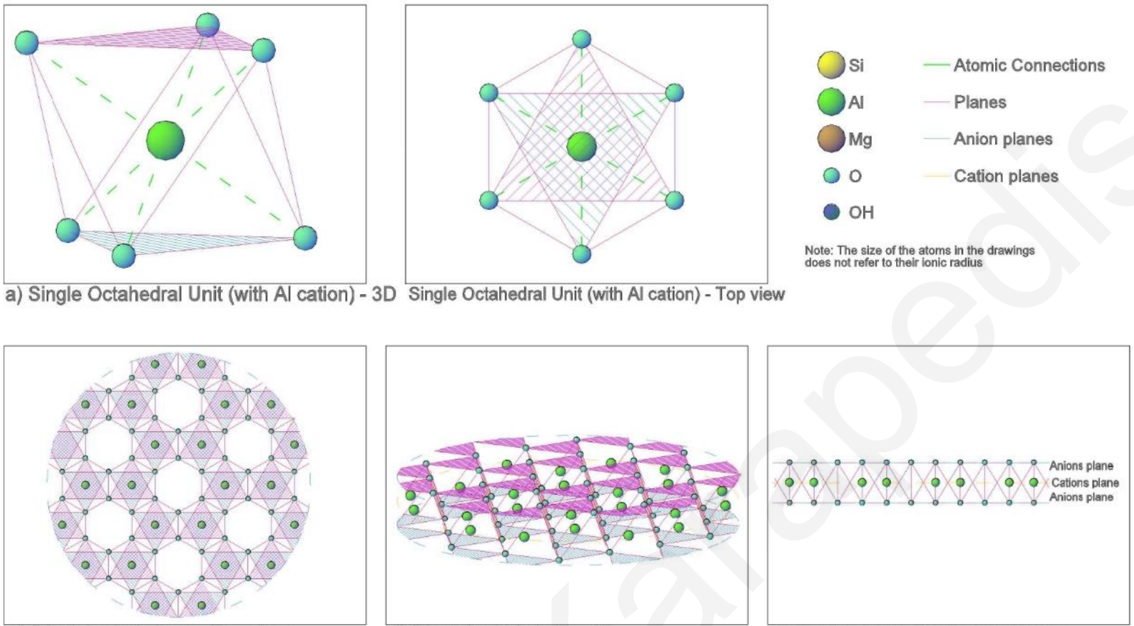


a) Single Tetrahedral Unit - 3D
b) Single Tetrahedral Unit - Top view
c) Tetrahedral sheet - Top view
d) Tetrahedral sheet - 3D
e) Tetrahedral sheet - Front view

Figure 3 – Single tetrahedral unit and tetrahedral sheet

2.1.2 Octahedral units and sheets

A single octahedral unit is formed by one Aluminium (Al^{3+}) or Magnesium (Mg^{3+}) cation, surrounded by six Oxygen (O^{2-}) or Hydroxyl (HO^{1-}) anions (Figure 4a-b and Figure 5a-b). Sometimes, the cation can be Iron (Fe^{2+}) (Al Ani & Sarapaa, 2008; Hillier, n.d.). Here again, oxygen or hydroxyl anions can be considered as the edges of the octahedron, and the planes defined by them, as the sides of it. All the anions of an octahedral unit are shared with other octahedral units, forming an octahedral sheet. Depending on the valency of the dominating cation, and in order to maintain an electric neutrality (Barton & Karathanasis, 2002; Hillier, n.d.), the octahedral sheet can be dioctahedral in the case of trivalent cations (such as Al^{3+}) or trioctahedral in the case of divalent cations (such as Mg^{3+} or Fe^{2+}). In a dioctahedral sheet (Figure 4c-e) every anion is shared by two octahedral units, while in a trioctahedral sheet (Figure 5c-e) every anion is shared by three octahedral units.

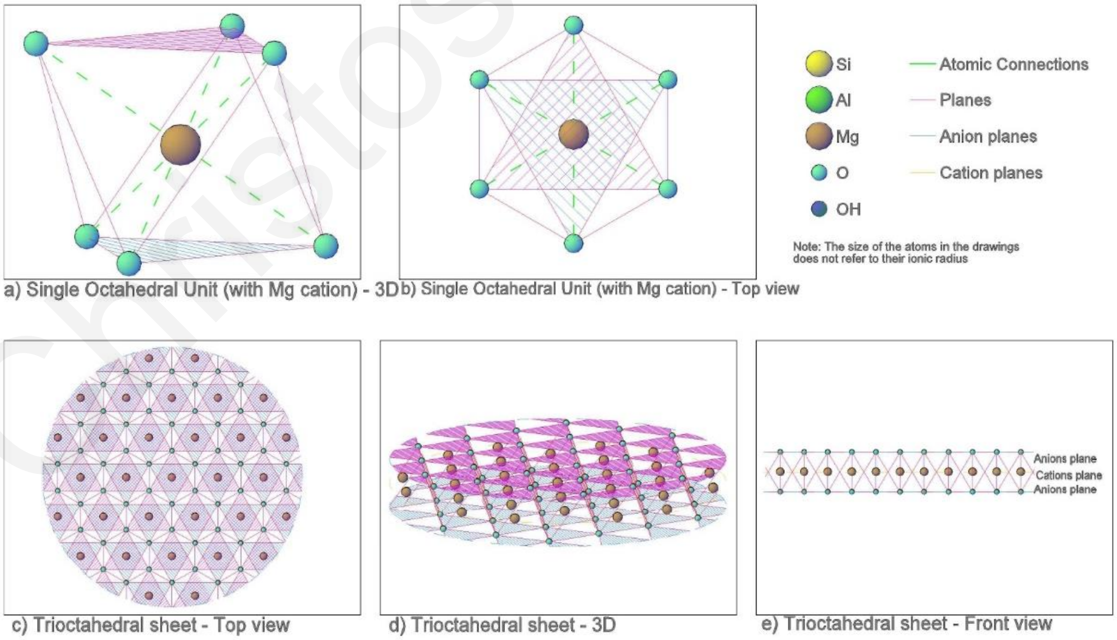


a) Single Octahedral Unit (with Al cation) - 3D Single Octahedral Unit (with Al cation) - Top view

c) Dioctahedral sheet - Top view d) Dioctahedral sheet - 3D e) Dioctahedral sheet - Front view

Note: The plane in magenta is an anion plane. A different color was chosen for visual reasons

Figure 4 – Single octahedral unit (with Al cation) and dioctahedral sheet



a) Single Octahedral Unit (with Mg cation) - 3D b) Single Octahedral Unit (with Mg cation) - Top view

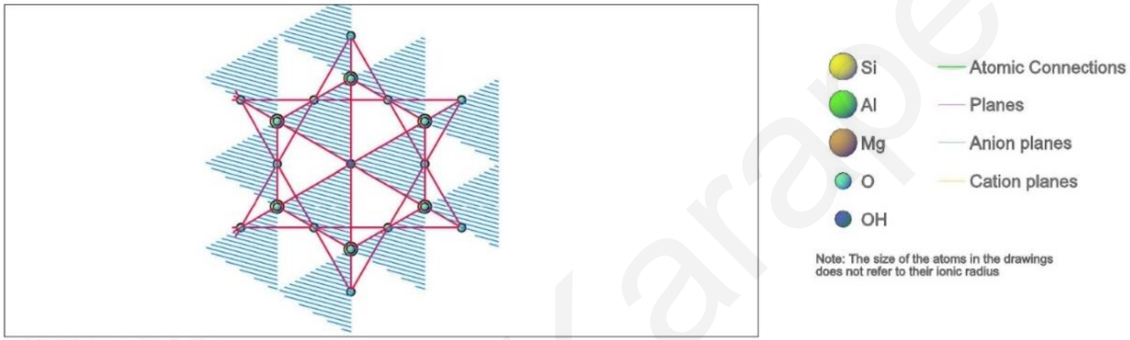
c) Trioctahedral sheet - Top view d) Trioctahedral sheet - 3D e) Trioctahedral sheet - Front view

Note: The plane in magenta is an anion plane. A different color was chosen for visual reasons

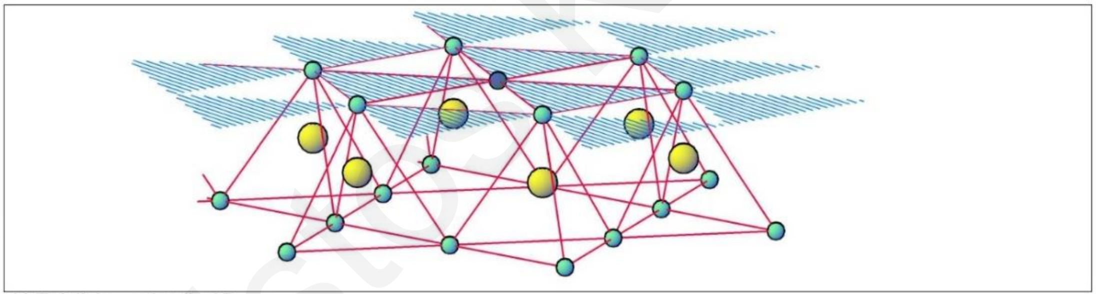
Figure 5 – Single octahedral unit (with Mg cation) and trioctahedral sheet

2.1.3 Layer formation

The lateral dimensions of tetrahedral and octahedral sheets are approximately equal; so, it is possible for them to join (Guggenheim, n.d.), even though some structural adjustments are needed in order to achieve the linkage (Hillier, n.d.). This linkage is achieved by oxygen sharing; apical oxygens of the tetrahedra are shared with two out of three oxygen atoms of the octahedra (Figure 6). The remaining anion is a hydroxyl, which cannot be shared between the two sheets (Guggenheim, n.d.; Hillier, n.d.), and is located at the center of the hexagonal network of the tetrahedron, in the apical oxygen plane (Christidis, 2011; Hillier, n.d.). This way, an aluminosilicate layer is formed.



a) T-O linkage detail - Top

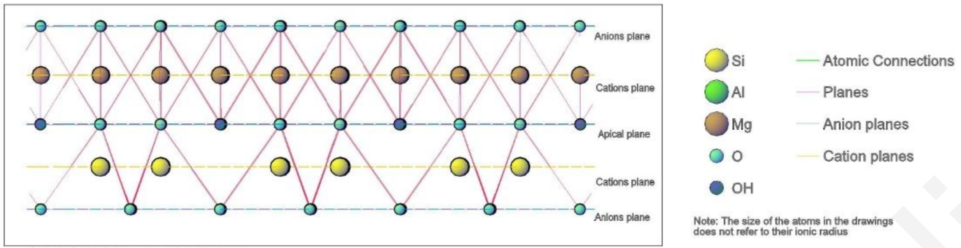


b) T-O linkage detail - 3D

Note: Planes with blue represent the anion plane of the octahedron

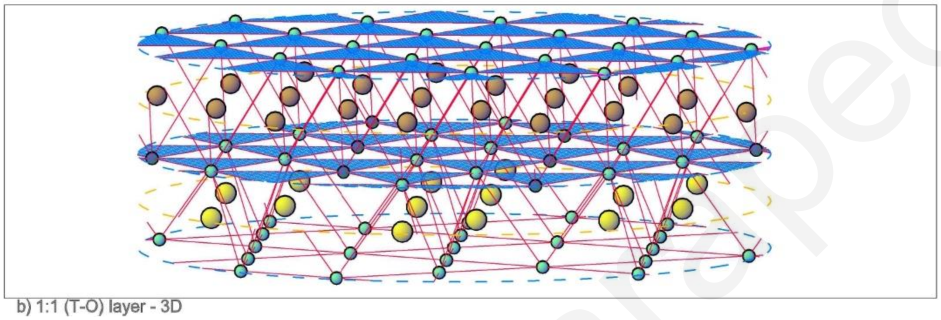
Figure 6 - Tetrahedral and Octahedral linkage detail

Tetrahedral and octahedral sheets may link in two ways. One way is for one tetrahedral sheet to link with one octahedral sheet, forming the what is called a 1:1 (or T-O) layer (Figure 7), and the other is for two tetrahedral sheets to link with one octahedral sheet, forming a 2:1 (or T-O-T) layer (Figure 8). In the last case, the tetrahedra are linked to the opposite surfaces of the octahedra. Figure 9 shows a simplified way to illustrate tetrahedral and octahedral sheets, and the consequential way to illustrate 1:1 and 2:1 layer. In a clay particle, 1:1 or 2:1 layer is placed together in the horizontal and vertical dimension. Two layers, one over the other, are bonded through hydrogen or Van der Waals bonds and the space between them is called an "interlayer". A structural unit is the sum of a layer and an interlayer (Christidis, 2011). Figure 10 is relevant.



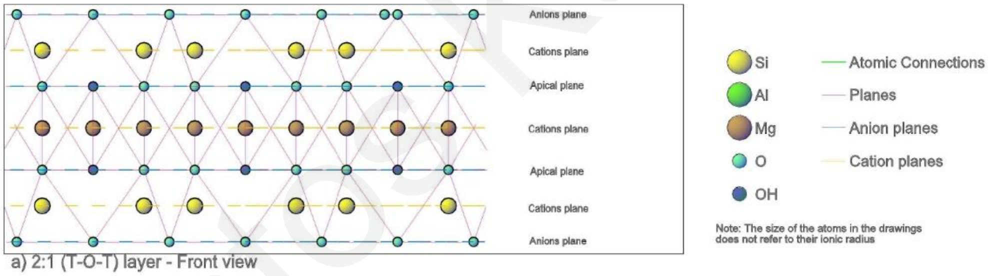
Note: The size of the atoms in the drawings does not refer to their ionic radius

a) 1:1 (T-O) layer - Front view



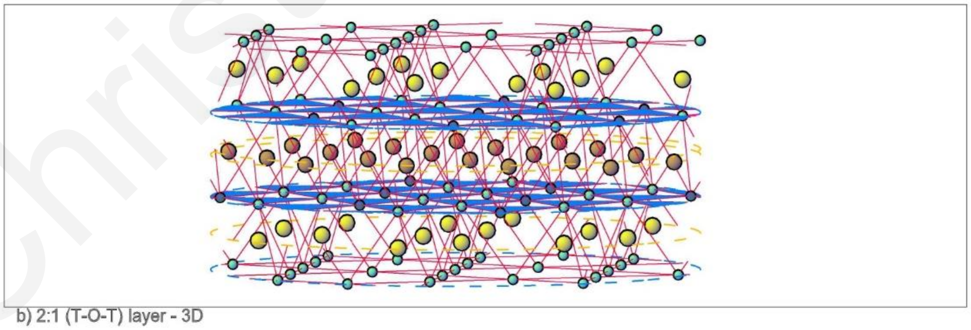
b) 1:1 (T-O) layer - 3D

Figure 7 - 1:1 (T-O) layer



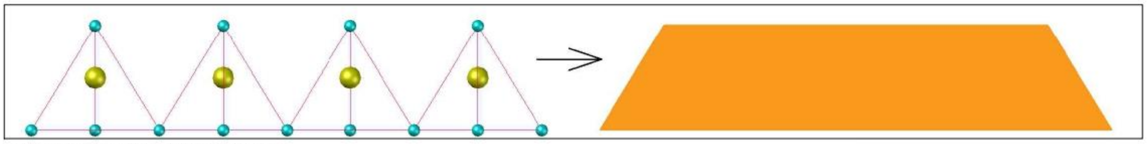
Note: The size of the atoms in the drawings does not refer to their ionic radius

a) 2:1 (T-O-T) layer - Front view

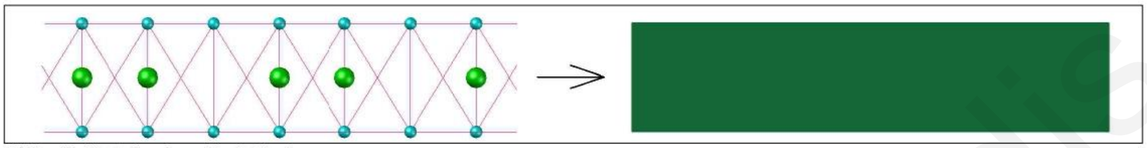


b) 2:1 (T-O-T) layer - 3D

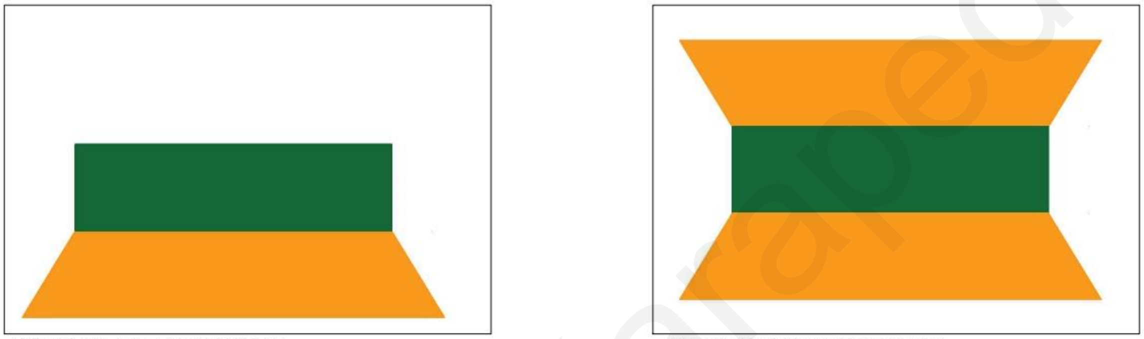
Figure 8 - 2:1 (T-O-T) layer



a) Simplified illustration of a tetrahedral sheet



b) Simplified illustration of an octahedral sheet



c) Simplified illustration of an 1:1 (T-O) layer

d) Simplified illustration of a 2:1 (T-O-T) layer

Figure 9 - Simplified illustrations of sheets and layers

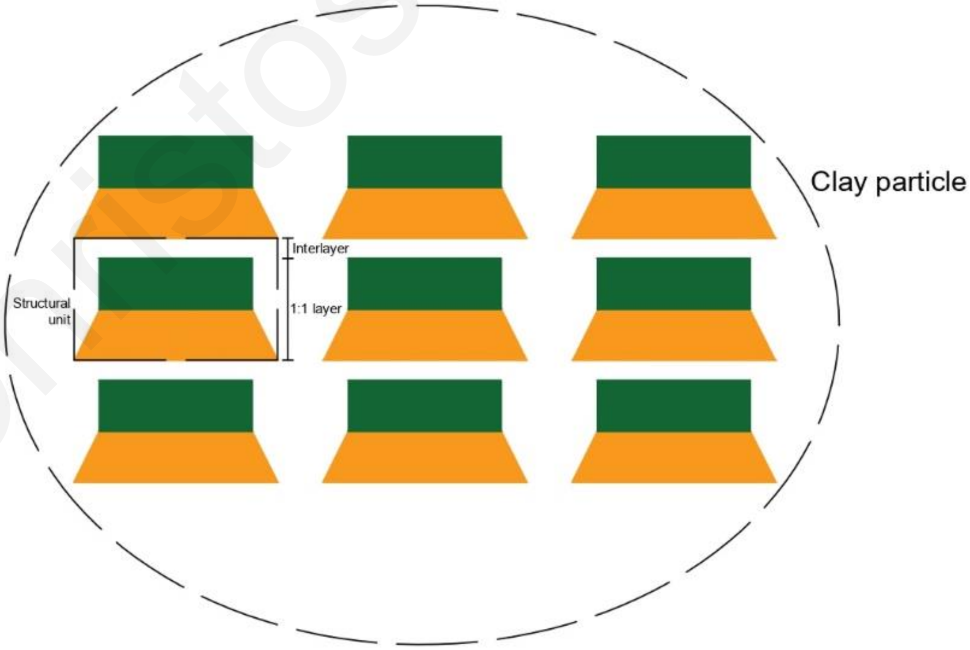


Figure 10 - Schematic representation of related terms

2.2 *Electric charge of layers*

As stated above, octahedral units connect with each other in order to keep all single unit neutral, while the tetrahedral sheet's negative charge is neutralized after the linkage with an octahedral sheet. However, it is possible for cations to replace each other, as long as the ionic radii of the two cations are similar, so that no different coordination on the replacing cation is needed and no disruption on the sheets will occur (Al Ani & Sarapaa, 2008). In tetrahedra, the usual substitution is for Al^{3+} and Fe^{3+} to replace Si^{4+}, while in octahedra, the usual substitution is for Fe^{3+}, Fe^{2+}, Zn^{2+} to replace Al^{3+} and Mg^{2+} (Al Ani & Sarapaa, 2008; Kumari & Mohan, n.d.). After that, and since the structure of the units does not change, deficiencies in electrical charge of the layers occur, leading to negative or positive electric charge of layers. In 1:1 layers, the substitutions that take place are usually in that manner, and the overall charge of the layers is still neutral (Hillier, n.d.). On the other hand, substitutions in 2:1 layers usually lead to an overall negative charge (Barton & Karathanasis, 2002; Hillier, n.d.).

Following that, attraction of cations on layer surfaces (interlayer) may happen, in order to keep the total layer charge neutral. The quantity of the needed cations depends on the initial negative charge of the layers and the specific surface area of the minerals, and is expressed by the Cation Exchange Capacity. Such cations may be Calcium (Ca^{2+}), Sodium (Na^{1+}), Magnesium (Mg^{2+}) or Potassium (K^{1+}), or even hydrous cations (i.e. cations with water molecules attached) (Christidis, 2011; Guggenheim, n.d.). The latter is of significant value, because some clay minerals react intensely under the presence of water, giving rise to very important phenomena, such as swelling/shrinkage behavior.

2.3 *Classification of clay minerals*

The classification of phyllosilicate clay minerals takes into consideration (i) the layer type, (ii) the nature of the octahedral cation, (iii) the magnitude of the net negative charge and (iv) the interlayer cation. Following that, clay minerals are separated in three categories depending on their layer type (1:1, 2:1 and 2:1:1), each of those categories containing groups of similar layer charge. Each group is divided into two subgroups, one containing minerals with dioctahedral sheets and the other minerals with trioctahedral sheets. Table 2 presents the most important clay minerals, following the aforementioned classification.

Table 2 also includes the Sepiolite-palygorskite group, which is a fibrous clay mineral group. It differs from the phyllosilicate group in two ways: (a) in each tetrahedral sheet, under a specific number of units, rotation of apical oxygen takes place and (b) the octahedral sheets are continuous in two dimensions only. Due to their structure, minerals included in this group are fiber-like.

Table 2 - Clay minerals: Classification and Interlayer cation

Layer type	Electric charge	Group	Subgroup	Mineral	Interlayer cations	References
1:1	q≈0	Kaolin-Serpentine	Kaoline	Kaolinite	None	Machida et al., 2021
				Halloysite	Water molecules	Haq et al., 2008
				others: Dickite, Nacrite		
			Serpentine	Lizardite	None	Mellini, 1982
				others: Antigorite, Chrysotile		
2:1	q≈0	Pyrophyllite-talc	Pyrophyllite	Pyrophyllite	None	Drits et al., 2012 and Sakuma and Suehara, 2015
			Talc	Talc	None	Drits et al., 2012 and Sakuma and Suehara, 2016
	0.2 < q < 0.6	Smectite	Di. Smectite	Montmorillonite	Hydrated cations (such as Na^{1+}, Ca^{2+}, K^{1+}, Cs^{1+})	*Yotsuji et al., 2021 and ** Uddin, 2018
				others: Beidellite, Nontronite		
			Tri. Smectite	Saponite	Na^{1+}, K^{1+}, Rb^{1+}, Nh4, $0.5Ca^{2+}$, $0.5Ba^{2+}$, $0.5Mg^{2+}$, $0.33Al^{3+}$	Kloprogge and Ponce, 2021
				others: Hectorite, Stevensite, Sauconite		
	0.6 < q < 0.9	Verrmiculite	Di. Vermiculite	Vermiculite	Mostly Hydrated Mg^{2+}, also Ca^{2+}, Na^{1+}, K^{1+}	Valaskova dn Martynkova, 2014
			Tri. Vermiculite	Vermiculite	Mostly Hydrated Mg^{2+}, also Ca^{2+}, Na^{1+}, K^{1+}	Valaskova dn Martynkova, 2014
	q≈1	Micas	Di. Mica	Illite	K^{1+}	Ghasemi and Sharifi, 2021
				Muscovite	K^{1+}	Sakuma and Kawamura, 2009
				others: Boromuscovite, Glauconite, Paragonite		
			Tri. Mica	Biotite	K^{1+} and Na^{1+}	Min et al., 2018
				others: Phlogopite, Lepidolite		
2:1:1	q: varies	Chlorite	Di. Chlorites	Members: Donbassite	Gibbsite-like sheet	Eggleston and Bailey, 1967*
			Di/Tri. Chlorites	Members: Cookeite, Sudoite	Brucite-like sheet	Eggleston and Bailey, 1968*
			Tri. Chlorites	Members: Clinclore, Nimite	Brucite-type octahedral layer	Zanazzi et al., 2006*
Fibrous clays (2:1 inverted)	q: varies	Sepiolite-palygorskite	Sepiolite	Sepiolite	-	Post et al., 2007
			Palygorskite	Palygorskite	-	Chen et al., 2021

*Articles refere to the subgroups and not to each mineral of the subgroup separately

2.4 Swelling/shrinkage

When clay minerals get in touch with water (liquid or vapor), they show a tendency to increase their volume, while when water is expelled, decrease of volume is observed. This phenomenon is known as swelling/shrinkage, and is activated as soon as clay particles contact water molecules (Aksu et al., 2015). The swelling and shrinkage behavior of clay minerals is attributed to the absorption of water molecules between their layers and particles. Figure 11 shows the swelling of a clay fraction in a porous medium and Figure 12 presents a soil sample after suffering swelling and shrinkage. The purpose of this sub-section is to present the basics of the swelling/shrinkage phenomenon and to highlight the most important factors affecting its intensity, which are related to the scope of the present thesis.

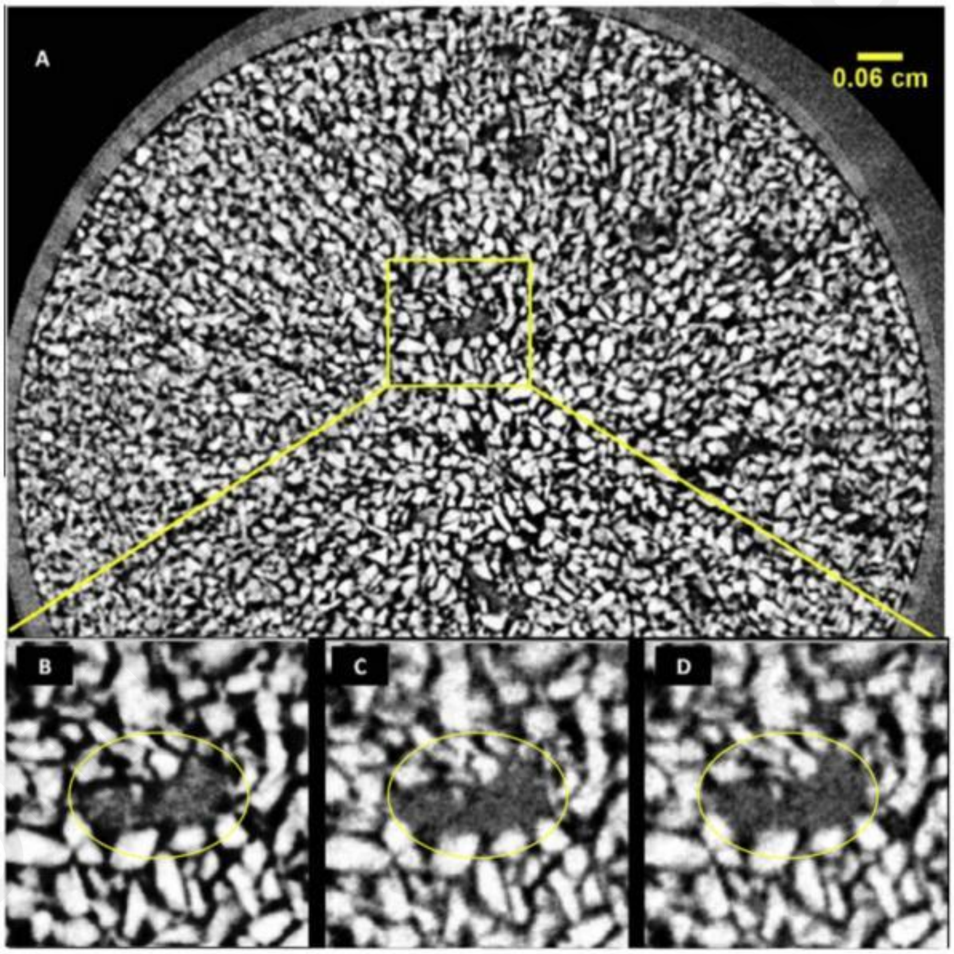


Figure 11 - Swelling of a clay fraction in a synthetic porous media. A) Region of interest, (B) Clay fraction in dry condition, (C) Clay fraction in saturated condition and (D) Clay fraction after 4 hours of saturation (from Aksu et al. (2015))

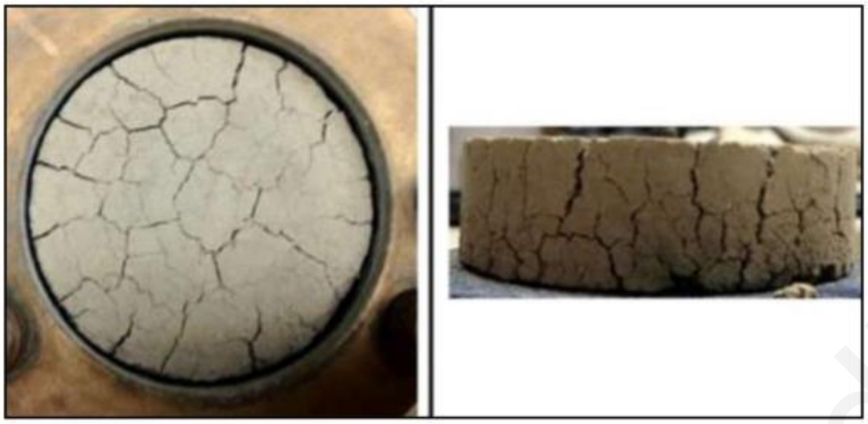
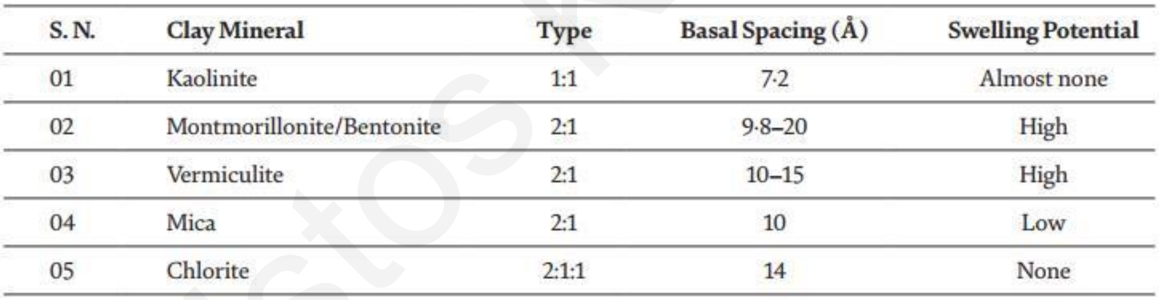


Figure 12 - Top view (left) and side view (right) of a soil sample after shrinkage (from Firoozi et al. (2016))

Generally, almost every clay mineral may suffer from swelling, but not all clay minerals exhibit the same degree of swelling. Table 3 presents the swelling potential of the basic clay mineral groups, after Kumari & Mohan (n.d.) . According to authors, smectite and vermiculite groups present the highest swelling potential, micas show a low swelling potential, while kaolinite presents almost no swelling behavior. Chlorite is supposed to be non-expandable.

Table 3 - Swelling potential of clay mineral groups (from Kumari & Mohan (n.d.))



S. N.	Clay Mineral	Type	Basal Spacing (Å)	Swelling Potential
01	Kaolinite	1:1	7·2	Almost none
02	Montmorillonite/Bentonite	2:1	9·8–20	High
03	Vermiculite	2:1	10–15	High
04	Mica	2:1	10	Low
05	Chlorite	2:1:1	14	None

Two mechanisms of swelling activation have been found: (a) Crystalline swelling and (b) osmotic swelling (Madsen & Muller-Vonmoos, 1989):

Crystalline swelling: As mentioned above, some clay minerals exhibit negative electric charge on their layer surfaces, resulting in the attraction of cations into the interlayer space. Some clay mineral groups attract hydrous cations (see Table 2), due to their electric polarity, resulting in the formation of a number of water layers around them. The formation of water layers around interlayer cations pushes the layers apart, resulting in the increase of interlayer space and of the bulk volume of clay particle. This mechanism is known as *crystalline swelling* (Figure 13).

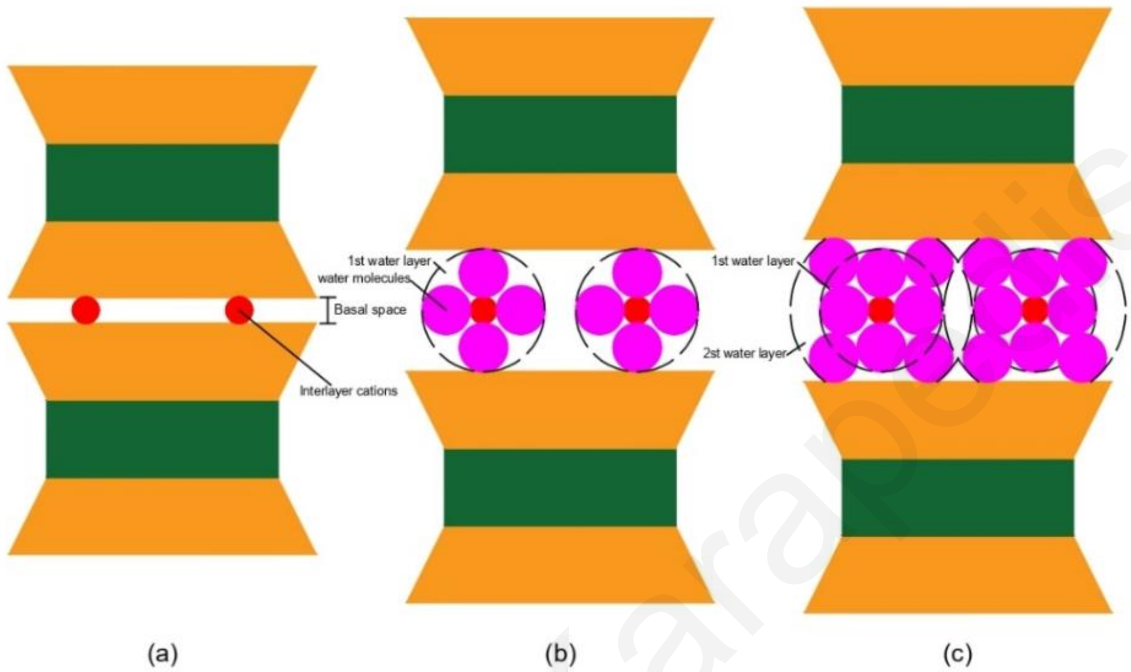


Figure 13 - Crystalline swelling in 2:1 clay mineral. (a) interlayer with unhydrous cation, (b & c) interlayer with 1 and 2 water layers, respectively

The intensity of crystalline swelling depends on many factors, such as the bonding strength between the layers (Elbrahmi & Zoukaghe, 2016), the charge density of clay surfaces (Alcázar-Vara & Cortés-Monroy, 2018; Anderson et al., 2010), the hydration energy of cations (Alcázar-Vara & Cortés-Monroy, 2018) and the number of water layers formed around them (Anderson et al., 2010), as well as the cation size (Anderson et al., 2010) and valency (Anderson et al., 2010; Madsen & Muller-Vonmoos, 1989). Therefore, the crystalline swelling behavior of each clay mineral is different; even the same minerals can present different behavior, depending on the interlayer cation. Figure 14 presents data from Sato et al. (1992), in which the different crystalline swelling behavior of montmorillonite, beidellite and nontronite (all part of the smectite group) with different interlayer cation is presented. Figure 15 also presents the behavior of montmorillonite with different interlayer cations, at different temperatures (Morodome & Kawamura, 2009, 2011).

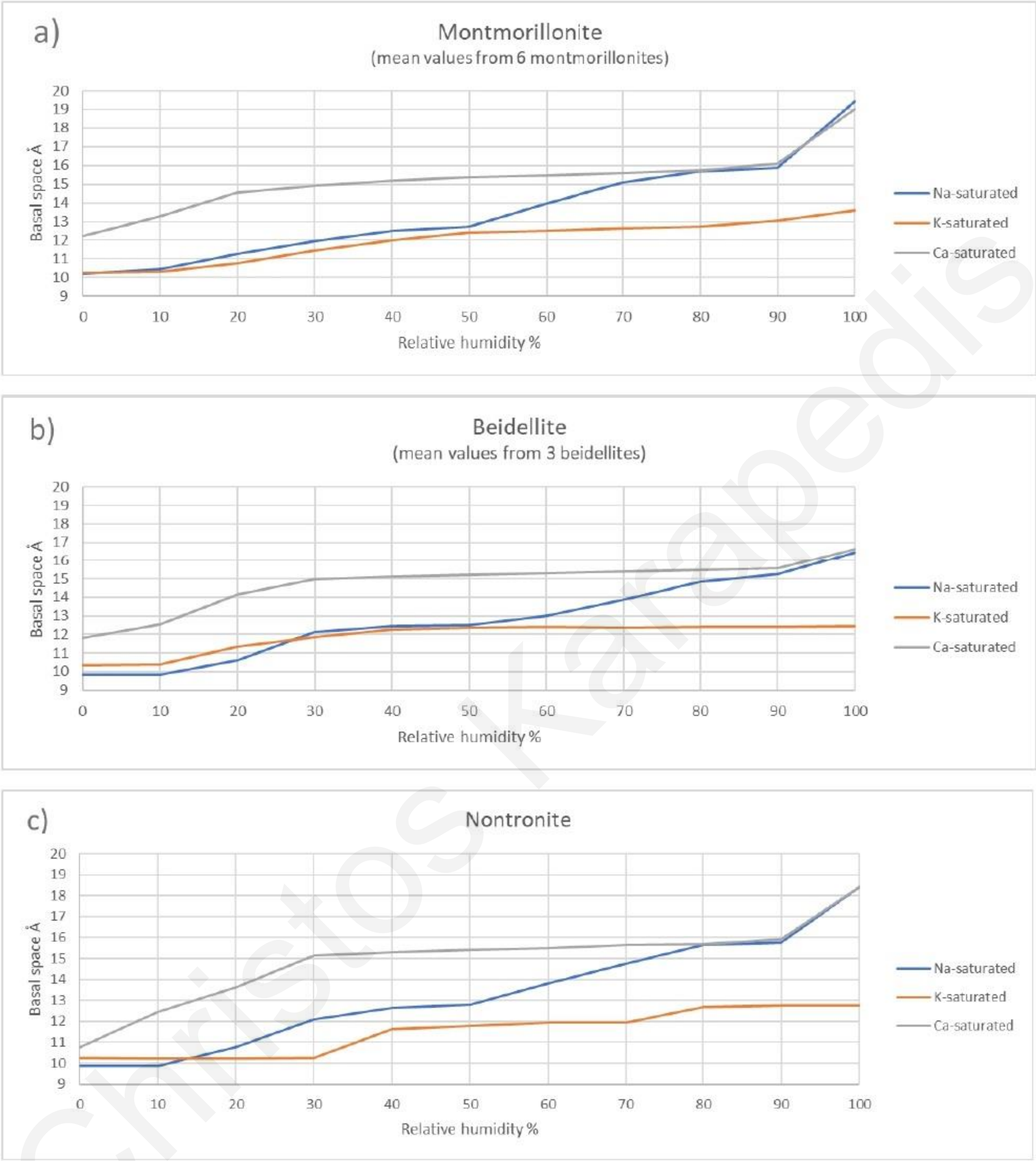


Figure 14 - Basal space vs Relative humidity in smectites. Data obtained from Sato et al. (1992). Graphs in (a) are the mean values of 6 montmorillonites, in (b) are the mean values of 3 beidellites and in (c) of 1 nontronite

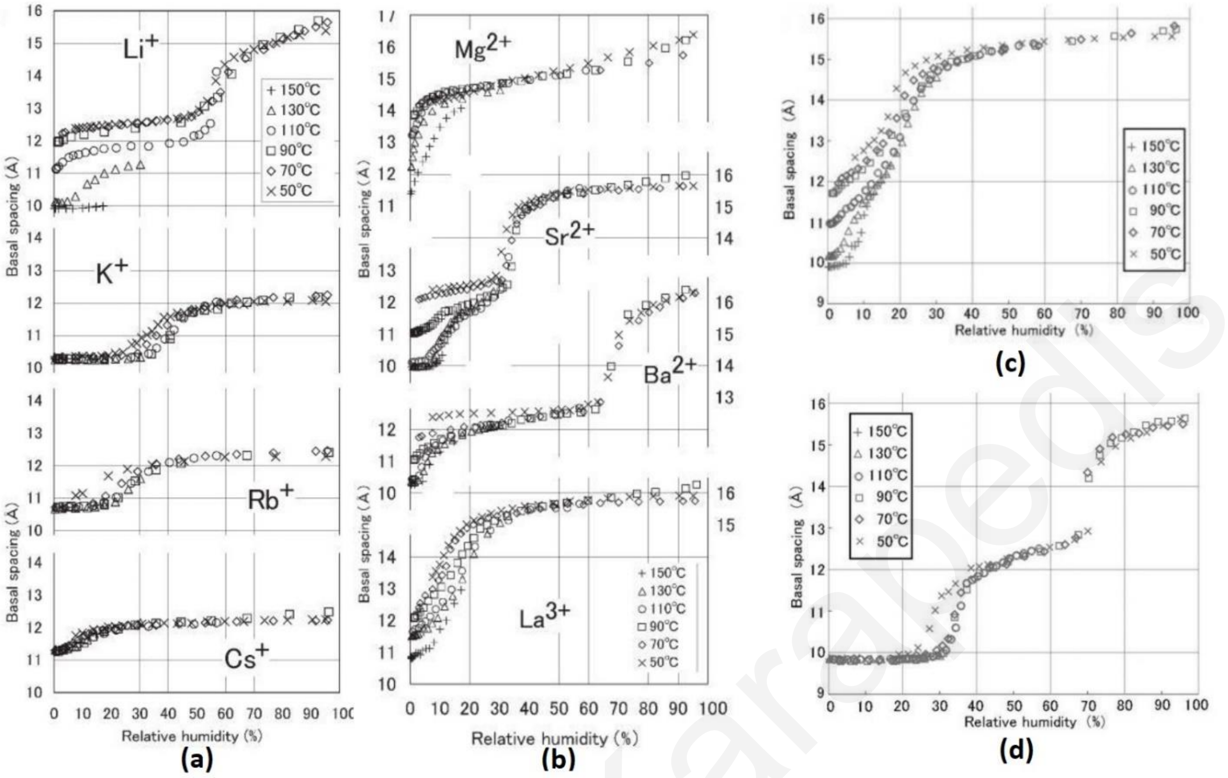


Figure 15 - Basal space vs Relative humidity in montmorillonite, at different temperatures, with different interlayer cations: (a) Li, K, Rb and Cs, (b) Mg, Sr, Ba and La, (c) Ca, (d) Na (from Morodome & Kawamura (2009) and Morodome & Kawamura (2011))

It is observed, that crystalline swelling in montmorillonite is more intense, compared to nontronite and beidellite, which show similar behavior. What is more, it can be stated that the presence of potassium (K), rubidium (Rb) and caesium (Cs), can limit the basal space increase of smectites, while sodium (Na) have the opposite effect. Lithium (Li), magnesium (Mg), strontium (Sr), barium (Ba), Lanthanum (La) and calcium (Ca) may present similar basal space at ~95% RH with sodium, but the basal space at 0% RH is smaller with sodium. Thus, the percentage of crystalline swelling in Na-smectites is larger than that of smectites with other interlayer cations. Similar trends can be observed in molecular simulations on montmorillonite and beidellite (Teich-McGoldrick et al., 2015), in the case of sodium, caesium, calcium and magnesium in the interlayer. The only exception is for the case of caesium, where the basal space at high hydration was found to be higher than the other cations. Another important conclusion, is that, in order for crystalline swelling to occur, there is no need for liquid water; vapor is enough to provoke this type of swelling.

Osmotic swelling: The negative charge of clay mineral particles results in the high concentration of cations close to their surfaces. The concentration of cations decreases with increasing distance from the particle, while the concentration of anions increases. This way, a diffuse double layer is formed, with its limit being where cation and anion concentration begin to be equal. When the diffuse double layers of different clay particles overlap, clay particles repel (Madsen & Muller-Vonmoos, 1989). What is more, in order for the equilibrium of cation concentration to be achieved, water molecules diffuse into the interlayer space, to dilute its ion concentration (Alcázar-Vara & Cortés-Monroy, 2018). With both ways combined, repulsive forces between clay particles are developed, resulting in what is known as *osmotic swelling*. Figure 16 illustrates osmotic swelling. In contrast with crystalline swelling, osmotic swelling needs water in liquid form in order to occur.

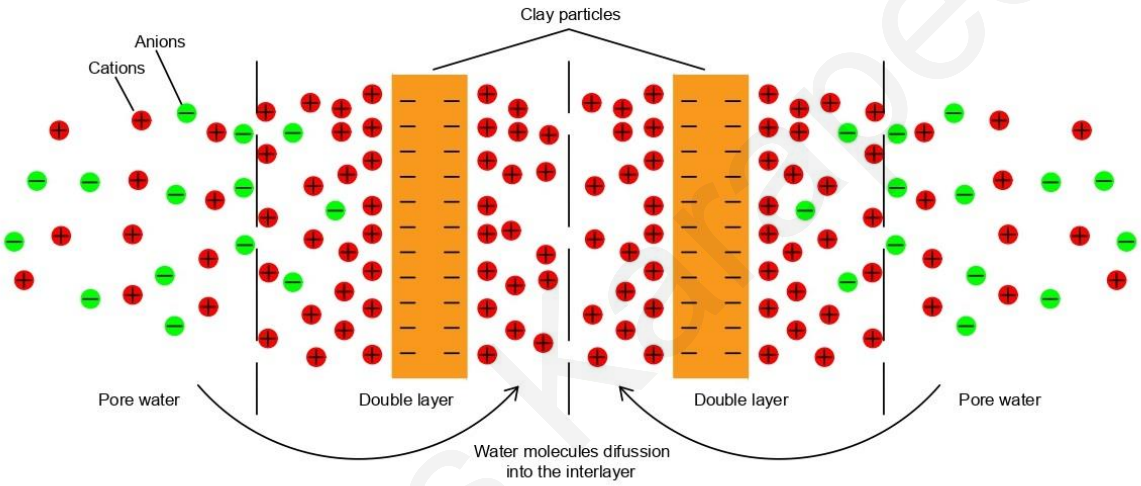
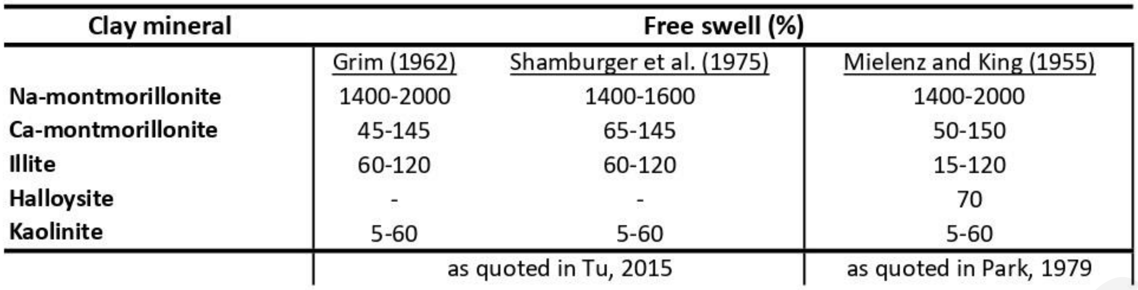


Figure 16 - Osmotic swelling of clay particles

The intensity of osmotic swelling mechanism depends on the charge of clay surfaces, the type and concentration of the cations in the interlayer space and the ionic concentration of pore water. In general, osmotic swelling promotes higher swelling values than crystalline swelling (Alcázar-Vara & Cortés-Monroy, 2018; Elbrahmi & Zoukaghe, 2016). Table 4 presents a general picture of the magnitude of free swelling in montmorillonite, illite, halloysite and kaolinite. Note that, "free swelling" refers to the unconfined swelling under the presence of liquid water; thus, in such measurements, samples exhibited both swelling mechanisms, since liquid water can activate crystalline and osmotic swelling as well.

Table 4 - Free swelling of the most common clay minerals. (from Grim, 1962, Shamburger et al., 1975 and Mielenz and King, 1955 as quoted in Tu (2015) and Park (1979))



Clay mineral	Free swell (%)		
	Grim (1962)	Shamburger et al. (1975)	Mielenz and King (1955)
Na-montmorillonite	1400-2000	1400-1600	1400-2000
Ca-montmorillonite	45-145	65-145	50-150
Illite	60-120	60-120	15-120
Halloysite	-	-	70
Kaolinite	5-60	5-60	5-60
	as quoted in Tu, 2015		as quoted in Park, 1979

Also, the importance of ionic concentration of the wetting liquid can be witnessed in Figure 17, which presents the swelling of a clayey soil, wetted with distilled water and with water and sodium chlorite solution with different cation concentrations. It is observed that as cation concentration of the solution increases, swelling decreases, since less water needs to diffuse between the clay particles in order for ion concentration to get equal. Note that in this case, osmotic swelling occurred only when the soil was saturated with distilled water and 0.1M NaCl solution, since a cation concentration of the soil was 0.25M.

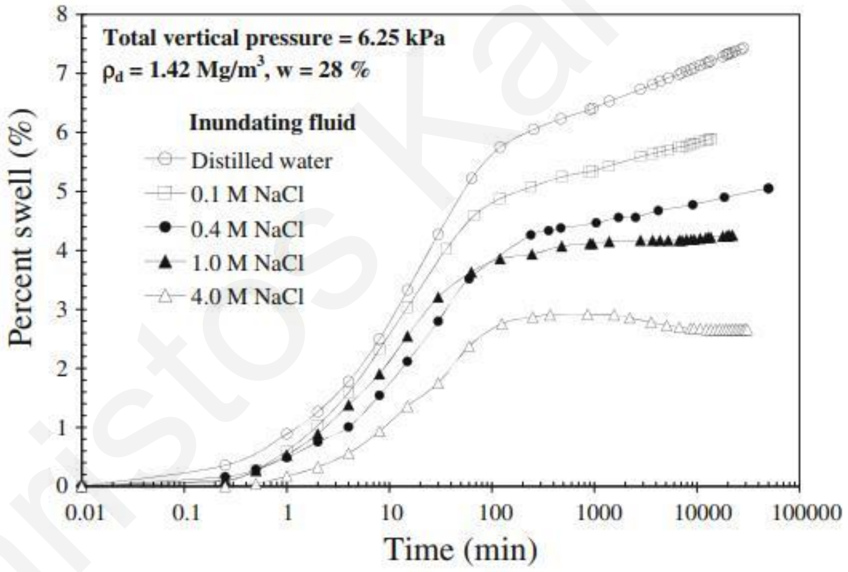


Figure 17 - Swelling of a clayey soil vs time, for solutions with different NaCl concentrations (from Rao et al. (2013))

2.5 Friction loss

Aside from the attraction of water into the interspace, clay minerals attract water molecules on their external surfaces as well (even from air humidity), forming water layers around particles and grains (Hatch et al., 2012). The presence of water layers in the interlayer and the external surface of the clay minerals, causes the friction coefficient (μ) to decrease from dry to wet state. Some reasons have been suggested to attribute this behavior, concentrating on the effect of surface water acting as a lubricant on contact areas (Kawai et al., 2015; Moore & Lockner, 2016), the interlayer water (and the consequential swelling) weakening interlayer bonds (Tetsuka et al., 2018) or a combination, in a manner that interlayer water causes structural deformations, which affect external surfaces of the mineral (Behnsen & Faulkner, 2013; Sakuma et al., 2022).

According to the interlayer space, it has been stated that wet friction coefficient in montmorillonites is related to the cations in the interlayer (Bird, 1984). Behnsen & Faulkner (2013) found that the same montmorillonite presented different friction coefficient values in the wet state, in the cases of potassium, sodium, calcium and magnesium cations in the interlayer, as Figure 18 presents (the role of effective stress will be discussed below). Similar observations were done by Tetsuka et al. (2018), in Na- and Ca- montmorillonites (Figure 19). Both test results are in accordance with the swelling behavior of montmorillonite. Firstly, from Figure 14 and Figure 15 we can see that at high relative humidity (>80%), basal space of Na-, Ca- and Mg- montmorillonites are approximately equal, while for K-montmorillonite is significantly less. Since basal space is related to bonding strength of montmorillonite, it is reasonable that values of friction coefficient in the wet state for Na-, Ca and Mg- montmorillonite are similar, while for K-montmorillonite is greater. Secondly, one can see that at low relative humidity, basal space of Ca-montmorillonite is quite bigger than that of Na-montmorillonite, and thus, the fact that at the same relative humidity Na-montmorillonite shows much greater friction coefficient values than Ca-montmorillonite, makes perfect sense.

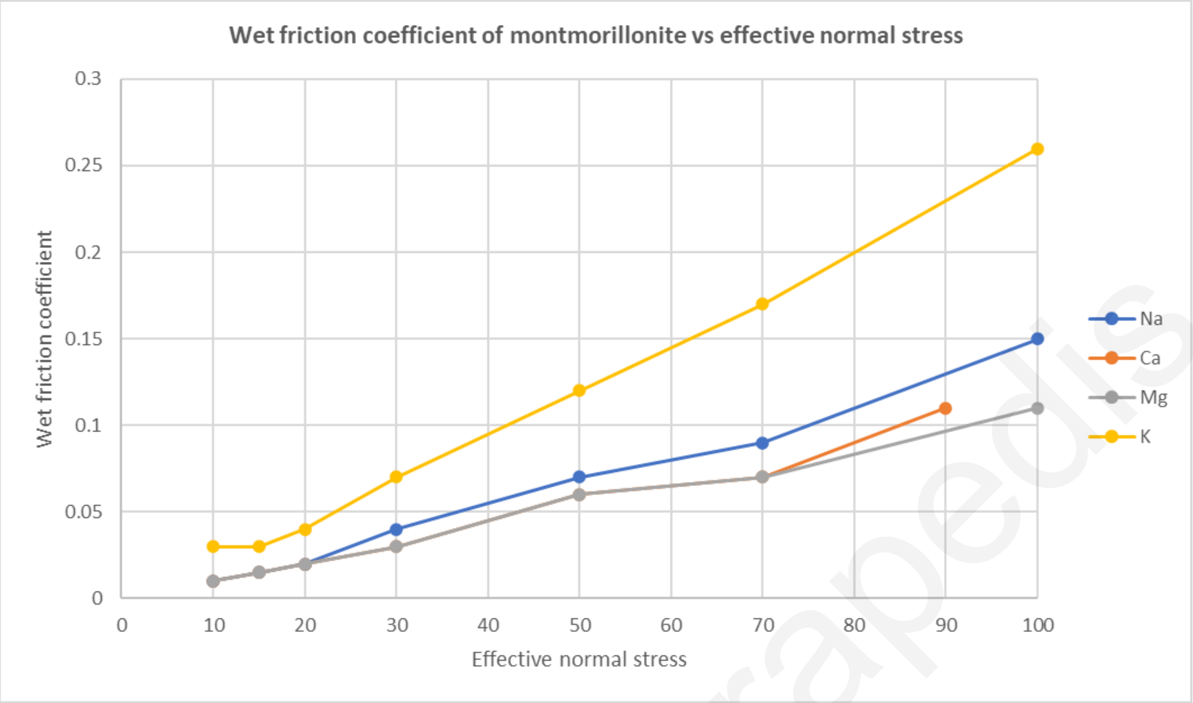


Figure 18 - Wet friction coefficient (μ) of Na-, Ca-, Mg- and K- montmorillonite vs effective normal stress (data from Behnsen & Faulkner (2013))

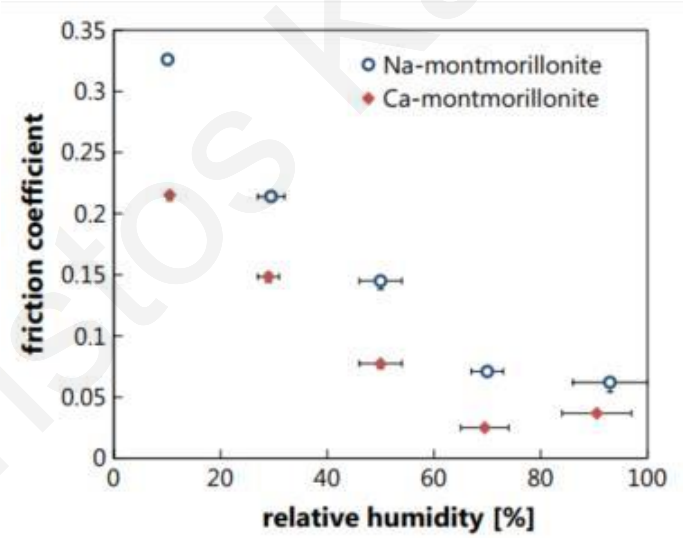


Figure 19 - Friction coefficient of Na- and Ca- montmorillonite vs relative humidity (from Tetsuka et al. (2018))

Another observation from Figure 19, is that for montmorillonite, the wet friction coefficient is significantly affected by the effective normal stress. The higher the effective normal stress, the higher the wet friction coefficient is. Such observations are common among researchers, for most of clay minerals, despite some differentiations regarding the behavior at low effective normal stress. This tendency is attributed to the fact that as effective normal stress arises, water among the particles or in the interlayer is driven off, and so, water films become thinner, reducing their lubrication effect (Moore & Lockner, 2016) and increasing the mineral contact areas (Behnsen & Faulkner, 2012). On the other hand, dry friction is reducing with increasing effective normal stress (Behnsen & Faulkner, 2012; Morrow et al., 2017). Wet and dry friction coefficients on effective normal stress values from 10 to 100Mpa, on Na-montmorillonites are presented in Figure 20. Morrow et al. (2017) performed extensive tests on Na-montmorillonite, for effective normal stress values up to 600-700Mpa and they found that while friction coefficient for dry state stabilizes at approximately 350Mpa, the values for wet state keeps increasing (Figure 21). Figure 22 presents test results on some other clay minerals, performed by Behnsen & Faulkner (2012). The tendency for the dry friction coefficient is to decrease until about 30MPa normal effective stress and then stabilizing, while for the wet state, a reduction from 5MPa to approximately 30MPa effective stress is observed and from that point and on, the values increase again.

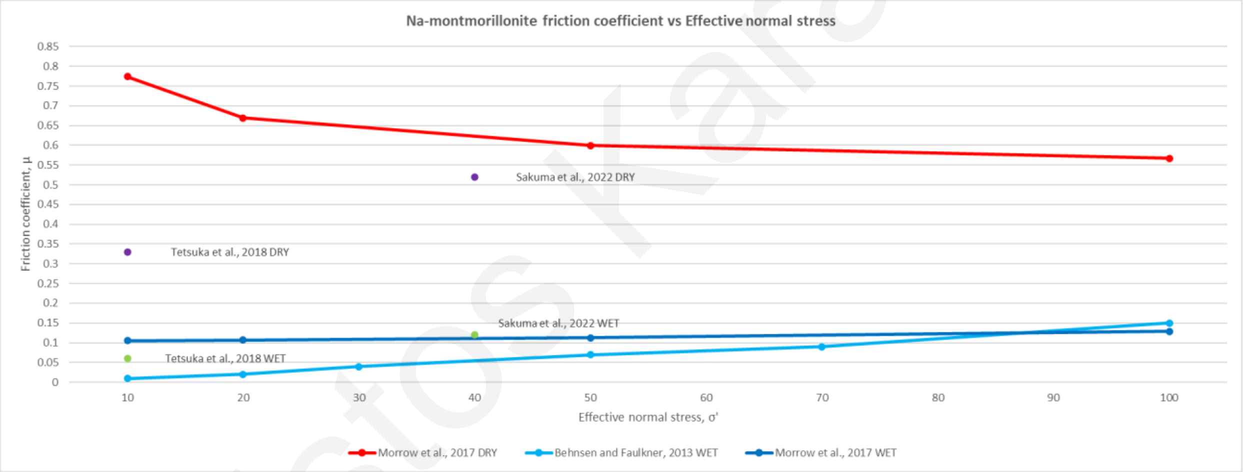


Figure 20 - Na-montmorillonite dry and wet friction coefficient vs effective normal stress (data from Morrow et al. (2017), Tetsuka et al. (2018), Sakuma et al. (2022) and Behnsen & Faulkner (2012))

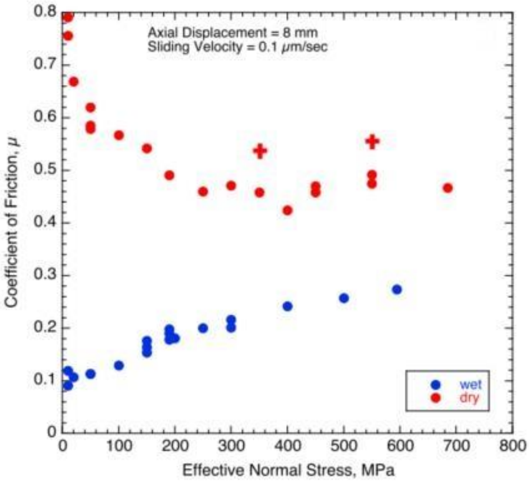


Figure 21 - Na-montmorillonite dry and wet friction coefficient vs effective normal stress from 10 to 685 Mpa (from Morrow et al. (2017))

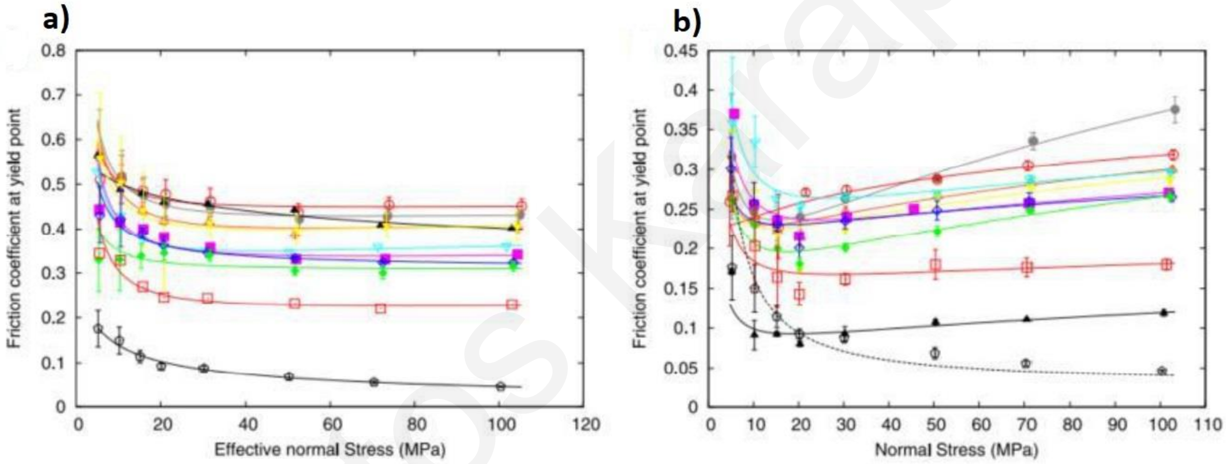


Figure 22 – (a) Dry and (b) wet friction coefficient vs effective normal stress, for different clay minerals (from Behnsen & Faulkner (2012))

Ikari et al. (2007), after performing frictional tests on samples with montmorillonite content at different normal effective stress vales (range from 5 to 100 MPa), while agreeing that dry friction coefficient decreases with increasing effective normal stress, they found that wet friction coefficient did not increase with increasing effective normal stress. However, on one hand this tendency became less significant as montmorillonite content of the sample increased, and on the other hand, values for 100% montmorillonite sample were very low (< 0.1) for the whole range of effective normal stress values, making the conclusions over this behavior difficult.

Figure 23 shows the friction coefficient reduction from dry to wet state with respect to effective normal stress, for Na-montmorillonite, chlorite and talc. Chlorite and talc were selected in order to give an indicative picture of the tendency, as the minerals with the highest (chlorite) and lowest (talc) friction coefficient in Behnsen & Faulkner (2012) Figure 23-excluding montmorillonite. We can conclude that the friction loss of clay minerals between dry and wet state on clay minerals becomes less, as effective normal stress increases.

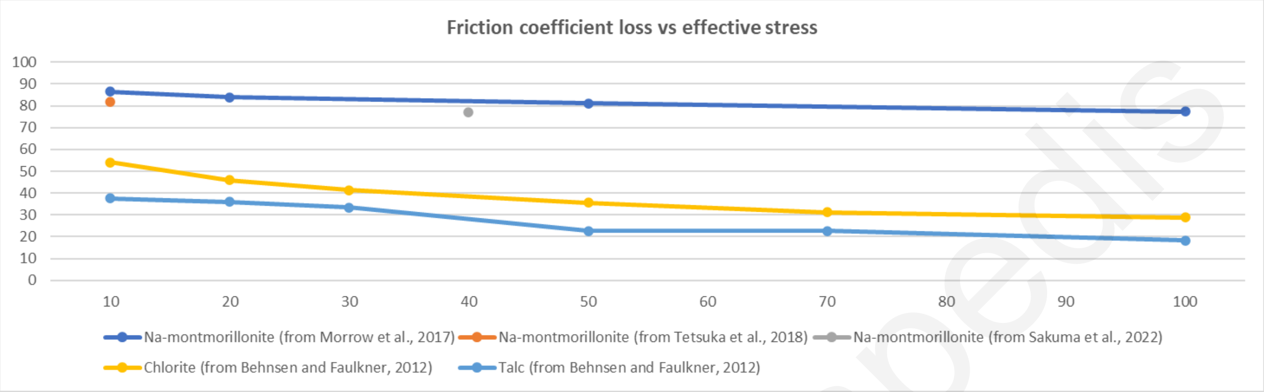


Figure 23 - Friction coefficient reduction vs effective normal stress for Na- montmorillonite, chlorite and talc. Data obtained from Morrow et al. (2017), Tetsuka et al. (2018), Sakuma et al. (2022) and Behnsen & Faulkner (2012))

Following the consideration that friction loss depends on the amount of water absorbed in the interlayer or adsorbed on clay mineral surface, it is easy to assume that decrease in friction coefficient also depends on the water content of the sample. Such a picture is given from Figure 24. As the water content of the sample (quartz and montmorillonite) increases, friction coefficient decreases. Also, one can observe that as the montmorillonite content increases against the quartz content, this reduction becomes more severe.

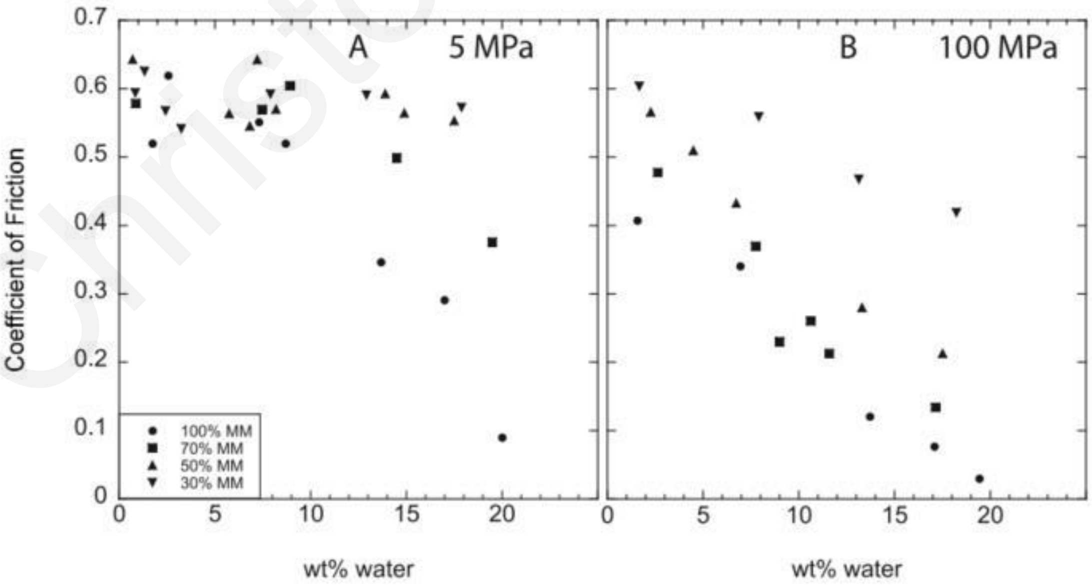


Figure 24 - Friction coefficient vs water content, from samples of quartz and montmorillonite (from Ikari et al. (2007))

As described above, clay minerals have the ability to attract water molecules from the air. Therefore, and as shown in Figure 19, relative humidity is capable enough to activate the friction loss mechanisms. Here, it is important to note that as a result of the above, tests on clay minerals on “dry” state, are not always achieved. In most procedures, researchers take measures in order to avoid the sample to interact with humidity; for instance, dry tests are performed exactly after the taking samples out of the oven or they perform tests in vacuum conditions. Even thought, some saturation can occur, thus, one test result described as “dry” could be partially saturated and correspond to low water content.

Other factors affecting dry and wet friction coefficient, could be the velocity of the friction test, i.e. shear deformation to time needed, or the salinity of the wetting fluid (Moore & Lockner, 2016), but these factors may be of secondary importance for most cases. For instance, Ikari et al. (2007) found that in order for friction coefficient to show a noticeable increase, the velocity of the test should be 100-300 mμ/s while usual tests performed at 0.5 - 5.0 mμ/s. What is more, Sakuma et al. (2022) found that friction coefficient of Na-montmorillonite saturated with 1M sodium chloride would be just 0.02 lower than that saturated with distilled water, while for K-montmorillonite would be 0.015 for the case of distilled water and 1M potassium chloride. Similar observations were done by Kawai et al. (2015) for the case of muscovite, saturated with distilled water, 0.5M caesium chloride and 0.5 sodium chloride, where the difference in friction coefficients were imperceptible.

Figure 25 presents friction coefficient values for dry and wet state, for other clay minerals, except montmorillonite. Figure 26 presents the friction coefficient reduction for the same minerals. Data were obtained from Morrow et al. (2000), Moore & Lockner (2004), Kawai et al. (2015) and Behnsen & Faulkner (2012). Note that: (i) these values refer to the peak frictional coefficient and not the residual one, (ii) values refer to effective stress of 100MPa, except for the values from Kawai et al. (2015) for muscovite, which refer to effective stress of 24MPa, (iii) values for muscovite from Kawai et al. (2015) refer to the case of powder samples (and not the single crystal samples also tested), (iv) in the cases of muscovite and phlogopite in Moore & Lockner (2004) and muscovite in Kawai et al. (2015), where 2 samples were tested, the mean value of them is presented, (v) for the calculation of percentage reduction, values for dry and wet friction coefficient refer to the mean of the available values for each mineral.

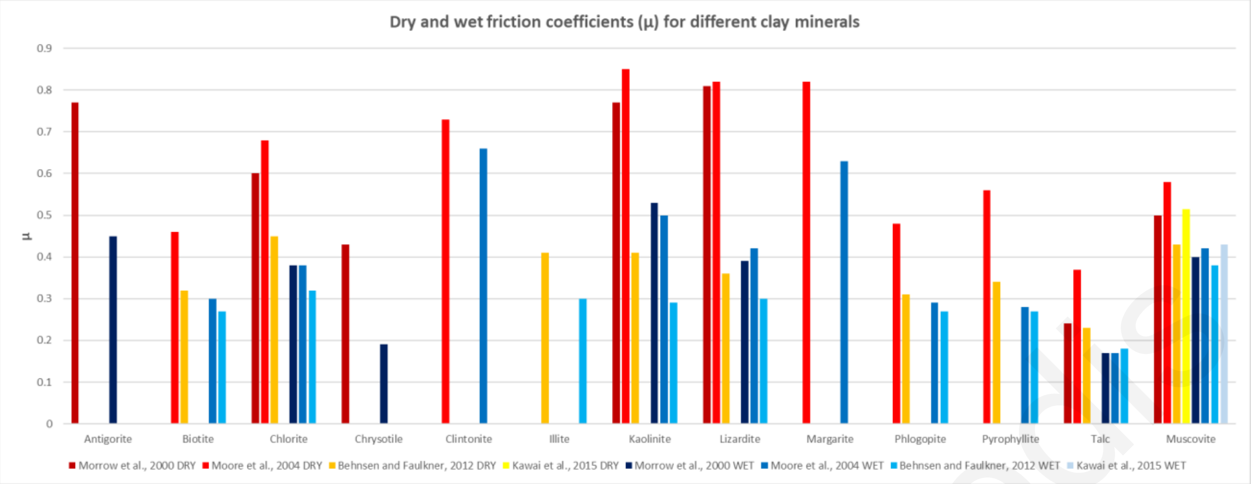


Figure 25 - Dry and wet friction coefficient for different clay minerals (data from Morrow et al. (2000), Moore & Lockner (2004), Kawai et al. (2015) and Behnsen & Faulkner (2012))

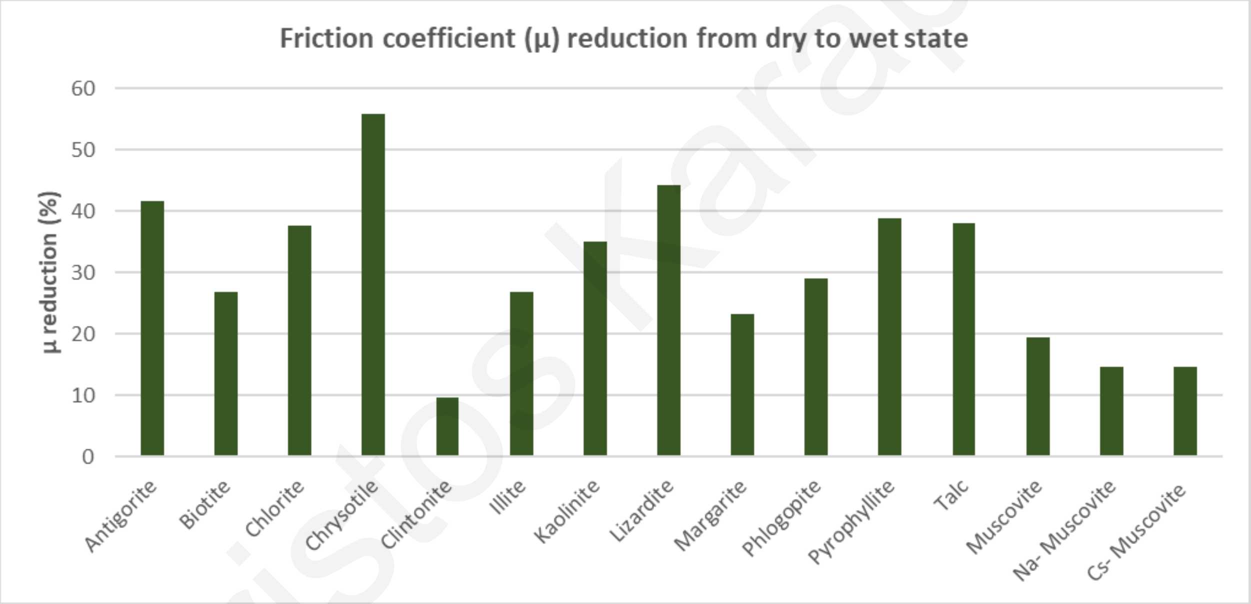


Figure 26 - Percentage of friction coefficient reduction for different clay minerals (data from Morrow et al. (2000), Moore & Lockner (2004), Kawai et al. (2015) and Behnsen & Faulkner (2012))

Chapter 3 Clay minerals in rocks: The case of rocks with high clay content

Mudstones, Siltstones and Marls are sedimentary rocks, characterized by the high clay mineral content in their mineralogy. Thus, these rocks are highly reactive when in contact with water. Their wet behavior is interesting, since it gives a good picture of the influence of clay minerals in wet rock strength.

Figure 27 shows the UCS loss in such rocks, between dry and wet condition, with data obtained from Erguler & Ulusay (2009), Jiang et al. (2014), Lu et al. (2017), B. Li et al. (2019) and Chen et al. (2022). Unfortunately, for that family of rocks, not enough data in literature are available. However, from the available data, it is obvious that rocks with high clay content are susceptible to high strength reduction when saturated, where in some cases is can reach values of even 90%.

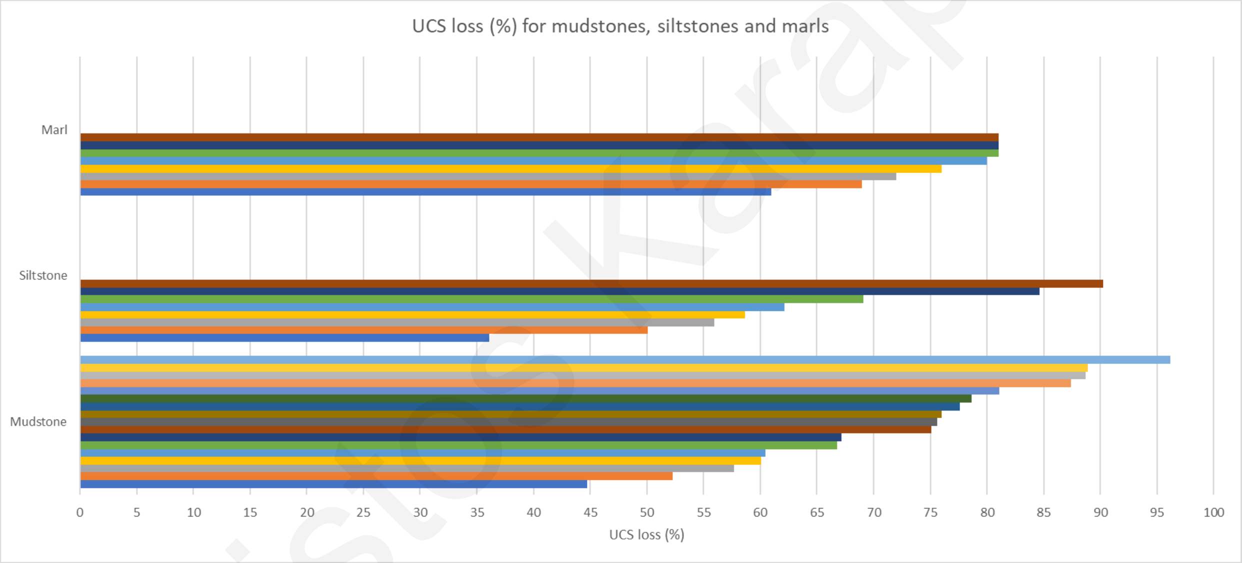


Figure 27 - UCS loss (%) for mudstones, siltstones and marls (Data obtained from Erguler & Ulusay (2009), Jiang et al. (2014), Lu et al. (2017), B. Li et al. (2019) and Chen et al. (2022))

3.1 Weakening mechanisms

During saturation, the structure of such rocks exhibits changes, mostly due to the reactivity of clay minerals. The procedure is, more or less, as follows: (1) During saturation, water finds its way into the rock, via the cracks of the sample (Jiang et al., 2014; B. Li et al., 2019). (2) As soon as water is in touch with clay minerals, these minerals expand. (3) Since clay minerals are predominant, their swelling does not cause the closure of pores (as it happens in sandstones - see Chapter 4), but instead, porosity increases (Lu et al., 2017) as pores are getting larger (M. Zhou et al., 2021). What is more, since different clay minerals with different swelling capacities are present, as well as clay particles have irregular shapes, swelling is not uniform into the rock body. (4) Consequently, contact between grains may change and transform from face-to-face, to point-to-face or even point-to-point (Jiang et al., 2014; B. Li et al., 2019), resulting in the rise of internal local stresses (Van Eeckhout, 1976) and the reduction of contact area, thus, friction resistance between grains reduces. The latter should be considered in addition to the fact that friction coefficient of clay minerals reduces under saturation (see 2.5). (5) Cracks start to appear and micropores connect to each other, forming in one hand, larger pores, and on the other hand, a network of open pores (Jiang et al., 2014; B. Li et al., 2019; M. Zhou et al., 2021), allowing water to penetrate even more into the rock body. In addition, “close pores” could become “open” (Figure 28) (M. Zhou et al., 2021). (6) All these, give access to water in regions which were not accessible before, and thus, more clay minerals get in touch with water and react, resulting in a new cycle of reactions. The aforementioned progressive procedure is responsible for the changes in the structure and the strength reduction of such rocks.

(**a**) Initial pores

(**b**) Expanded and connected pores

Figure 28 - Expansion and connection of pores & "close" pores became "open" (from M. Zhou et al. (2021))

3.2 Time dependency

In mudstones, this procedure does not stop until water is expelled from rock pores; otherwise, total disintegration of the rock sample may occur. Jiang et al. (2014) and Chen et al. (2022)

performed tests on mudstones to observe the crack propagation process. In Jiang et al. (2014), mudstone sample had a clay content (chlorite and illite) of about 45%, and it was immersed in water for 1 year. Figure 29, shows the crack propagation in the sample relative to the time of immersion. It can be seen, that most of the cracks evolved in the first 75 hours, but this evolution kept on going, until the specimens were taken out of water. Figure 30, shows the condition of four specimens, after one year of immersion; the specimens were totally disintegrated. Note that, in this experiment, samples were hold in plexiglass containers, so, total disintegration could have happened earlier, if the samples were “free” to swell.

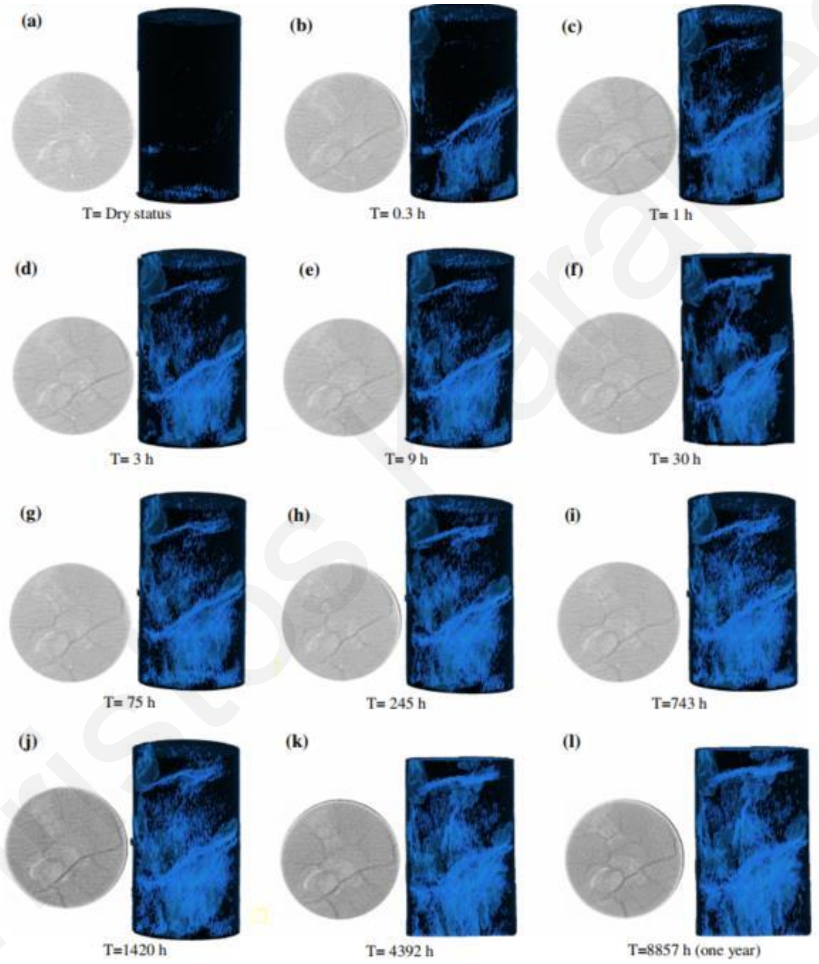


Figure 29 – Images presenting the crack propagation of mudstone with time of immersion. Voids of sample are represented with blue (from Jiang et al. (2014))

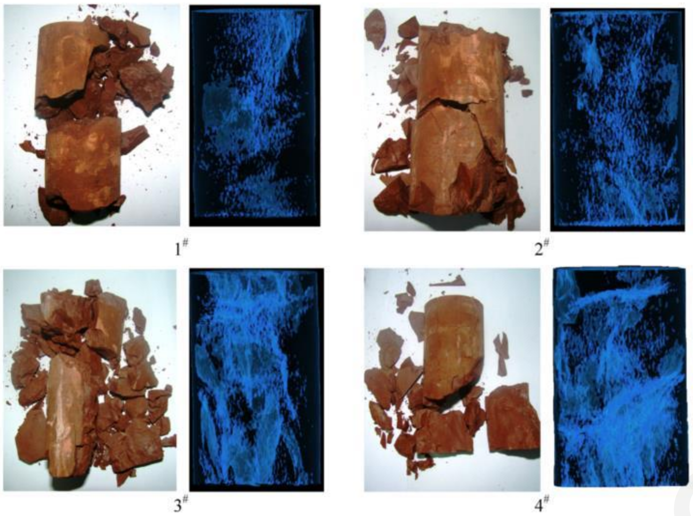


Figure 30 - Disintegration of mudstone after 1 year of immersion (from Jiang et al. (2014))

In Chen et al. (2022), mudstone had a clay content of about 50.3%, 90% of which was smectite. Figure 31, shows the disintegration progress of the sample for 26 hours of immersion in water. It can be seen that after 10-20 minutes, the sample broke in smaller, detached pieces.

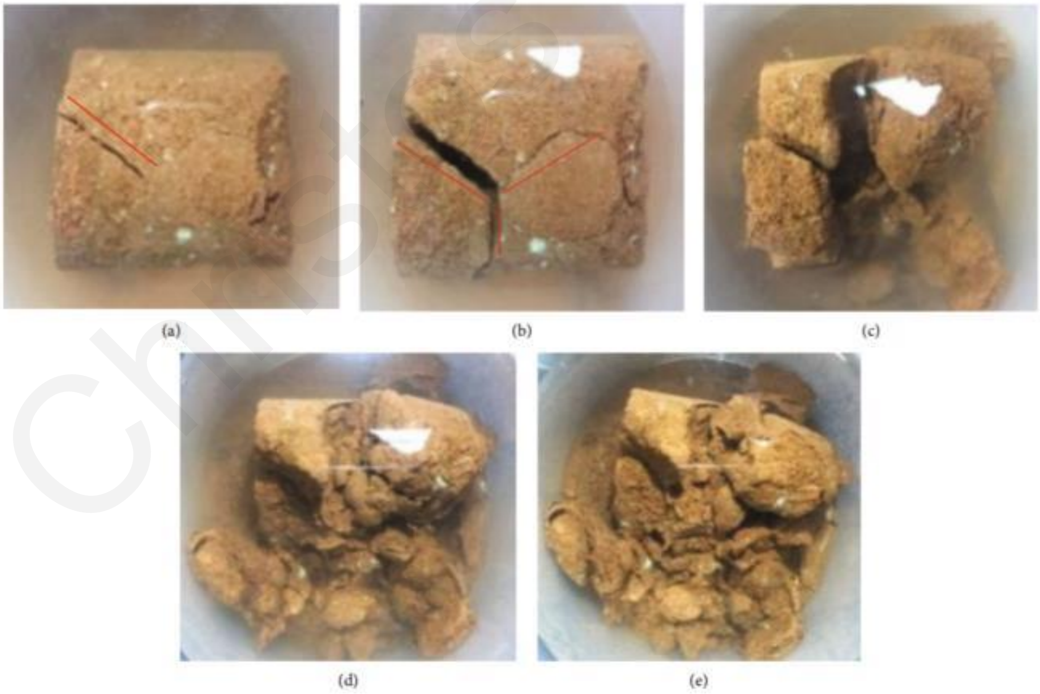


Figure 31 - Disintegration process of a mudstone with high smectite content, after (a) 1 minute, (b) 10 minutes, (c) 20 minutes, (d) 60 minutes and (e) 26 hours of immersion (from F. Chen et al. (2022))

The difference in the disintegration progress of these experiments, should be attributed to the smectite content of the mudstones. Therefore, one could state that immersion time in combination with the smectite content, is an indicator of the strength reduction in rocks with high clay content. Siltstones and marls, should behave accordingly, since clay content is similar.

In rocks containing large portion of calcite, such behavior could be more intense, because calcite is a soluble mineral (Appendix B). As long as soluble minerals are in contact with water, part of their surface dissolves into the water and contact areas weaken. Consequently, part of the skeleton is lost, distance between particles increase, and bonding forces (for instance Van der Waals forces) weaken (B. Li et al., 2019).

3.3 Wetting and drying cycles

In order to simulate natural conditions where rocks suffer from alternate wetting and drying (as in water reservoirs), experiments with cycles of wetting and drying in mudstone and siltstone have been performed in order to study the swelling (Doostmohammadi et al., 2009; José Pejon & Valentin Zuquette, 2002), porosity increase (M. Zhou et al., 2021) and UCS reduction (S. Huang et al., 2018); no relative experiments were found for marl.

Figure 32, shows the reduction of UCS after 12 cycles of wetting and drying of a mudstone, with data from S. Huang et al. (2018). Figure 33 shows the swelling strain of mudstone after time and wetting and drying cycles tests performed by José Pejon & Valentin Zuquette (2002), and Figure 34 shows the evolution of porosity in siltstone, after wetting and drying cycle tests performed by M. Zhou et al. (2021).

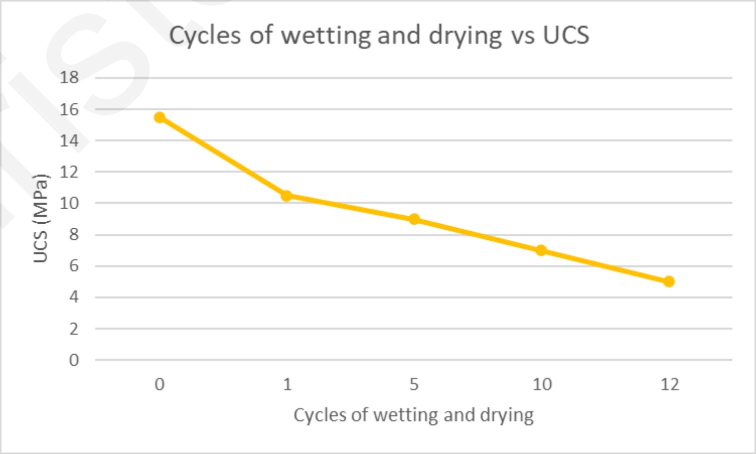


Figure 32 - UCS vs number of wetting and drying cycles, for mudstone (Data from S. Huang et al. (2018))

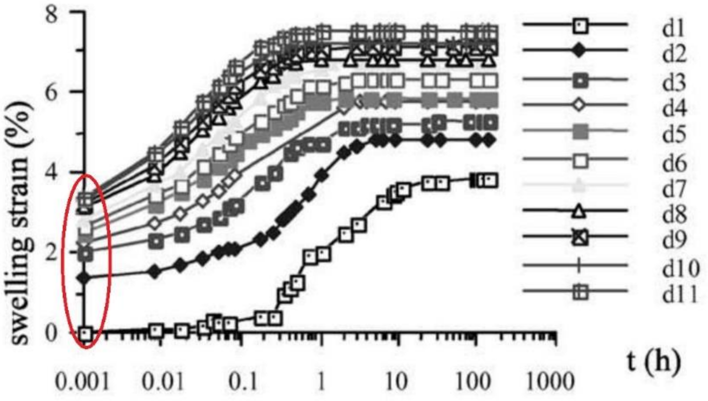


Figure 33 - Typical swelling vs time and wetting and drying cycles for mudstone. Red circle indicates that part of the swelling is irreversible. (from José Pejon & Valentin Zuquette (2002))

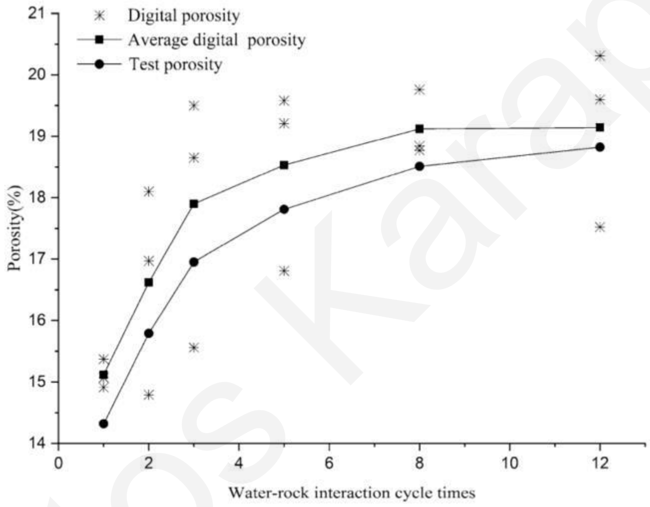


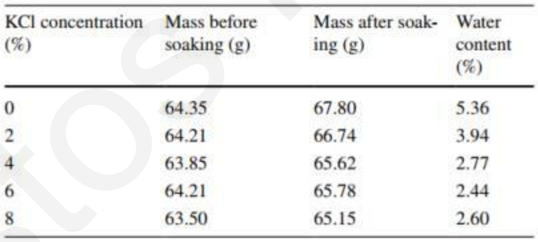
Figure 34 - Porosity vs wetting and drying cycles (from M. Zhou et al. (2021))

The behavior after wetting and drying cycles can be explained under the following steps: (1) during saturation, the samples swell and its structure suffers changes, and as a result, porosity increases. The mechanism is the same as explained above. (2) During drying, only a part of the total swell (and porosity increase) is reversible. This is indicated from the difference in initial swelling strain in each cycle, as shown into the red circle in Figure 33. Similar observations are be made in Doostmohammadi et al. (2009). (3) Consequently, the next cycle starts from a new point, where more surface area is accessible to water, meaning that more clay minerals may react. (4) As cycles continue, total swelling of the sample increase in every cycle, resulting in porosity increase. (5) After some cycles, no extra swelling is observed, and porosity remains constant. However, total disintegration of the sample may occur before reaching this number of cycles. (6) Since UCS loss is attributed to the cumulative structure changes, the reduction of UCS after wetting and drying cycles should be explained in the same way.

3.4 Saturation with Sodium and Potassium Chloride

X. Chen et al. (2020), performed experiments on mudstone, where saturation liquid was a potassium chlorite solution (KCl) in 0% (pure water), 2%, 4%, 6% and 8% concentration. The mudstone had a clay content of 27.4%. They found that, for saturated samples, as the percentage of KCl concentration increased, water content and swelling capacity decreased (Table 5 and Figure 35). The reason could be explained in addition to what was discussed in 2.4 and 2.5: i) the presence of potassium in the interlayer of clay minerals, is related to the limited basal space increase during water absorption, and ii) as concentration of cations in water increases, less water is needed between particles to equilibrate the concentrations in the interparticle space and pore water (osmotic swelling). Figure 36, shows the UCS loss for each KCl concentration, for the same experiment. As KCl concentration increases, UCS loss decreases, and this could be explained in the same manner; less swelling, leads to less structure changes, and thus, less reduction in UCS. Note that in this experiment, test for the case of pure water (0% KCl) could not be accomplished, because of the sample's disintegration.

Table 5 - Water content with different KCl solutions for mudstone (from Chen et al., 2020)



KCl concentration (%)	Mass before soaking (g)	Mass after soaking (g)	Water content (%)
0	64.35	67.80	5.36
2	64.21	66.74	3.94
4	63.85	65.62	2.77
6	64.21	65.78	2.44
8	63.50	65.15	2.60

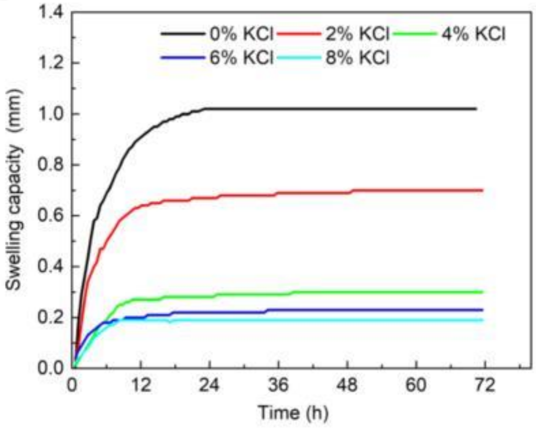


Figure 35 - Swelling capacity with time for different KCl solutions for mudstone (from X. Chen et al. (2020))

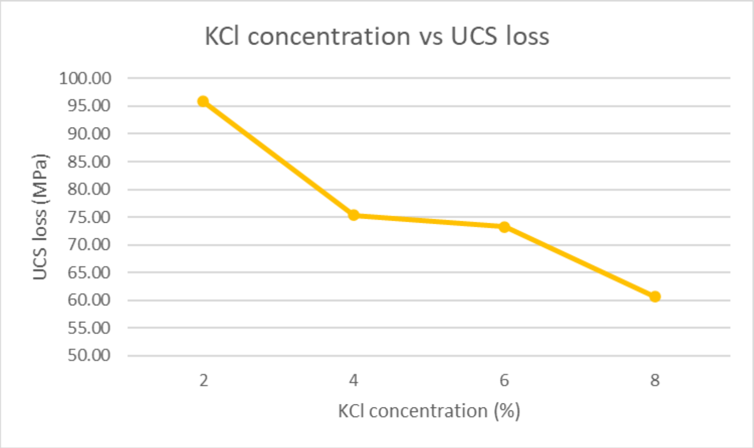


Figure 36 - UCS loss for different KCl solutions (data obtained from X. Chen et al. (2020))

Anne Perera et al. (2018) and Wanniarachchi et al. (2019) studied the influence of sodium chlorite (NaCl) on strength of siltstone. The research team was pretty much the same in both cases. Siltstone in both cases was taken from the same region and had a clay content of about 40-45%, but dry strength was different. Figure 37, shows the variation of UCS for dry and saturated samples at different NaCl concentrations, including pure water, from Anne Perera et al. (2018) experiments. It can be seen that as NaCl concentrations increase, the UCS loss decreased, although the difference is insignificant. The exception here is for the case of pure water, where the reduction in strength was the lowest observed. The latter does not agree with the previous observations in mudstone and KCl solutions (Figure 36). According to authors, high ion concentration in pore water can cause corrosion in the rock structure. This difference between NaCl solutions and pure water, could mean that the negative influence of ion corrosion is greater than the aforementioned positive ones. Authors also referred to the positive influence of NaCl crystallization into rock pores, which could add extra strength to the rock. But this statement needs more examination, since crystals may also cause exfoliation or add extra internal stress, increasing the internal damage of the rock, especially in rocks weak as the investigated ones.

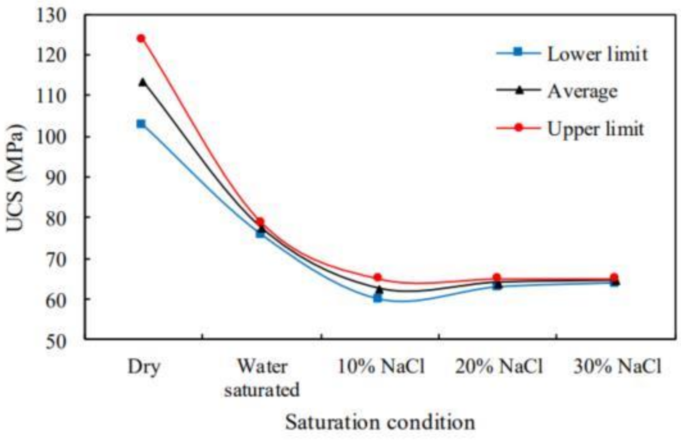


Figure 37 - UCS variation in siltstone, under dry and wet conditions, for the case of water and different NaCl solutions (from Anne Perera et al. (2018))

Figure 38, presents the UCS test results of Wanniarachchi et al. (2019). In this case, NaCl concentration of 10%, 20% and 25% were compared. Test showed that UCS loss for all three cases were very close; the greatest UCS loss was found for 10% concentration, 20% showed the lowest one and 25% was in the middle, a fact that is interesting. According to authors, the negative influence of NaCl for the case of 25% concentration is such that it overcomes the positive ones compared to the 20% concentration; but these test results are not in agreement with the previous experiment. So, safe conclusions cannot be made, and further research should take place.

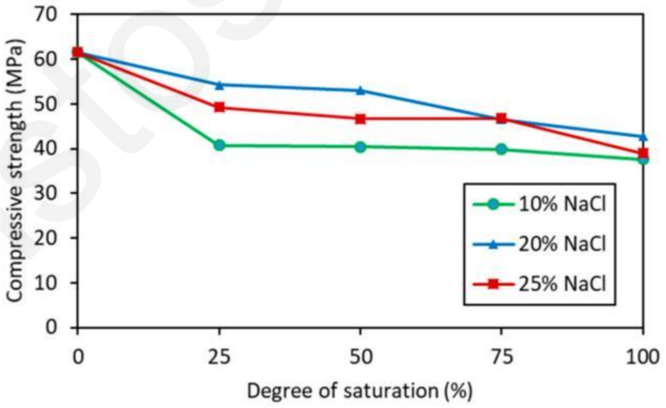


Figure 38 - UCS variation in siltstone, versus degree of saturation for different NaCl solutions (from Wanniarachchi et al. (2019))

3.5 *Water content (or saturation degree) dependency*

Except from the saturation period, the amount of water in the rock body is also important. After many tests on different sandstones, Hawkins & Mcconnell (1992) proposed that the relation between UCS and water content is expressed by Equation 1:

$$\sigma_c = a \cdot e^{-b \cdot w} + c \qquad (1)$$

with σ_c being the UCS, w the water content, and a, b and c constants unique for each rock, and for the estimation of them, one needs to perform a series of UCS tests in different water contents. UCS in dry state is equal to c, while in full saturation it is equal to a + c. The constant b expresses the steepness of the curve. After experiments on mudstones by Lashkaripour & Ajalloeian (n.d.), Jiang et al. (2014), Zhang et al. (2017) and on siltstone by B. Li et al. (2019), it was confirmed that the relationship between UCS and water content, is also valid for rocks with high clay content.

Figure 39, shows the relation between saturation degree and the percentage of total UCS loss, with data from the aforementioned articles. Vergara & Triantafyllidis (2016) found another relation for UCS and water content for, that follows a parabolic curve (Figure 40).

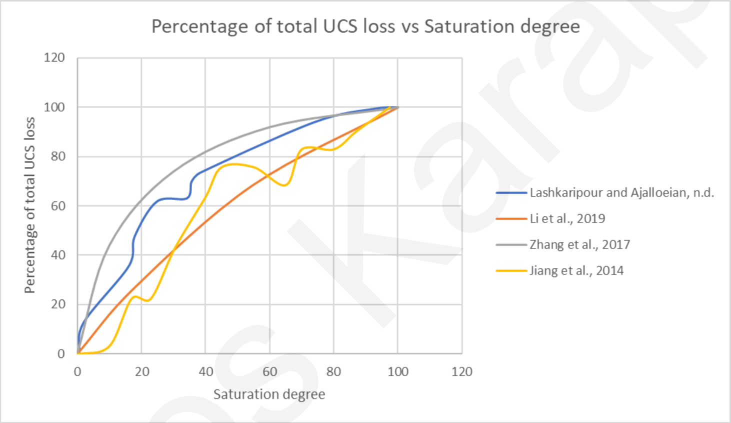


Figure 39 – Saturation degree vs percentage of total UCS loss for mudstones and siltstone (data Lashkaripour & Ajalloeian (n.d.), Jiang et al. (2014), Zhang et al. (2017) and B. Li et al. (2019))

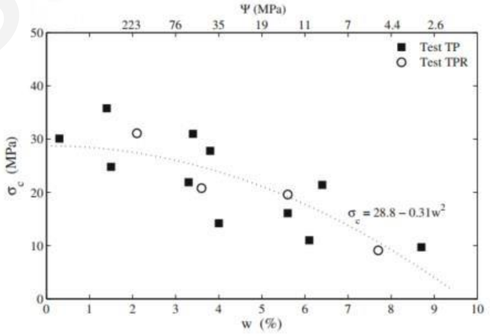


Figure 40 - UCS vs water content for marl (from Vergara & Triantafyllidis (2016))

All curves indicate that UCS decreases as water content increases, as expected. From clay mineral concern, greater amount of water into the rock sample means that, on one hand, more water is “available” to take place in the reactions (for instance the formation of more water layers, the activation of osmotic swelling etc), and on the other hand, that more clay particles react with water. The difference between the three rock types, is that mudstones and siltstones lose most of their UCS at lower saturation degrees, and as saturation increases, the rate of UCS loss decreases. The same trend was found for sandstones. On the other hand, marls seem to follow the exact different trend; the rate of UCS loss increases as saturation degree increases. This may be due to the capillary phenomena, since capillarity is lost at higher saturation degrees (Appendix E). The aforementioned should be considered in addition to Appendix C, regarding the water distribution in rock sample. Concluding, increasing saturation degree means that more parts of the sample get in touch with water and more pores get fully filled, so clay mineral reaction in rock is more intense, causing greater internal damage.

3.6 Discussion over the approachability of clay minerals from water

From the previous discussion, it was concluded that clay minerals have a significant influence on UCS loss, as does porosity. Note that, in Erguler & Ulusay (2009), most of the mudstones and siltstones had a very low smectite content, however, they presented the aforementioned swelling and strength reduction behavior. What is more, and as pointed in 2.5, all clay minerals present a friction coefficient reduction in wet state. Thus, in the present study, clay minerals are studied as one, and when “clay content” is referred, all clay minerals content is taken into account. Figure 41 and Figure 42, show the dependence of UCS loss in mudstone from clay content and porosity respectively, with data from Erguler & Ulusay (2009). Figure 43 and Figure 44, show the dependence of swelling strain after 1 cycle of wetting and drying, from the same parameters, with data from José Pejon & Valentin Zuquette (2002). The first cycle was chosen instead of maximum swelling after a number of cycles, because the porosity given in the article refers to the starting porosity (i.e. the porosity of the sample before any cycle).

For both cases, clay mineral content is not a good indicator for the examined variable. Porosity is a good indicator for swelling strain, with R^2 being equal to 0.89, while for UCS loss the R^2 equals to 0.22. If the specimen L16-1 is excluded, the R^2 value are equal to 0.54 (the reason for this exclusion will be explained later).

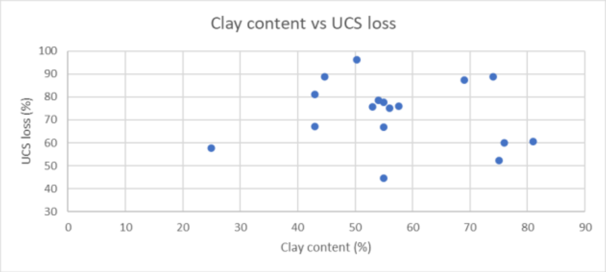


Figure 41 - Clay content vs UCS loss for mudstone (Data from Erguler & Ulusay (2009). Note that clay content here is the sum of clay content and mica content in the article)

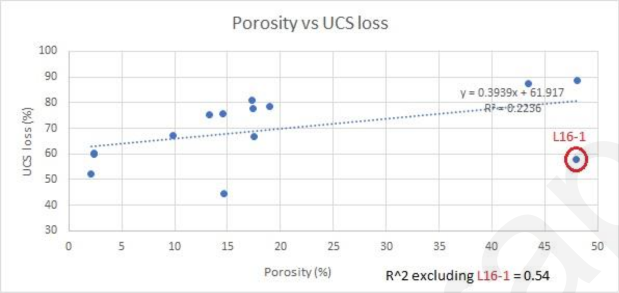


Figure 42 - Porosity vs UCS loss for mudstone (Data from Erguler & Ulusay (2009). Note that clay content here is the sum of clay content and mica content in the article)

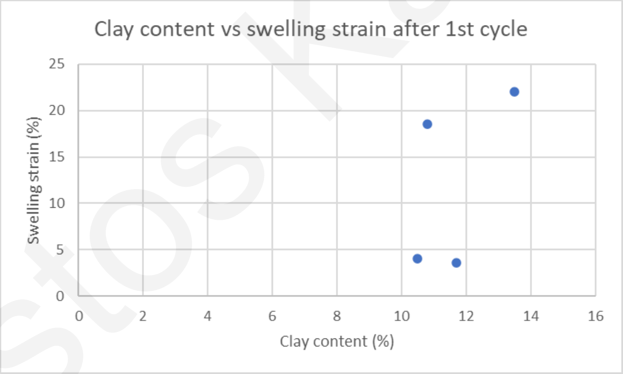


Figure 43 - Clay content vs Swelling strain for mudstone (Data from José Pejon & Valentin Zuquette (2002))

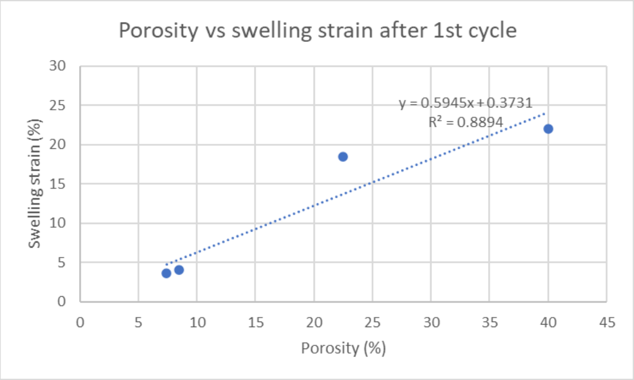


Figure 44 - Porosity vs Swelling strain for mudstone (Data from José Pejon & Valentin Zuquette (2002))

The fact that clay content seems totally irrelevant from both UCS loss and swelling strains, does not seem reasonable. So, trying to find a variable that would include clay content and would be in agreement with the general conclusions, the idea of approachability of clay minerals from water came up. As mentioned, the increase in porosity gives new routes for water to penetrate, and new regions of rock body get saturated, resulting in new clay minerals to take place in the reaction and the alteration of rock structure. So, according to this, there would be some parts of clay content which are not in contact with water, and thus, do not react. So, changes in rock structure could not depend just on the clay content or porosity, but to the proportion of clay content which is approachable by water, or else, the percentage of clay minerals that react.

The easiest way to simulate the approachability of clay from water, i.e., “how much of clay content is in touch with pore water”, is to consider the ratio of porosity over clay content. Figure 45 and Figure 46, present the relation between this ratio and the examined mechanical properties. As one can see, the correlation in both cases increased, compared to the case of porosity alone (R^2 for UCS loss increased to 0.60 from 0.54 and for swelling strain increased to 0.96 from 0.89). The basic advantage of this suggestion, is not the increase in R^2, which is not significant, but the fact that clay content is taken under consideration. Note that, relative exercise was made for the case of siltstone and marl, with data from Erguler & Ulusay (2009), but for both cases, not three of clay content, porosity and porosity / clay content showed a good correlation with UCS loss. Sample L16-1 was chosen to be excluded as it was considered as extreme value.

Simulating approachability this way shows some weaknesses. First of all, porosity refers to the whole volume of the sample, while clay content refers to the volume of solid part. Transforming clay content as a ratio of the volume of sample may give more accurate results. Additionally, with this way, the nature of voids and clay, their size and their specific surface area are not taken under consideration. Estimating the whole surface area of the voids (i.e. the interface between clay and water under saturation) and the whole surface of clay part, would give more analysis options. Furthermore, assuming the porosity constant over saturation time is not correct, as discussed before. So, a more precise approach should consider the fact of porosity increase during saturation time or wetting and drying cycles. Finally, one doubt that comes in mind according to this simplified approach, is that, for two different rocks with a given porosity, the one with the lower clay content would give a greater porosity over clay content ratio, and so, a greater UCS loss. The simplified approach would for sure give a wrong result, in the case of a very big porosity value, over a relatively small clay content. Estimating the upper limit of clay content over which no extra clay part can be in contact with water (meaning that 100% of pore surface is in touch with clays), or the lower limit of porosity for which all clay parts will be in touch with water, so any increase in porosity would not have any attribution, could help to exclude extreme values. However, the simplified method can give an easy and quick picture, and in any way, supports the idea that approachability is something that could get more attention; as shown in Figure 45 and Figure 46, the greater the approachability, the greater the reduction in UCS.

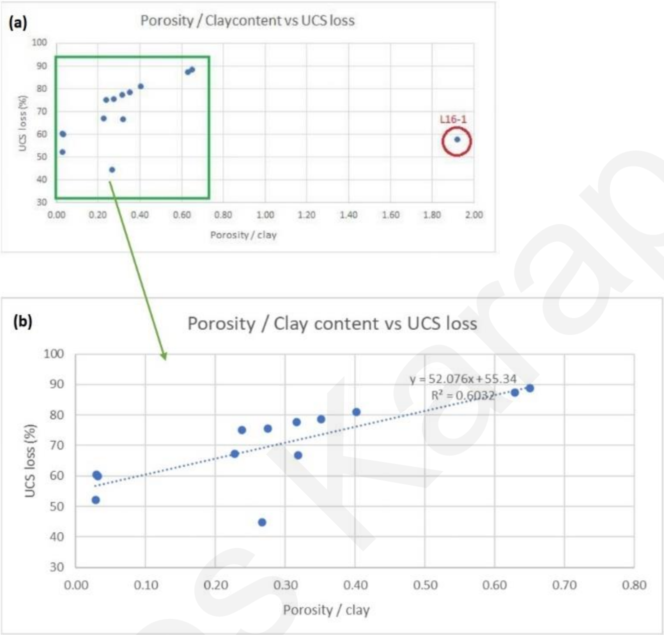


Figure 45 - Porosity / clay content vs UCS loss for mudstone. (a) For all the samples and (b) excluding sample L16-1 (Data from Erguler & Ulusay (2009))

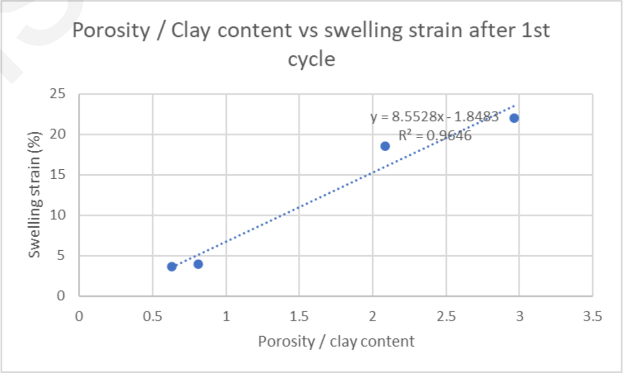


Figure 46 - Porosity / clay content vs swelling strain (Data from José Pejon & Valentin Zuquette (2002))

Chapter 4 Clay minerals in rocks: The case sandstones

Sandstone is a very common rock type, consisting mainly of sand-size grains (0.0625 to 2.00 mm). It is composed mostly of quartz and feldspar, while calcite may also be found. Clay minerals are very common in sandstones, and their content could be anywhere between 0% and 20-25%. A large variety of other minerals can be found, but usually in small amounts.

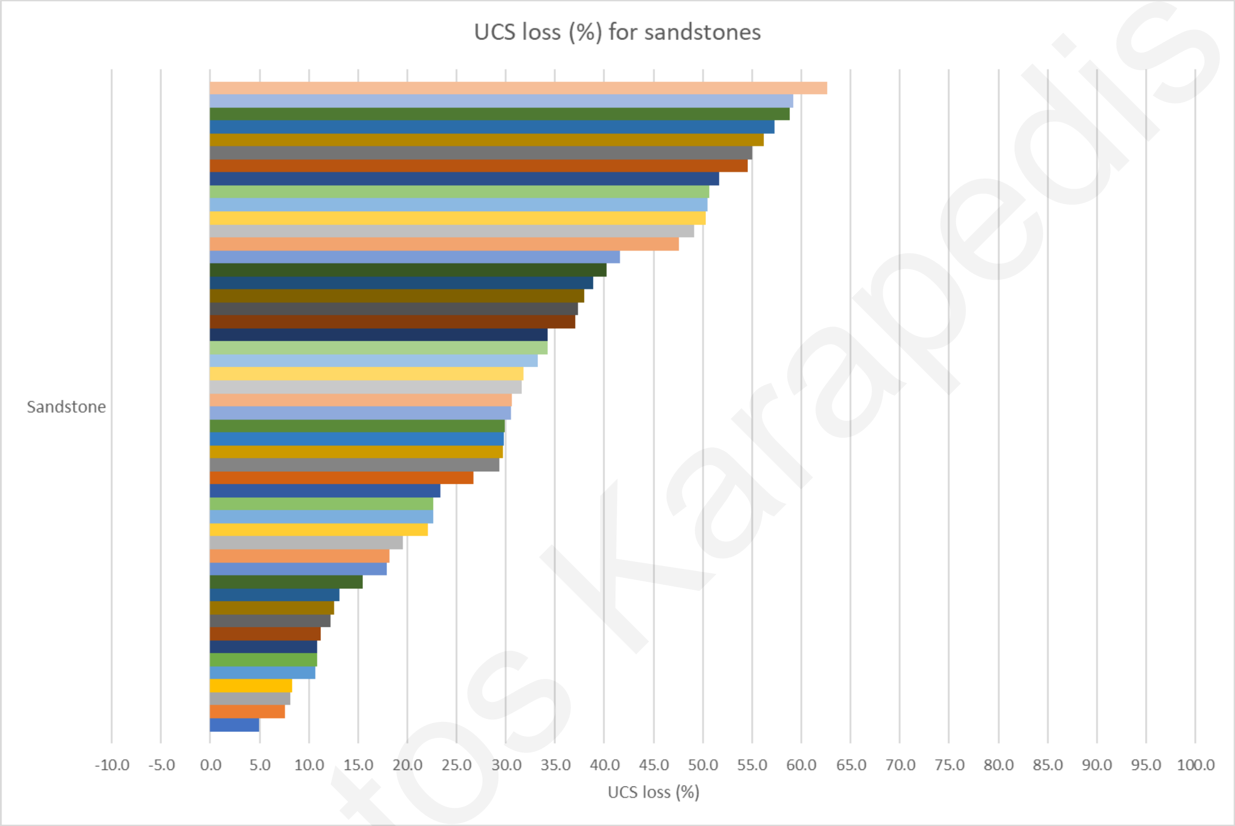


Figure 47 - UCS loss (%) for sandstones (Data from Hawkins & Mcconnell (1992), Shakoor & Barefield (2006), S. Huang et al. (2010), Verstrynge et al. (2014), Z. Zhou et al. (2016), ZHOU et al. (2016), Lu et al. (2017), Masoumi et al. (2017), Tang (2018), Wasantha et al. (2018), Cai et al. (2019) and Noël et al. (2021))

Figure 47 shows the reduction of UCS in sandstones, between dry and wet state. Data were obtained from Hawkins & Mcconnell (1992), Shakoor & Barefield (2006), S. Huang et al. (2010), Verstrynge et al. (2014), Z. Zhou et al. (2016), ZHOU et al. (2016), Lu et al. (2017), Masoumi et al. (2017), Tang (2018), Wasantha et al. (2018), Cai et al. (2019) and Noël et al. (2021). It can be seen that i) the fluctuation of strength percentage loss is great, with the values covering evenly all the range from 4.9% to 78.1%, and ii) the strength loss is not so intense compared to the case of mudstones. Note that, in Hawkins & Mcconnell (1992), one sandstone (Ashdown Sandstone) showed an increase in its UCS under saturation, which is strange. Its properties are similar to other rocks, and no similar observation according UCS increase under saturation has been reported in the literature, even in other rock types. Authors do not provide information about the number of this sandstone's samples tested, and so, this record was excluded from this report, as well as the following ones. What is more, in Noël et al. (2021), the Fontainebleau sandstone showed a great deviation in its measured mechanical characteristics, and it was also excluded from all reports.

4.1 Weakening mechanisms

Because of clay swelling, during saturation, the expanded clays fill more pore space than in dry condition, resulting in the decrease in porosity (Aksu et al., 2015; Jiménez-González et al., 2008), unlike in mudstones and siltstones. Clay minerals will swell "freely", until other grains block their expansion. In this case, an extra internal stress will develop, known as *swelling stress or swelling pressure,* and will apply on rock grains*.* Figure 48 is relevant.

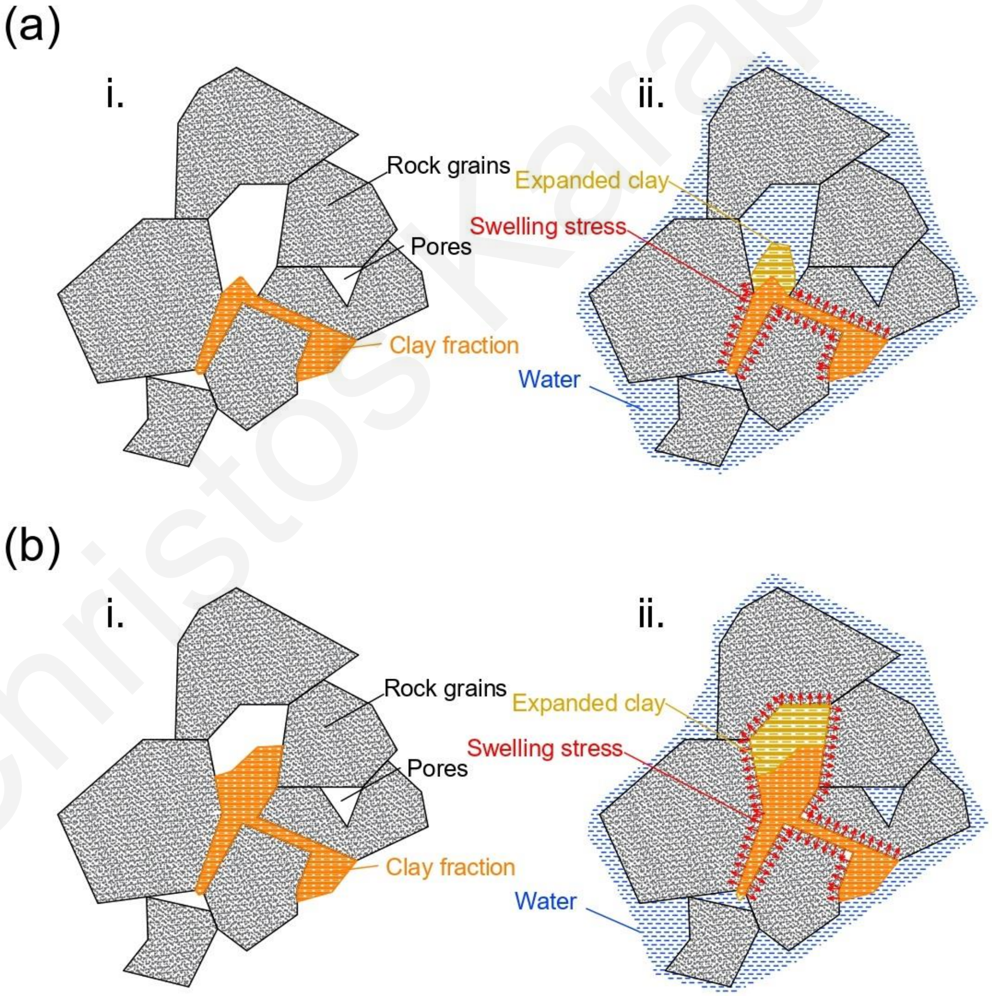


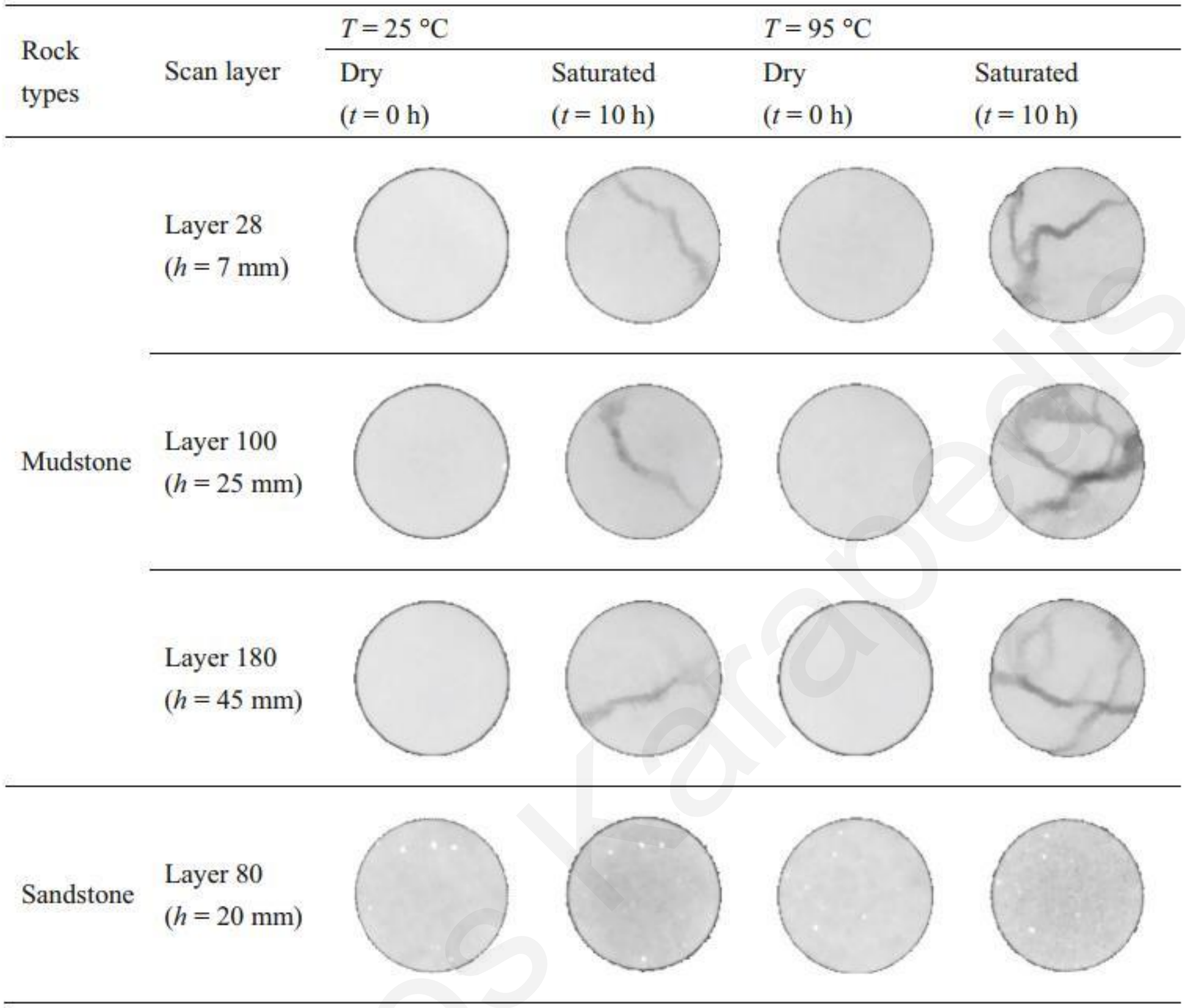
Figure 48 - Expansion of clay minerals in sandstones. (a) Free swelling and (b) developement of swelling stress

Typical values of swelling stress for different clay minerals are given in Table 6. It can be seen that the values of swelling stress are in the order of some kPa to approximately 0.5-0.6 MPa. Only montmorillonite shows a swelling stress value slightly over 1.00 MPa. Similar value (about 0.80 MPa) of swelling stress was estimated by Jiménez-González et al. (2008), after tests in sandstone sample.

Table 6 - Swelling stress for different clay minerals. Data obtained from Kohno (2020), (2021); Kohno et al. (2018)

Smectite/ Montmorillonite	**Kaolinite**	**Halloysite**	**Pyrophyllite**	**Talc**	**Mica**	**Chlorite**	**Vermiculite**	**Reference**
	0.17	0.24		0.01	0.02			Kohno et al., 2018
0.98	0.37	0.56	0.01	0.01	0.02	<0.002	0.13	Kohno, 2020
1.10	0.42	0.63	0.02	0.01	0.03	<0.002	0.15	Kohno, 2021

Tensile strength of sandstones is usually higher, and considering that the stress is applied only in the part of the cross section where clay is present (depending on the clay content of the sample), one could conclude that swelling stress alone cannot cause the total breakage of a sandstone, at least in most of the cases. What is more, there is no available data in the literature to confirm the opposite. On the contrary, in such experiments, no obvious damage or fracture has been observed after saturation of sandstones. Figure 49 is indicative.



Rock types	Scan layer	$T = 25$ °C		$T = 95$ °C	
		Dry ($t = 0$ h)	Saturated ($t = 10$ h)	Dry ($t = 0$ h)	Saturated ($t = 10$ h)
Mudstone	Layer 28 ($h = 7$ mm)				
	Layer 100 ($h = 25$ mm)				
	Layer 180 ($h = 45$ mm)				
Sandstone	Layer 80 ($h = 20$ mm)				

Figure 49 - Images of mudstone and sandstone after different time of immersion and temperature (from Lu et al. (2017))

However, swelling stress can still cause some internal damage in the sample, and can help with the propagation of cracks (Cai et al., 2019; H. Li et al., 2019). Figure 50 illustrates some possible mechanisms. In one hand, swelling stress can lead to the total detachment of rock grains, when the developed swelling stress is higher than the friction between grains, or when the clay cementation of grains swells and its friction is reduced. On the other hand, a grain can exhibit breakage under the swelling stress, through its weaker part. The latter should be considered in addition to the clay migration; during saturation and water flow inside the pores, clay could suffer some abruption, and so, clay fragments can insert into cracks, and build up new fractions where no clay was present before (Figure 51). In addition, in this case, the same amount of clay in the rock sample will attract more water than before (H. Li et al., 2019), since the surface area of the whole clay fraction would be more. Thus, osmotic swelling would be greater and swelling stress could appear in new points.

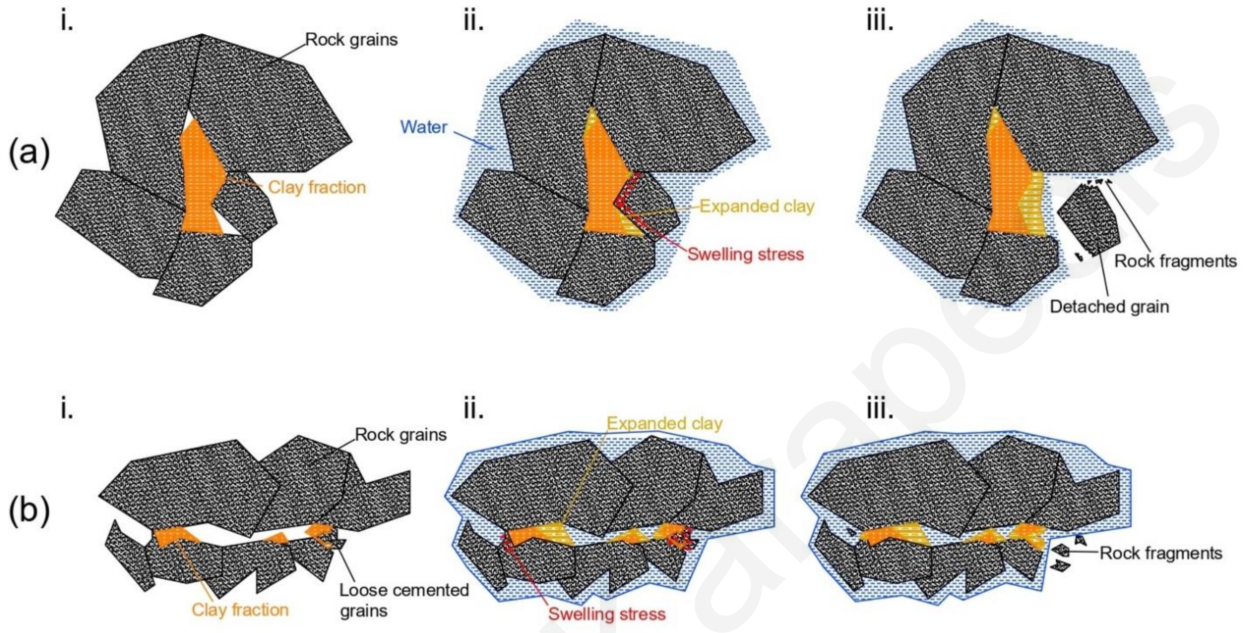


Figure 50 - Mechanisms of internal damage in sandstones, due to clay swelling

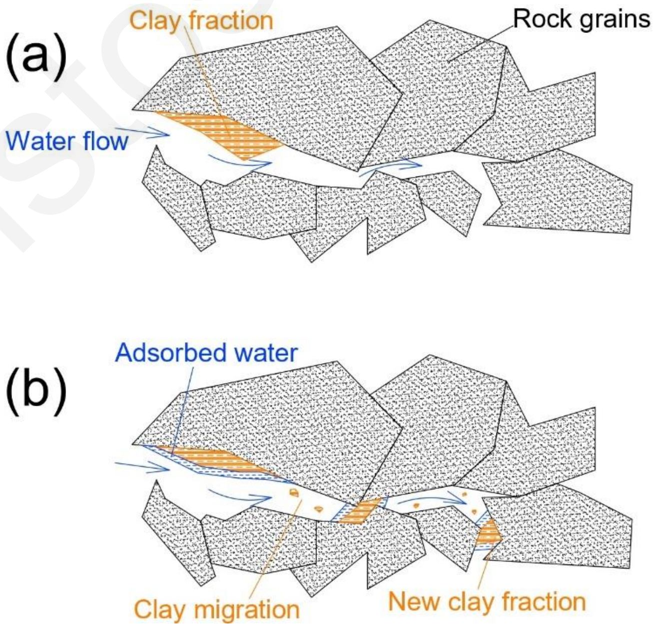


Figure 51 – Migration of clay particles after water flow in rock pores. (a) Clay fraction before water flow and (b) during water flow. See how new clay fractions form

Aksu et al. (2015), performed experiments in artificial sandstone, to examine the reduction of permeability after swelling. They found that for samples with the same clay content, permeability reduction decreased as pore size increases, and for high pore size, this reduction was insufficient. They suggested that this happened because, for bigger pores, swelling was not enough to cause the total closure of the pores, and thus, water could still flow through them. So, permeability reduction is proportional not just to the clay content, but also to the pore size.

Extending this thought for the case of compressive strength reduction, seems interesting. When in pores with lower size, swelling of clays will not be “free”, since there will not be enough space to do so. Instead, swelling stress will develop, resulting in internal damage mentioned before. On the other hand, for higher pore sizes, swelling will be unconfined and swelling stress development will be limited. So, for the same clay content, the higher the pore size of the rock, the lower the influence of clay in UCS reduction. Unfortunately, in the articles concerning the UCS loss, the pore size distribution of the samples is not given. The only data available is in Noël et al. (2021); comparing Bentheim and Adamswiller sandstones, the only ones from the four tested sandstones with similar clay content (8.0 and 9.2% respectively) and porosity (24.0% and 24.3% respectively), one could see that Adamswiller sandstone with 6-60μm pore diameter suffered a 23.4% UCS loss, while the Bentheim sandstone, with a pore diameter of 20-100μm, suffered a 7.5% of UCS loss. Once again, the available data are not sufficient enough to confirm this hypothesis. Note that, porosity itself is not enough to indicate the available space of clay to swell; two rocks with the same porosity could have different pore size distributions. Also, in the case of sandstones, where clay is usually not in high proportions, it could be stated that while rock is saturated, whole of the clay fraction gets hydrated - unlike in mudstones.

Jiménez-González et al. (2008) examined the natural damage induced in historical building constructed from swelling sandstone, highlighting the effect of clay minerals. One interesting mechanism proposed, related to clay swelling, was the buckling of the wet parts of the rock. As explained in Appendix C, when saturation of the sample is not complete, there are parts that are saturated and others that remain dry. As clay swelling happens in parallel and perpendicular direction to bedding, wet layers exhibit buckling (Figure 52). Buckling produces internal stresses, resulting in the degradation of the rock. Especially when initial flaws are present, buckling can also result in the detachment of grains and reduction of contact areas. Of course, grain contact areas can also be reduced by clay swelling itself, as presented in Figure 53.

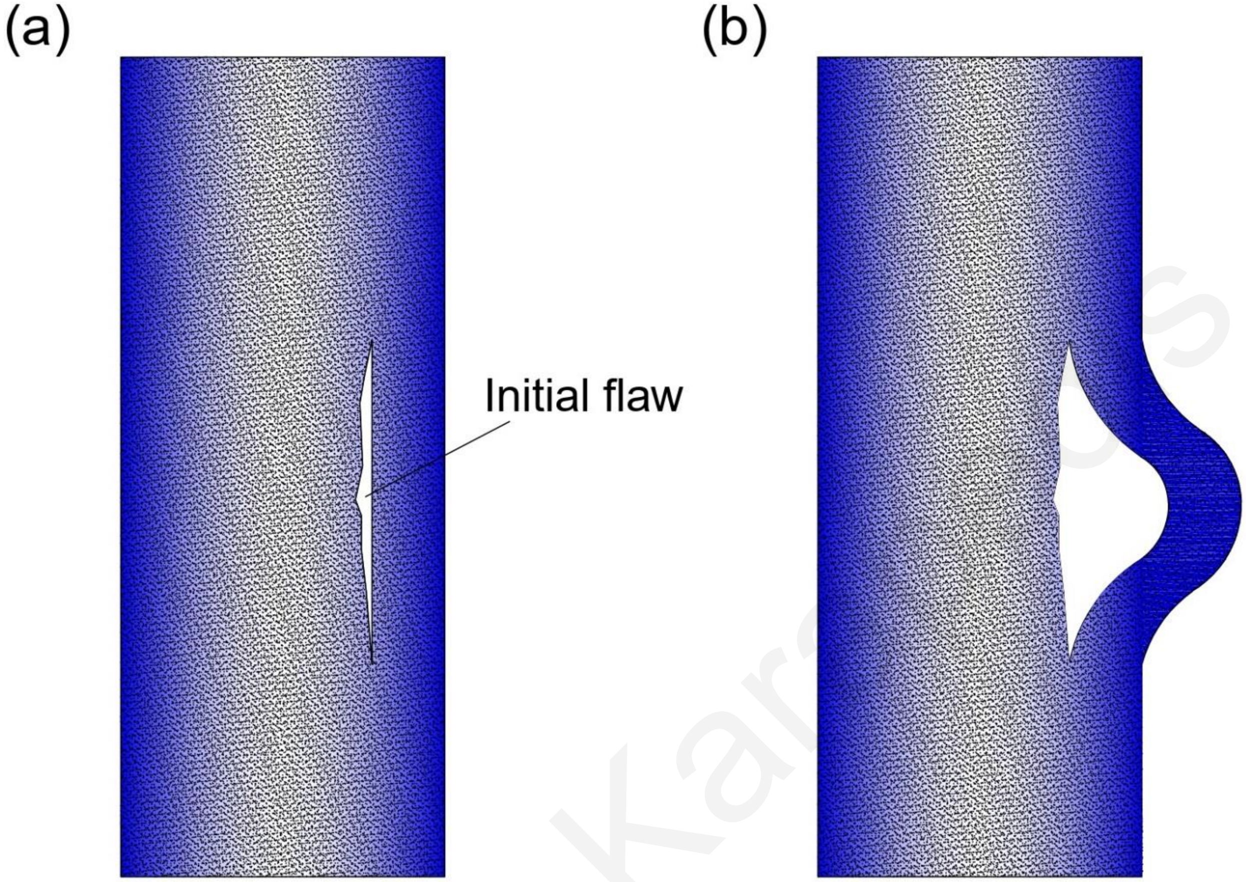


Figure 52 - Buckling in sandstone. (a) before and (b) after buckling

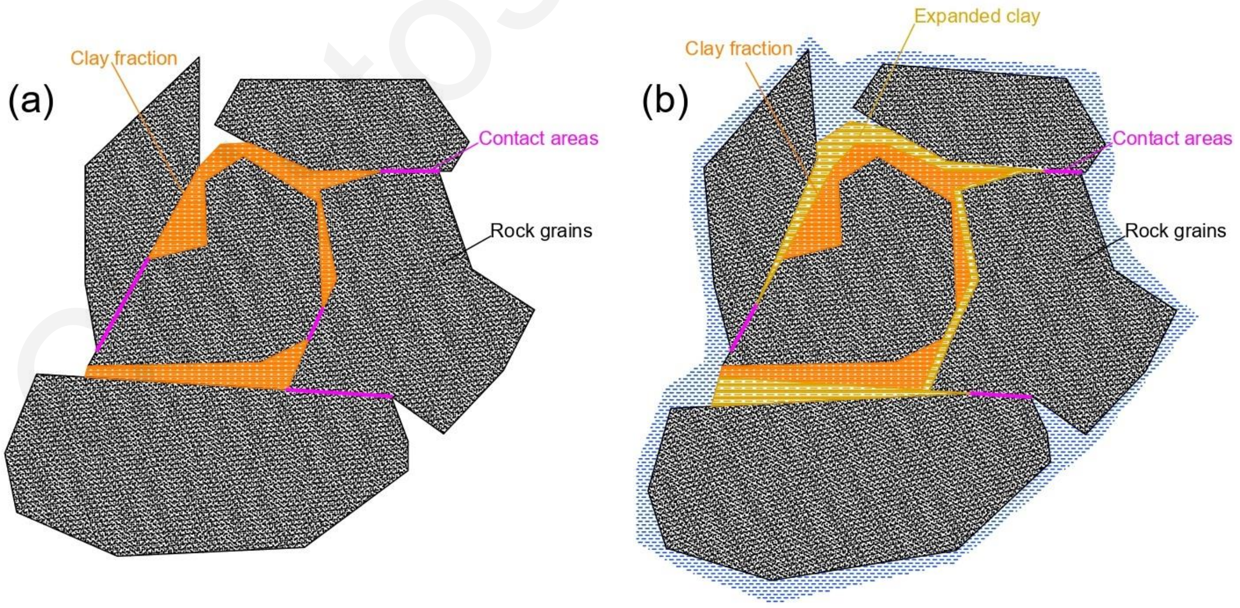


Figure 53 - Reduction of grain contact areas due to clay swelling. (a) before and (b) after clay swelling

The reduction of contact areas should be considered of high significance, as it has been proved by Dyke & Dobereiner (1991), that there is a great correlation between wet UCS and grain contact area (Figure 54). As grain contact area increase, wet UCS also increases. Reduction in grain contact area will greatly affect the wet UCS.

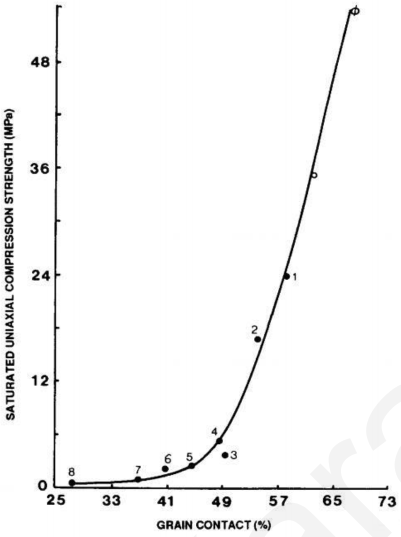


Figure 54 - Relation between saturated UCS and grain contact area (from Dyke & Dobereiner (1991))

Lu et al. (2017), performed tests on sandstones at different water temperatures. They observed that as temperature increased, the maximum water content decreased, indicating that porosity decrease was greater for higher water temperatures, and thus, swelling was more intense in higher temperatures. At the same time, for higher temperatures, the wet UCS was higher than for lower temperatures (Figure 55). Unfortunately, the pore size of the sample is not provided.

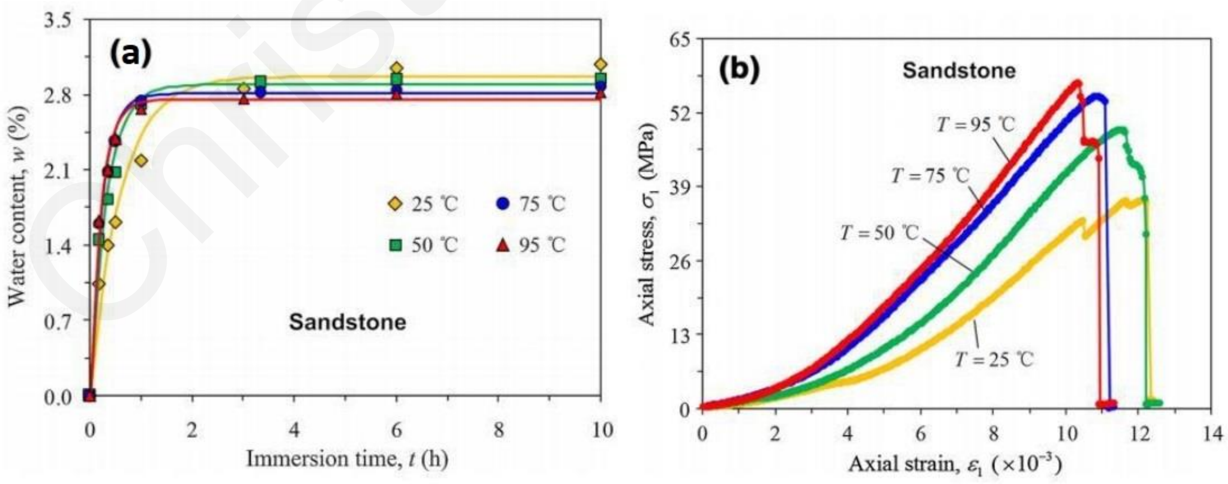


Figure 55 - (a) water content vs immersion time and (b) axial stress vs axial strain in sandstones for different temperatures (from Lu et al. (2017))

On one hand, UCS is usually expected to be higher for lower porosity values, but on the other hand, as swelling is more intense, swelling stress is expected to increase. In this case, the negative effects of the extra swelling seem to be overruled by some positive ones. The positive effects could be explained as following: As clay expands into the pore, it will get in contact with other grains. In this case, new contact areas will be created and the compressive stress into the rock will be reduced, since the total contact area will increase. On the other hand, extra friction resistance will develop on the new contact points of grains (Figure 56). These new contact points and the extra resistance they provide, could explain why UCS reduction was lower for more intense clay swelling. Concluding, it could be suggested that, under specific conditions, clay swelling could also have a positive effect in wet UCS.

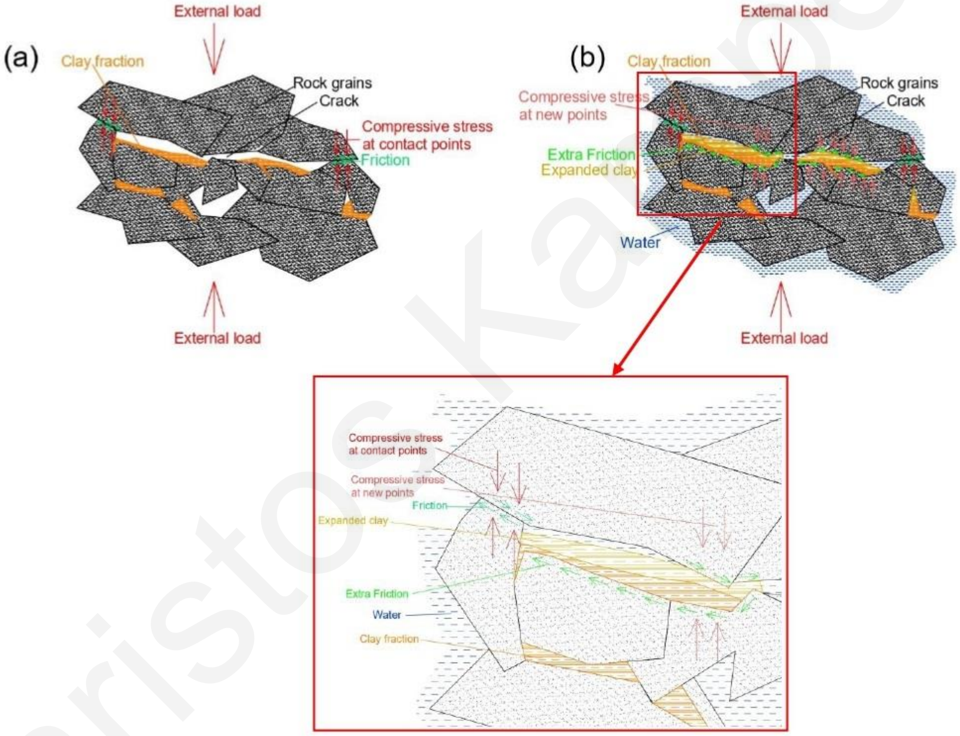


Figure 56 - Formation of new contact points after clay swelling. Clay (a) before and (b) after swelling

Another aspect concerning the wet behavior of clays and their influence in sandstone's wet UCS, is the reduction of friction coefficient of clays, especially when clay acts as a cement material in the rock (Lu et al., 2017). In some experiments, the failure type of sample transformed from tensile or brittle, to shear, as saturation increases (Cai et al., 2019; Tang, 2018). What is more, in other experiments, it was observed that while the cracks of the dry samples were mostly intra-granular, in the wet state, inter-granular cracks prevailed (Figure 57) (H. Li et al., 2019; Tiennot et al., 2019). Such observations give rise to the importance of the friction developed between rock grains and clay fraction, especially when clay acts as cementation.

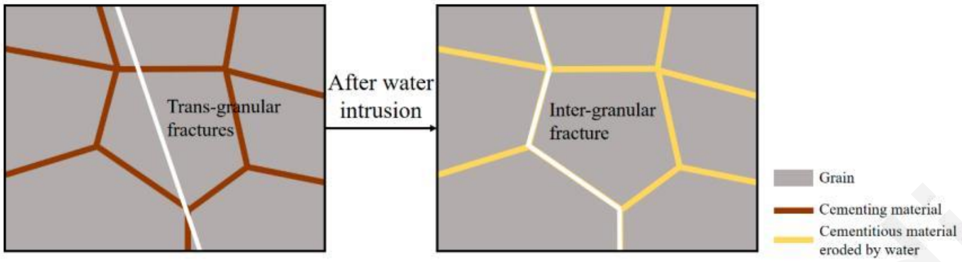


Figure 57 - Intra-granular (or trans-granular) and inter-granular cracks (from Tiennot et al. (2019))

Lin et al. (2005) made similar observations in rocks with high cementation proportion; for rocks with less cementation and higher grain proportion, the crack type remained mostly the same. This seem reasonable, since in order for inter-granular crack to propagate and to result in the breakage of the sample, a “cement path” needs to be available, and as the cementation increases, such paths are more likely to exist.

Figure 58, shows the of UCS loss in sandstone, versus their clay content. Note that, for the same reasons discussed in 3.6, the term “clay content” includes all of the clay minerals content. The correlation does not seem strong, as R^2 value is equal to 0.1361 (not shown in the chart). This fact may indicate two things: (i) apart from clay related mechanisms, other mechanisms should have a great impact on UCS loss in sandstones (see Appendices), and (ii) clay content alone is not enough to express the effect of clay minerals in UCS loss in sandstones. Clay content should be considered in addition to other rock characteristics, like the pore size distribution described before. In any case, no safe conclusions can be made with the existing literature data.

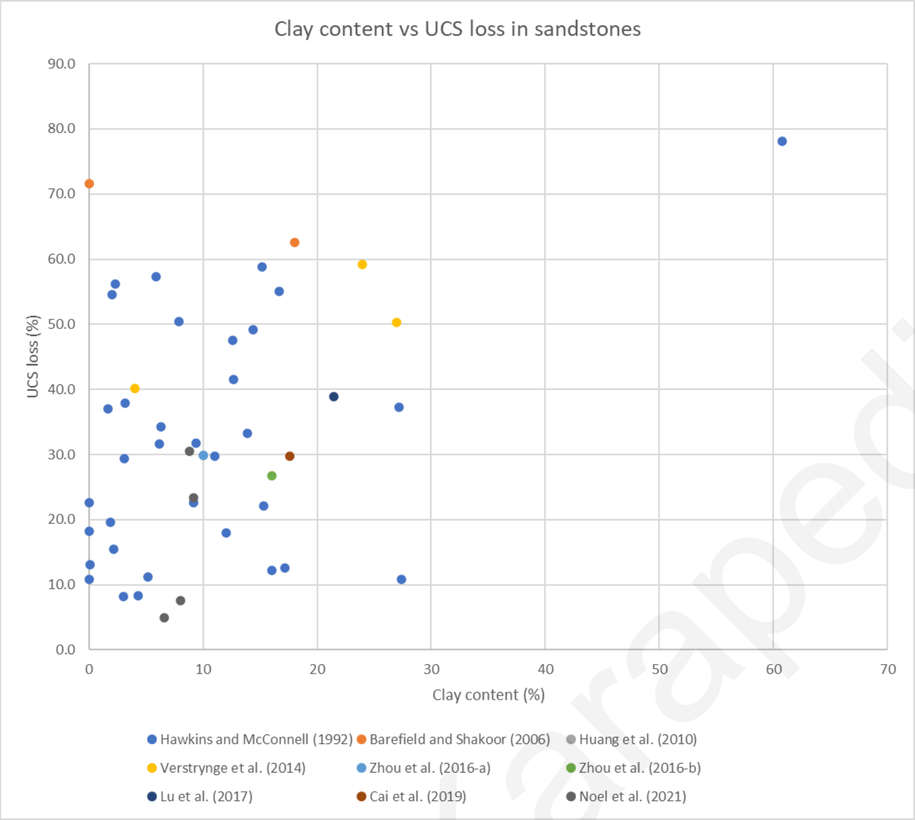


Figure 58 - Clay content vs UCS loss in sandstones. Data obtained from Hawkins & Mcconnell (1992), Shakoor & Barefield (2006), S. Huang et al. (2010), Verstrynge et al. (2014), Z. Zhou et al. (2016), ZHOU et al. (2016), Lu et al. (2017), Masoumi et al. (2017), Tang (2018), Wasantha et al. (2018), Cai et al. (2019) and Noël et al. (2021)

4.2 Time dependency

In spite of rocks where clay minerals are dominant (like mudstones and siltstones examined before), sandstones will not collapse, or totally disintegrate, after some immersion time. As long as swelling of clay is completed, no other weakening mechanism further proceeds with immersion time, except from the dissolution of calcite. But as explained in Appendix B, when the immersion liquid is pure water distilled water, a very long time is needed to dilute an important amount of calcite. Also, calcite is usually in small amounts in sandstones. So, it can be assumed that clay swelling is the only controlling factor to determine the time dependency of sandstone's UCS reduction.

Figure 59, presents the UCS of sandstones in wet condition, after different immersion times (Tang, 2018). In this case, swelling of clay completed after 48 hours. It can be seen that the UCS of sandstone reduced for the first 48 hours, as swelling proceeded. After 48 hours, UCS remained the same; see that for 960 hours immersion time, the UCS was equal to that of 48 hours. Note that, in this sandstone, calcite was presents, and its content was 4.29%.

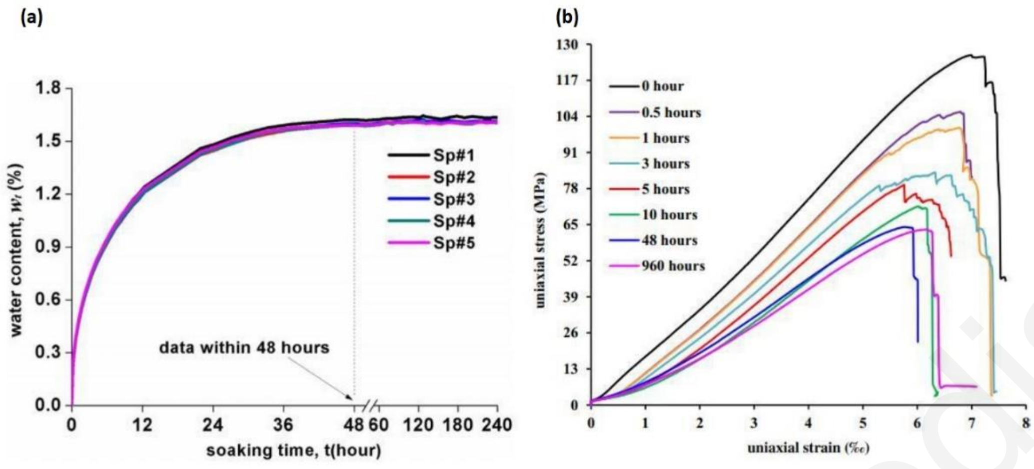


Figure 59 - (a) soaking time vs water content, (b) axial strain vs axial stress for samples immersed in water for different time (from Tang (2018))

No other experiments that investigate the reduction of UCS with immersion time, were found in the literature, at least under author's knowledge.

4.3 Wetting and drying cycles

The general behavior of sandstones under wetting and drying cycles, is that the UCS is reduced gradually for some number of cycles, and as their number increase, reduction becomes mild, until a point where it gets insufficient. Figure 60, shows the reduction of dry UCS after wetting and drying cycles for different sandstones. Data were obtained from Liu et al. (2018), Xie et al. (2018), S. Huang et al. (2018), Yao et al. (2020), Liu et al. (2021), X. Li et al. (2021), Zhao et al. (2021).

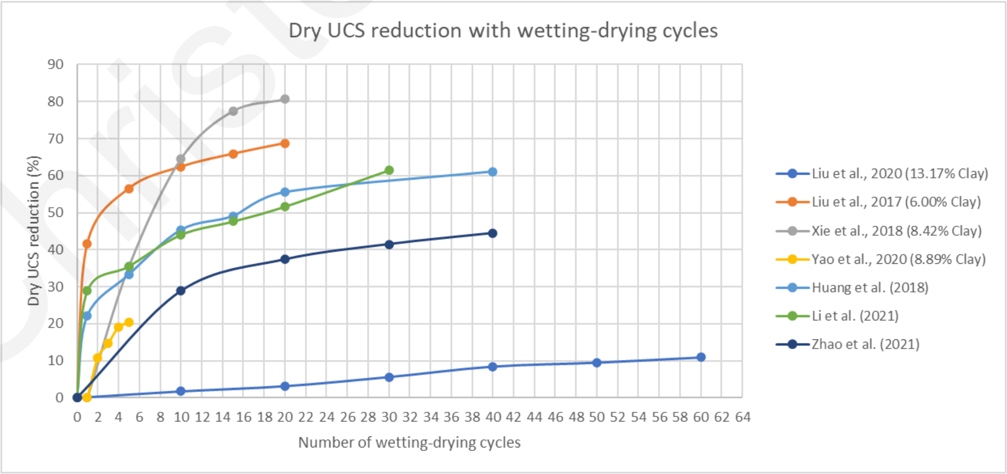


Figure 60 - Reduction of dry UCS vs number of wetting-drying cycles. Data obtained Liu et al. (2018), Xie et al. (2018), S. Huang et al. (2018), Yao et al. (2020), Liu et al. (2021), X. Li et al. (2021), Zhao et al. (2021)

For the cases where mineralogy is given, the clay content is written in the legend, in order to observe the relation between clay content and the reduction of strength. Unfortunately, the existing data are limited, and no clear correlation can be made. However, it seems that there is no solid correlation; the sandstone with the highest clay content shows the least UCS reduction, while the samples with 8.42% and 6.00% clay content show a reduction of 81% and 69% on 20th cycle. The sandstone with 8.89% clay content, shows a reduction of 20% in the 5th cycle, while the sandstone with similar clay content (8.42%) shows a reduction of 35% on the 5th cycle. Here too, the idea to examine the clay content with respect to pore size distribution, seems attractive. However, no data are available in the literature.

Figure 61 shows the fluctuation of porosity with wetting and drying cycles, with data obtained from Z. Zhou et al. (2018), S. Huang et al. (2018) and Xie et al. (2018); clay content is not available in any of these articles. The tendency is the same as for the case of UCS reduction; porosity increases with decreasing grade, until a point where the increase is insufficient. What is interesting, is that for the only cases with available data for both porosity and UCS change, the change in these two characteristics is very similar. For the sandstone of S. Huang et al. (2018), the increase of porosity after 40 cycles is 58% and the reduction of UCS is 61%, and for the sandstone of Xie et al. (2018), the increase in porosity after 20 cycles is 85% and the reduction of UCS is 81%. In addition, the curves of porosity and UCS fluctuation with increasing wetting and drying cycles, look similar. The above indicates that dry UCS reduction after wetting and drying cycles is governed by the increase in porosity.

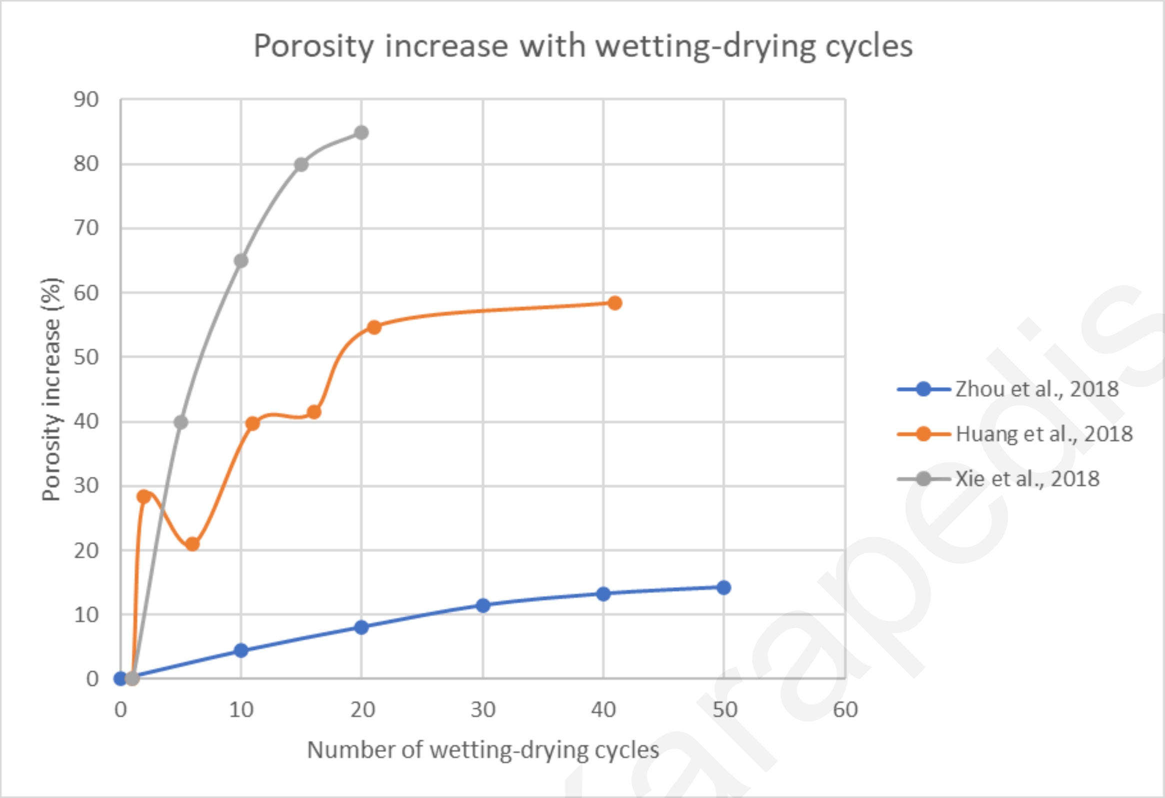


Figure 61 - Porosity increase vs number of wetting-drying cycles. Data obtained from Z. Zhou et al. (2018), S. Huang et al. (2018) and Xie et al. (2018)

The aforementioned internal changes, could be explained as follows: During wetting and drying cycles, microcracks start to appear. As cycles increase, their quantity increases as well, and they become wider and longer Z. Zhou et al. (2018). The formation and propagation of microcracks may be due to clay swelling and air-breakage phenomenon, i.e. during saturation, air is trapped into the pores and cannot escape, leading to an increase in air pressure, and thus, in the total internal pressure of rock (Doostmohammadi et al., 2009; José Pejon & Valentin Zuquette, 2002). At the same time, intergranular cracks show up, and increase in number with increase of cycles (Guo et al., 2021), until a point where there are hardly no grains without cracks on their boundaries (Guo et al., 2021; Z. Zhou et al., 2018). Figure 62, presents SEM images of a sandstone exposed at wetting and drying cycles, showing the internal changes in the microstructure at different cycles (Z. Zhou et al., 2018).

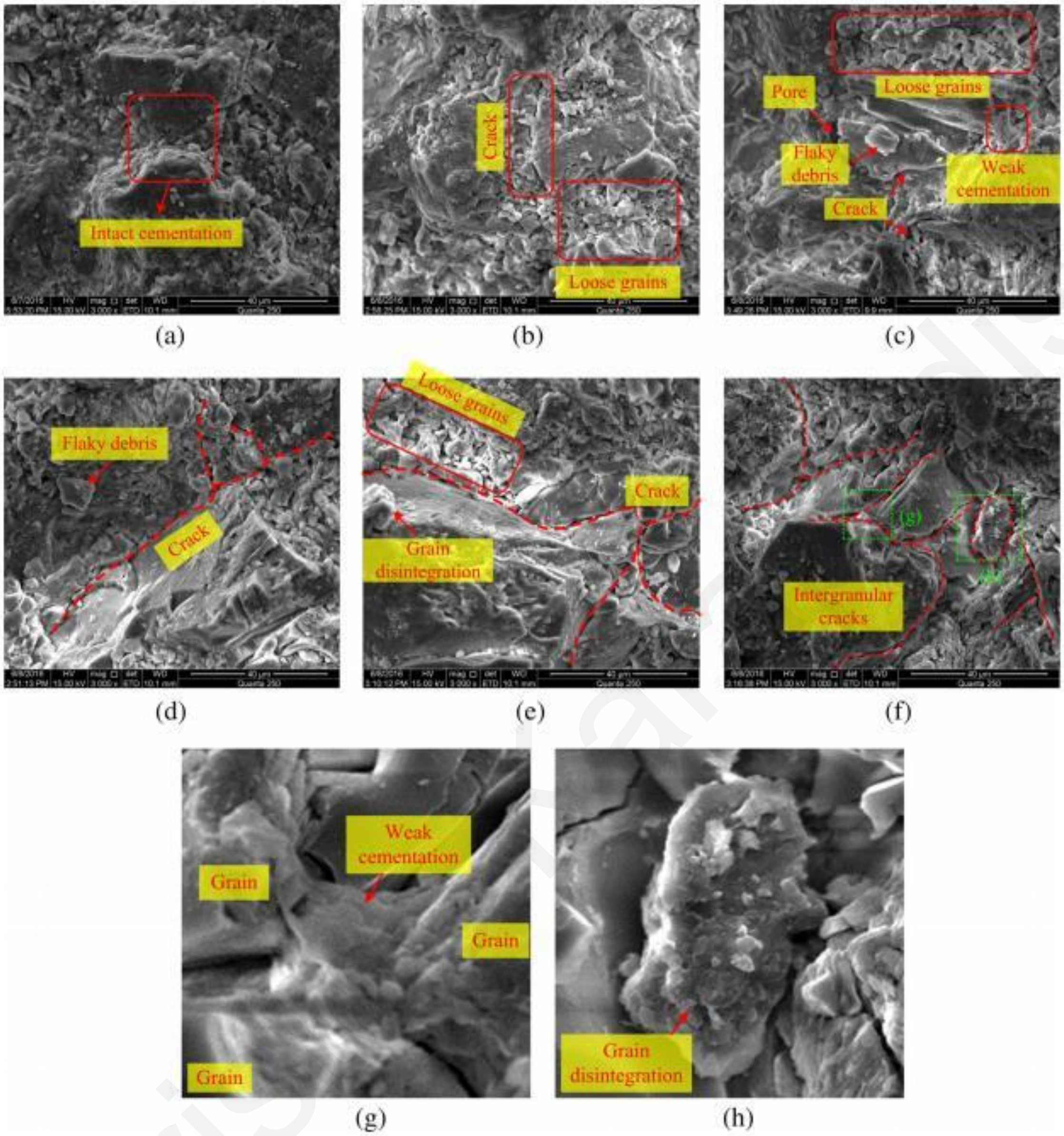


Figure 62 - SEM images of sandstones at different wetting-drying cycles. (a) - (f) present images after 0, 10, 20, 30, 40 and 50 cycles, (g) and (h) depict the magnified views marked with green boxes in (f) (from Z. Zhou et al. (2018))

What is more, clay fraction is found to be reduced after wetting and drying cycles (Xie et al., 2018; Yao et al., 2020), leading, on one hand in porosity increase, and on the other hand, in the decreasing of rock cementation. The clay fraction reduction can be attributed at clay leaching (Lin et al., 2005); as water flow into the pores, some clay fraction is washed out. This phenomenon was described above, for the case of a simple saturation, but it should be more intense in the case of wetting and drying cycles, since water flow is repeated many times. In Xie et al. (2018) and Yao et al. (2020), it was found that clay content reduced by 27% and 20% after 20 and 5 cycles, respectively.

4.4 Saturation with Sodium Chloride

Figure 63 presents the wet UCS for sodium chloride solutions for different NaCl concentrations (0% concentration refers to distilled water), after 5 relative experiments. Data were obtained from Shukla et al. (2013), Rathnaweera et al. (2014), Nasvi et al. (2014), Rathnaweera et al. (2015) and Y. H. Huang et al. (2018). Figure 64 shows the variation of wet UCS for the case of distilled water and different salinity concentrations, with data from the same experiments (the case of Rathnaweera et al. (2015) was excluded because no uniaxial compression test for distilled water was performed).

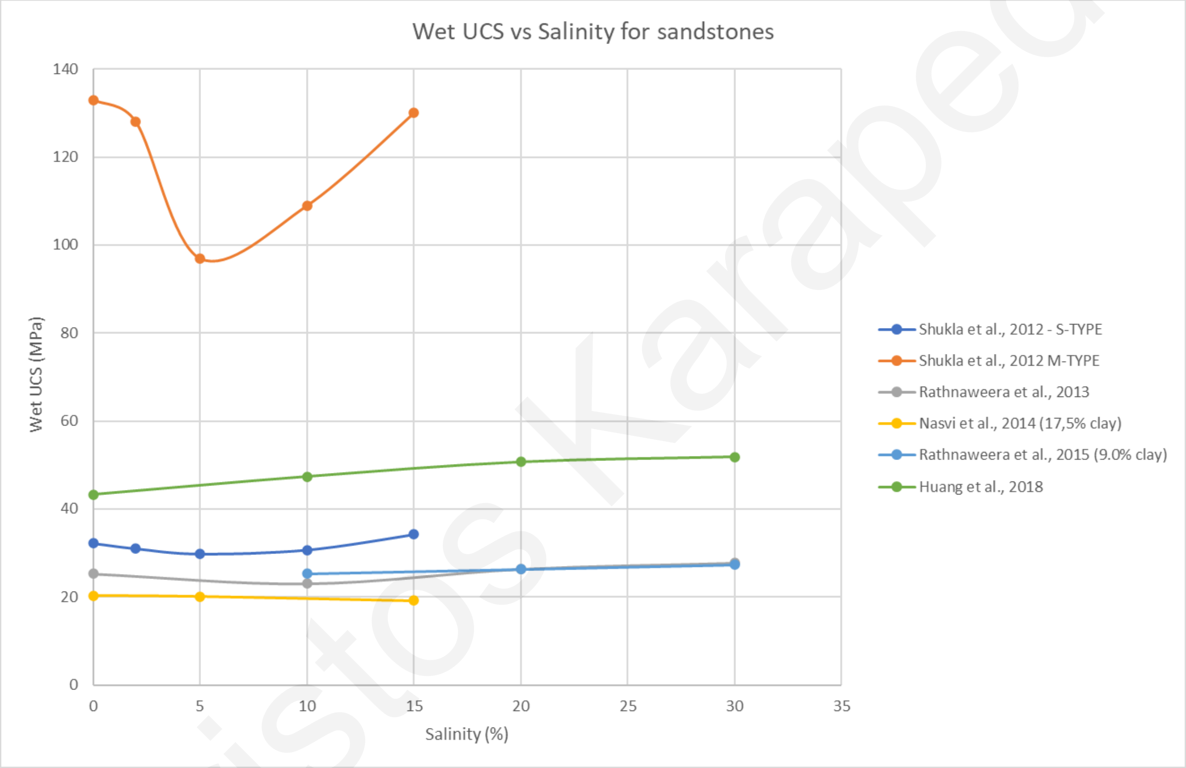


Figure 63 - Salinity vs wet UCS for sandstones saturated with solutions with different NaCl concentrations. Data obtained from Shukla et al. (2013), Rathnaweera et al. (2014), Nasvi et al. (2014), Rathnaweera et al. (2015) and Y. H. Huang et al. (2018)

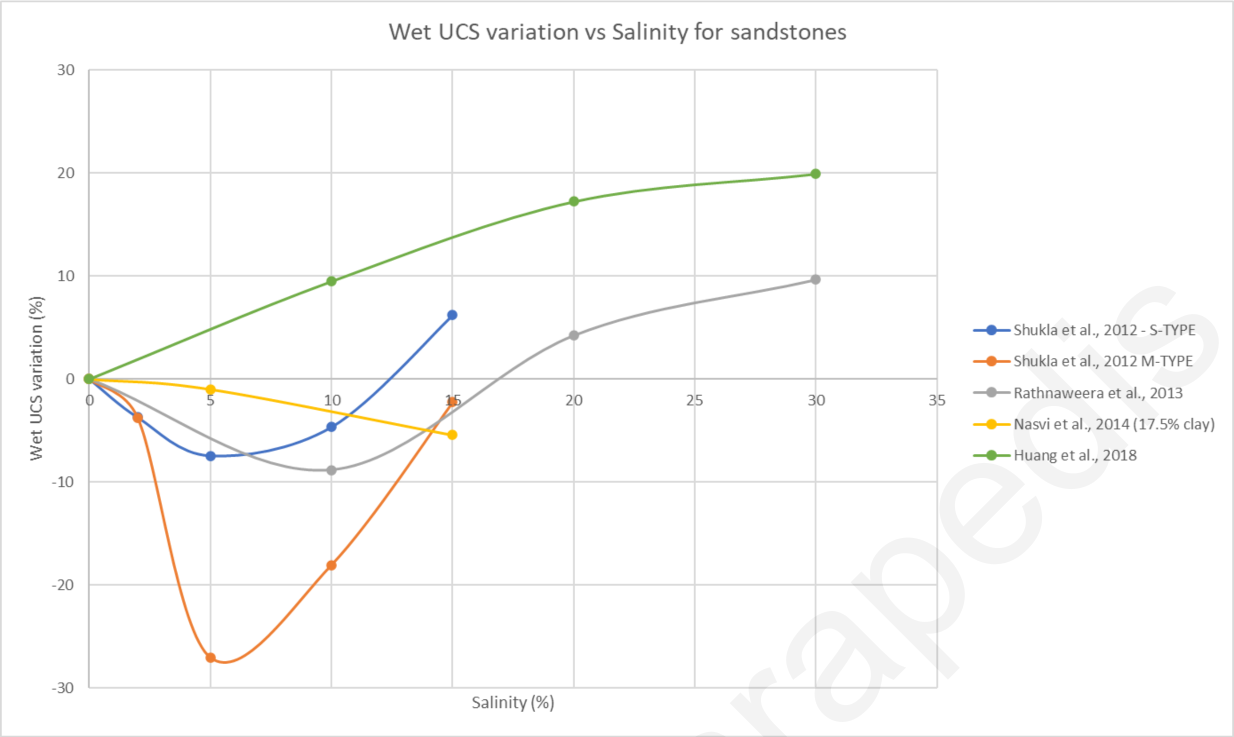


Figure 64 - Salinity vs wet UCS variation (comparing to pure water) for sandstones saturated with solutions with different NaCl concentrations. Data obtained from Shukla et al. (2013), Rathnaweera et al. (2014), Nasvi et al. (2014) and Y. H. Huang et al. (2018)

In general, for NaCl concentration above 10%-15%, the wet UCS of the sandstones is higher than that of distilled water, and for concentrations below that, it gets lower. The cases of Nasvi et al. (2014) and Y. H. Huang et al. (2018) do not show the same trend, but their tests did not involve solutions in the critical concentrations. Solutions with concentrations higher than 15% are absent in the former, and between 0% and 10% in the latter. If these cases were included, their curves could possibly follow the same trend. Note that, dry tests were only included in Shukla et al. (2013) and Rathnaweera et al. (2014), and in all three sandstones, the dry UCS was higher than any wet UCS.

It seems that there are both positive and negative effects of NaCl in sandstone's UCS, and that the positive ones overrule the negative ones, at concentrations 10%-15% and above. The positive effect is attributed to the formation of crystals into the pores of the rock. First, the formation of crystals decreases the pore space, and thus, porosity of the rock decreases (Rathnaweera et al., 2014). Rathnaweera et al. (2015), found that for NaCl concentrations of 10%, 20% and 30%, porosity showed values of 28.88%, 27.47% and 24.48% respectively. Figure 65, presents SEM photos of Rathnaweera et al. (2015) sandstone, after saturation with 0%, 10%, 20% and 30% NaCl concentrations. It is observed that crystals amount increases with increasing concentration. Furthermore, crystals between the grains, enhance the friction between grains (Rathnaweera et al., 2015), and especially at the case of loose grains, this could provide an extra friction resistance on the sample. What is more, crystals could provide an extra support at grains, since the compression strength of a crystal is higher than that of water.

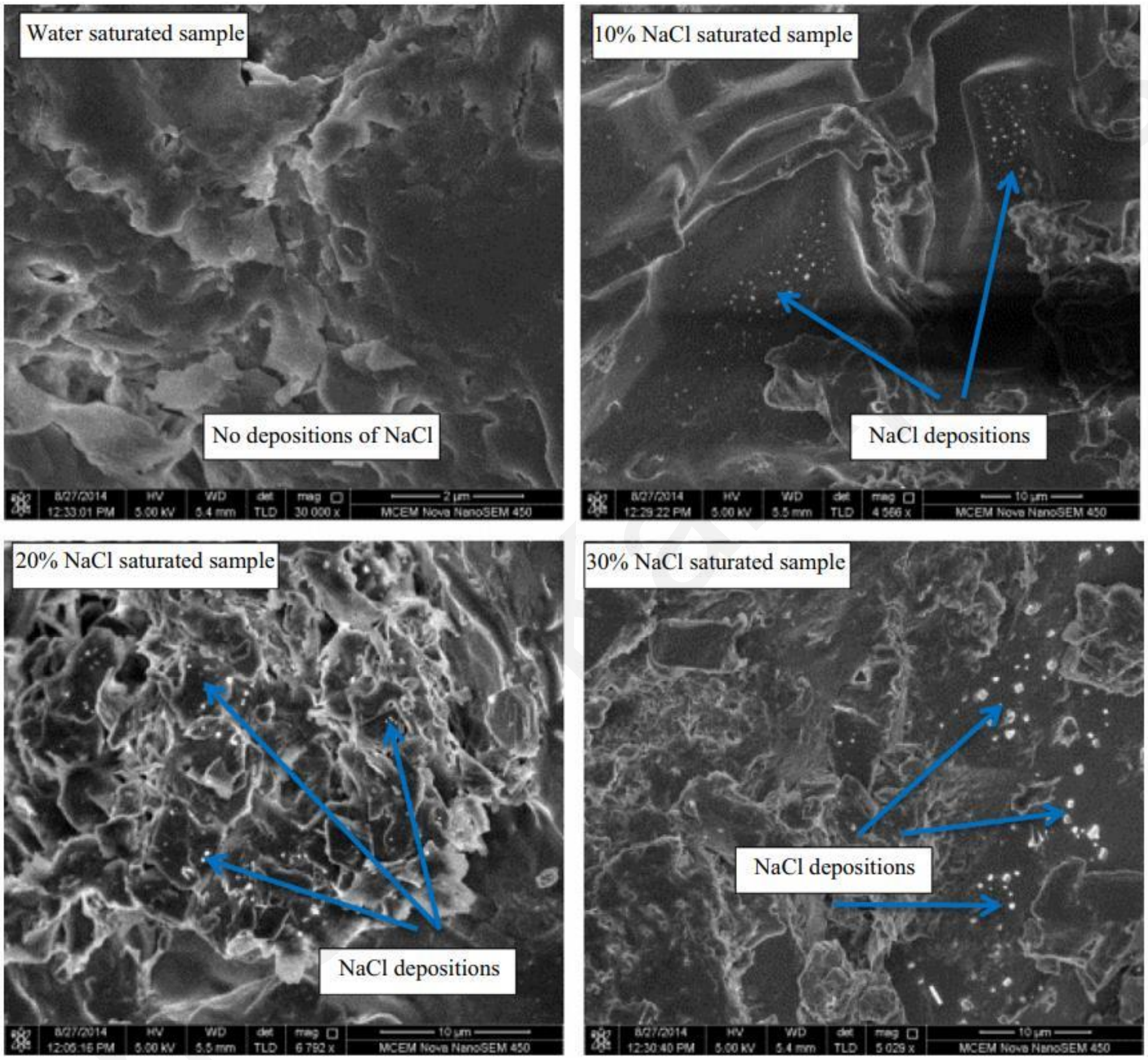


Figure 65 - SEM photos of sandstone after saturation with different NaCl concentrations (from Rathnaweera et al. (2015))

Another positive aspect, could be related to the limited swelling of clay minerals. As explained in 2.4, swelling of clay minerals is expected to be lower as cation concentration increases. This limited swelling, results in limited internal swelling stress, and so, changes in the microstructure should be less.

One possible explanation about the negative effect of NaCl could be the higher reduction of surface energy of the grains, in the presence of NaCl (Rathnaweera et al., 2014), or the higher corrosion of minerals. In any case, this phenomenon should be examined more.

4.5 Water content (or saturation degree) dependency

As described in 3.5, after many tests on different sandstones, Hawkins & Mcconnell (1992) proposed that the relation between UCS and water content, is expressed by Equation 1:

$$\sigma_c = a \cdot e^{-b \cdot w} + c \qquad (2)$$

Later, many researchers confirmed the validity of this Equation 1 on other tested sandstones, (Babets et al., 2020; Lin et al., 2005; Masoumi et al., 2017; Tang, 2018; Z. Zhou et al., 2016).

Figure 66 shows the curve between saturation degree and the percentage of the total UCS loss of sandstones, with data obtained from the aforementioned articles. It can be seen that, for all sandstones, most of the total UCS is lost at lower saturation degrees. 40% of the total UCS loss happens at about 20% saturation, while at 40% saturation, approximately 60% of the total UCS has occurred for 3 sandstones, while two of them exhibited 90% of their total UCS loss. After that point, the UCS keeps reducing, but with constantly reducing rate.

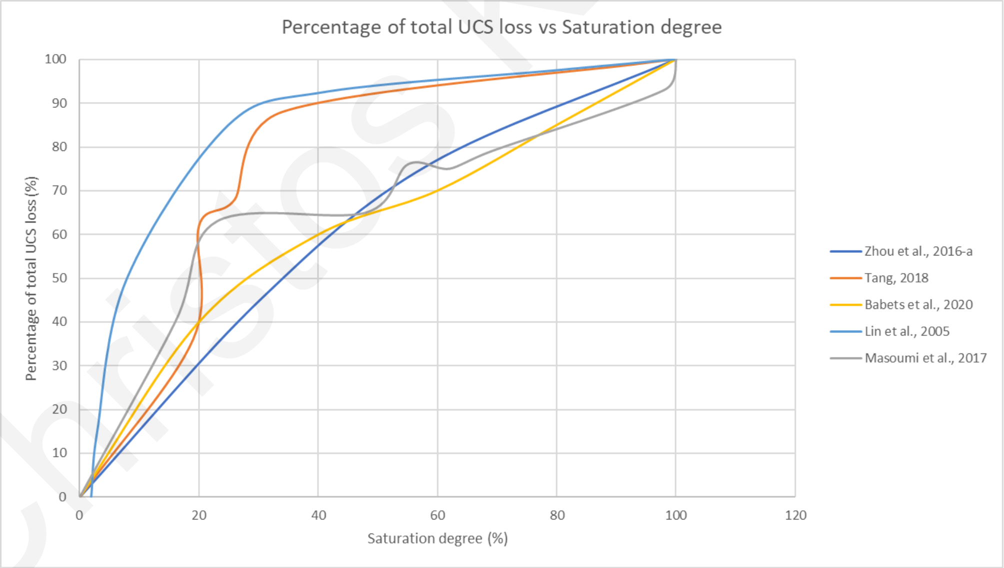


Figure 66 - Saturation degree vs percentage of total UCS loss. Data obtained from Lin et al. (2005), Z. Zhou et al., 2016), Masoumi et al. (2017), Tang (2018) and Babets et al. (2020)

Tang (2018) explained this behavior taking under consideration the water distribution in samples and their failure mode. He found that while the examined sandstone suffered shear failure at dry state, as soon as it got saturated even with a small amount of water, tensile or brittle failure occurred. As the degree of saturation increased, the failure mode turned to tensile-shear or shear, with shear failure becoming dominant at high saturation degrees. What is more, at lower saturation degrees, cracks developed near the surface of the sample, i.e. at the parts of the rock that were saturated. He suggested that this happened because, during compression, the saturated parts are more prone to lateral deformations than dry ones, and so, at the interface of dry and wet parts, tensile stresses developed, resulting in splitting of whole parts of the sample, and thus, in brittle or tensile failure.

Concluding, it could be stated that wet parts of the sample provide enough available space for cracks to develop, in lower compression stress comparing to dry parts. This fact indicates that the strength of the whole sample is determined by the behavior of the wet parts, even if these parts represent a small proportion of the whole sample. Under these terms, the trend of sandstones to lose most of their strength in the lower saturation degrees can be explained. Of course, as water in the rock body increases, the bigger pores start to fill with water, thus, more weakening mechanism activate (like the pore pressure increase), and this is why at higher saturation degrees the UCS still decrease.

Chapter 5 General conclusions

Because of their unique chemical structure, clay minerals are associated with the ability to absorb water molecules into their structure and their surface. Water absorption into the structure causes clay minerals to expand, a phenomenon called “swelling”. This phenomenon is partially reversible; when clay minerals expel the absorbed water, they shrink. Swelling of clay minerals has two components: crystalline and osmotic swelling. The magnitude of crystalline swelling a mineral will exhibit, highly depends on the interlayer cation. It is expected that when the interlayer is filled with sodium and calcium cations, the crystalline swelling will be greater, while the exact opposite behavior is expected to happen with potassium. Osmotic swelling depends mostly by the cationic concentration of the liquid; the higher the concentration, the lesser the osmotic swelling. Water adsorption on the surface of the clays, is related with friction loss.

Smectite group seem to suffer the most swelling and friction loss than any other clay mineral. Thus, many researchers focus only on the existence of smectite, like montmorillonite or beidellite, when examine the influence of clay mineral on saturated rock strength and other properties. However, with the present study, it is suggested that this approach is incorrect, because even rocks with no smectite content are found to suffer changes in strength when saturated, and also, all clay minerals present a reduction in friction coefficient under wet conditions.

In order to investigate the influence of clay minerals in rock uniaxial compressive strength, two rock groups examined. The first group was about mudstones, siltstones and marls, that is, rocks in which clay is the dominant mineral of their composition. The secons group was the sandstone type alone. Sandstones are rocks consisting mostly by grains the size of sands, and most of them contain a portion of clay minerals, usually in the order of 5%-10%. The percentage of UCS reduction is both groups is found to be in a large range of values. For the case of sandstones, the reduction may vary from 5% to 60%, while for mudstones, siltstones and marls may vary from 40% to even 95%. Thus, the first impression is that clay content indeed plays an important role on the reduction of UCS loss in rocks, since rock types with high clay content seem to suffer greater UCS loss.

Since mudstones and siltstones consist mostly of clay minerals, it is reasonable to state that the basic mechanisms responsible for their UCS reduction, are related with to the reactivity of clay with water. This is the common opinion between the relevant projects, and is something that has been confirmed experimentally. Through the observation of the structure of such rocks, and how it changes during saturation time, the basic conclusion is that clay swelling is responsible for severe internal damage inside mudstone and siltstone body. This damage leads to the decrease of rock quality, and can even result in total disintegration of the sample, if the sample is immersed in water for some time. Thus, the reduction of such rock's strength is time-dependent.

For the case of sandstones, it is hard to mention which mechanism is more significant, because all of the proposed mechanisms are activated (see also Appendices), and throughout the range of different sandstones tested, many different behaviors have been observed. However, clay reactivity is believed to be an important reason for sandstone's reduction in strength. Clay swelling causes internal changes in sandstones, too. However, there are two main differences in sandstones: (1) the grain skeleton is strong enough to resist the development of internal swelling stresses, and (2) as soon as swelling completes, no extra change happens inside sandstone's body, or else, the structure changes do not follow a progressive procedure. So, from that point of view, the reduction of UCS in sandstone is time-dependent, but only until clay swelling is over. After this point, the only mechanism progressing with time, is the dissolution of calcite, but it takes a great amount of time for calcite to dissolve in pure water. Reduction of friction coefficient of clay minerals should also contribute to strength reduction of both rock groups.

What is more, since clay swelling depends on the interlayer cation, one would expect that the liquid chemistry would affect the wet behavior of rocks with clay portion; and this is true. It has been found that sandstones and mudstones saturated with water with cationic concentrations, show a different behavior than that of pure water. It is interesting, though, that such a behavior is not clear; greater amount of NaCl or KCl in water does not necessarily mean a higher or a lower wet strength. It seems that there is a cationic concentration at which the wet UCS becomes minimum, comparing with pure water, and after this point, wet UCS increases and becomes higher than pure water. In any case, wet samples present a lower strength than dry ones.

The relation between UCS and water content (or saturation degree), follows a negative exponential curve (see Equation 1). This suggestion was made by Hawkins & Mcconnell (1992) after investigating the strength reduction of many sandstones. This relation has been confirmed by all subsequent studies on sandstones, but also applied in all mudstones and siltstones examined. Marls may not follow the same trend, but it needs more experiments to make a solid conclusion.

In addition, cycles of wetting and drying has been proved to affect the wet UCS in sandstones, mudstones and siltstones. The behavior of all rocks is similar. Wet UCS decreases by each cycle, with the rate of reduction being higher at lower cycle number, and reducing as cycles increase, until reaching a number of cycles where no extra reduction is observed.

From the aforementioned, one could expect that there would be a strong correlation between clay content and UCS loss in every rock. Such a correlation must exist, since it has been proven that clay reactivity affects the wet UCS. However, after processing the available data in literature, such a correlation does not seem strong (see Figure 58 and Figure 41). Maybe, clay content itself is not sufficient enough to indicate the loss of UCS, and needs to be examined in addition to some other rock property. In the present study, it is suggested that this combination should be between clay content and pore space. In the case of rocks with high clay content, there should be parts of clay fraction that do not get in touch with water when saturated, thus, one should consider the total interface between clay and pore water. In the case of sandstones, where it should be assumed that most of clay fraction gets in touch with water (low clay fraction, high porosity), the pore size distribution should be taken into account. That is because, in a rock with bigger pores, clay will swell “freely” and swelling stresses will not be developed, while in a rock with smaller pores, clay swelling will be confined, resulting in the development of swelling stresses. Unfortunately, the existing literature data are not enough to confirm such a hypothesis.

Appendix A Fracture toughness and Subcritical cracking

Fracture toughness refers to the ability of a rock, or any other material, to resist in the formation and propagation of cracks into its body. To describe the crack behavior of a rock, the intensity factor K_I has been widely used. Intensity factor K_I, expresses the magnitude of the stress concentrated on a crack tip, and is given by Equation 2 (Kean Atkinson, 1982)

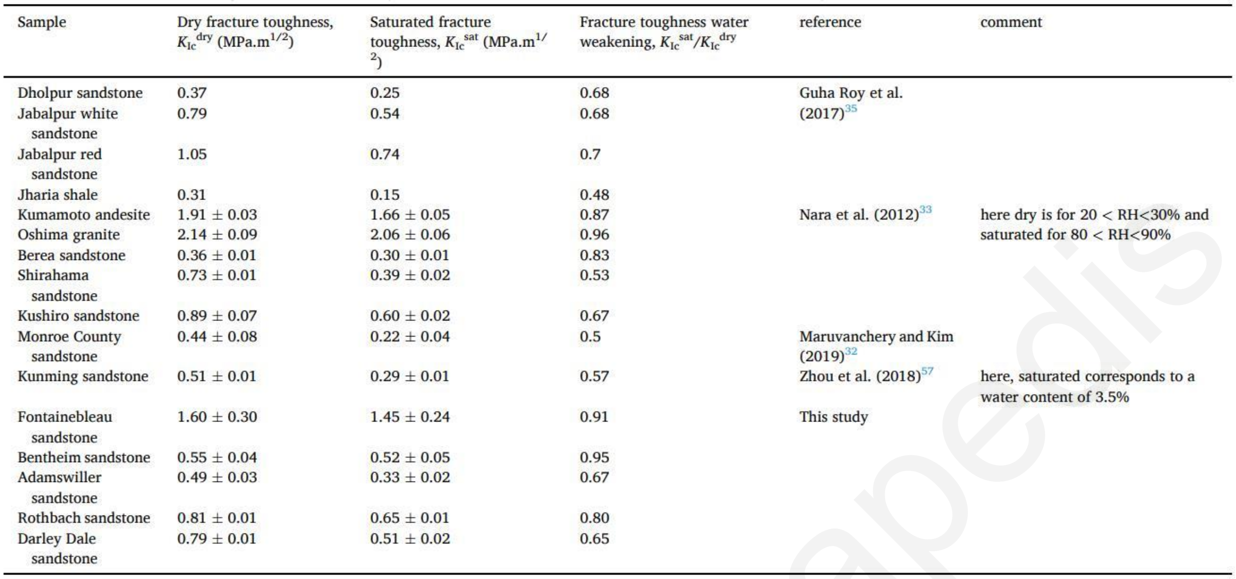
$$K_I = Y \cdot \sigma_a \cdot a^{1/2} \qquad (3)$$

where Y is a geometrical constant, σ_a is the applied stress and a is a characteristic crack length. When the intensity factor in a rock overcomes a critical value, K_{Ic}, cracks inside the rock body start to propagate. After observing Equation 2, it can be seen that, in order for the intensity factor to have values above 0, an initial crack must be present. However, rocks are considered to be heterogenous, due to their porous and grain structure, thus, initial cracks or flaws are always present (Eppes & Keanini, 2017).

According to Van Eeckhout (1976), fracture energy (i.e. the energy required for a crack to expand) depends on the surface energy of the media on which cracks propagate. The surface energy of the rock-water surface is lower than that of the rock surface itself, so, after water fills the pores of rock, the fracture energy reduces. Thus, it is expected that critical intensity factor of a rock decreases from dry to wet state, indicating that crack propagation starts at a lower compressive stress, under saturation.

Such observations were made by some researchers after relative experiments on sandstones, indicating that the critical intensity factor is indeed reduced under saturate conditions (Noël et al., 2021; Z. Zhou et al., 2018). Table 7 presents the data available on literature concerning the critical intensity factor reduction and Figure 67 represents the relation between water content and critical intensity factor, as found in Z. Zhou et al. (2018). It can be seen that the best fit curve is a negative exponential one, similar to that suggested from Hawkins & Mcconnell (1992) between water content and UCS. Note that in both Table 7 and Figure 67, the critical intensity factor is expressed just as “fracture toughness”.

Table 7 - Literature data of fracture toughness under both dry and water-saturated conditions (from (Noël et al. (2021))



Sample	Dry fracture toughness, K_{Ic}^{dry} (MPa.m$^{1/2}$)	Saturated fracture toughness, K_{Ic}^{sat} (MPa.m$^{1/2}$)	Fracture toughness water weakening, $K_{Ic}^{sat}/K_{Ic}^{dry}$	reference	comment
Dholpur sandstone	0.37	0.25	0.68	Guha Roy et al. (2017)[35]	
Jabalpur white sandstone	0.79	0.54	0.68		
Jabalpur red sandstone	1.05	0.74	0.7		
Jharia shale	0.31	0.15	0.48		
Kumamoto andesite	1.91 ± 0.03	1.66 ± 0.05	0.87	Nara et al. (2012)[33]	here dry is for 20 < RH<30% and saturated for 80 < RH<90%
Oshima granite	2.14 ± 0.09	2.06 ± 0.06	0.96		
Berea sandstone	0.36 ± 0.01	0.30 ± 0.01	0.83		
Shirahama sandstone	0.73 ± 0.01	0.39 ± 0.02	0.53		
Kushiro sandstone	0.89 ± 0.07	0.60 ± 0.02	0.67		
Monroe County sandstone	0.44 ± 0.08	0.22 ± 0.04	0.5	Maruvanchery and Kim (2019)[32]	
Kunming sandstone	0.51 ± 0.01	0.29 ± 0.01	0.57	Zhou et al. (2018)[57]	here, saturated corresponds to a water content of 3.5%
Fontainebleau sandstone	1.60 ± 0.30	1.45 ± 0.24	0.91	This study	
Bentheim sandstone	0.55 ± 0.04	0.52 ± 0.05	0.95		
Adamswiller sandstone	0.49 ± 0.03	0.33 ± 0.02	0.67		
Rothbach sandstone	0.81 ± 0.01	0.65 ± 0.01	0.80		
Darley Dale sandstone	0.79 ± 0.01	0.51 ± 0.02	0.65		

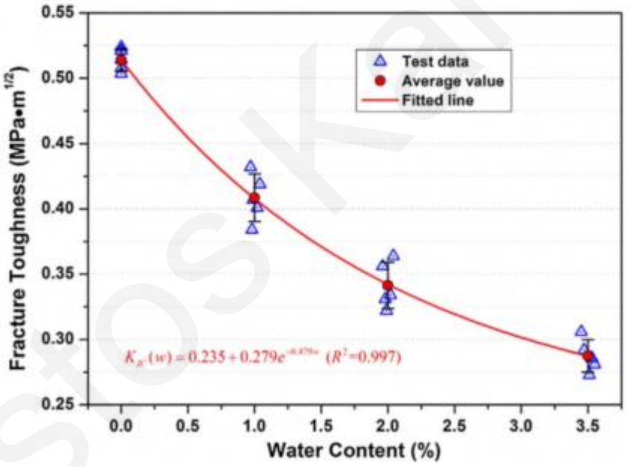


Figure 67 - Water content vs fracture toughness (from Z. Zhou et al. (2018))

However, cracks into the rock may propagate at intensity factor values below the critical ones (Hadizadeht & Law, 1991). This phenomenon is known as “subcritical cracking”, and it is mostly attributed on the hydrolysis of quartz (Atkinson & Meredith, 1981; Dyke & Dobereiner, 1991; Eppes & Keanini, 2017; Kean Atkinson, 1982). Quartz consists of one part silicon and two parts oxygen (SiO_2).

Figure 68 presents the relative mechanism. As shown, tensile stress concentration on crack tip is large. At time 1, molecules of quartz “stretch”, so bonds between silica (Si) and oxygen (O) weaken. At time 2, since Si-O-Si bonds are weaker, quartz molecules become chemically reactive with water molecules. Equation 3 expresses this reaction (Cai et al., 2019):

$$(Si\text{-}O\text{-}Si) + (H\text{-}O\text{-}H) \leftrightarrow \text{-}Si\text{-}OH\text{-}HO\text{-}Si\text{-} \qquad (4)$$

At time 3, the weak hydrogen bonds (OH-HO), which replace the stronger Si-O-Si bonds at the crack tip, are broken by subcritical stress, leading to the propagation of crack, even below stress values corresponding to the critical intensity factor. (Molaei & Kemeny, 2021), used a reactive molecular dynamic simulation to simulate the stress corrosion phenomenon on a "perfect" rock specimen consisting entirely of quartz. Their study confirmed the above proposed mechanism. Note that, hydrolysis of quartz can take place when water is either in liquid form or in vapour (Eppes & Keanini, 2017; Kean Atkinson, 1982), so relative humidity is enough to initial this mechanism.

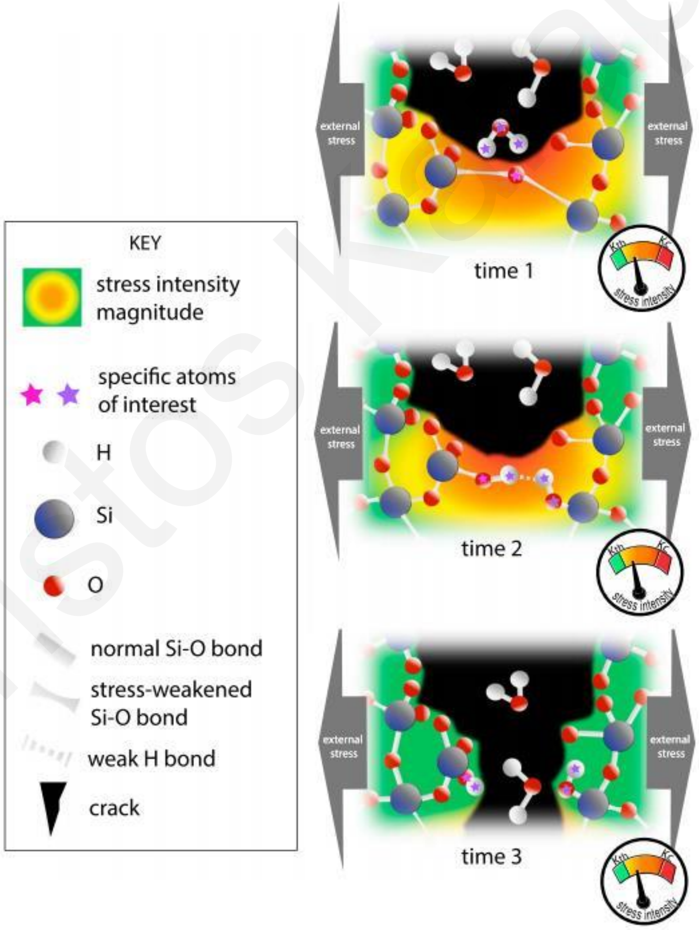


Figure 68 - Hydrolysis of quartz. K_{th} refers to a value under which no crack propagates, and it usually equals to 10-20% of the critical intensity factor (from Eppes & Keanini (2017)).

The weakening of rock strength due to quartz hydrolysis, seems to be important for both rock groups investigated, especially for sandstones where quartz is usually the dominant mineral. However, its influence should be limited in the cases where the wet samples suffer from intra-granular cracks instead of inter-granular ones (see 4.1), since hydrolysis affects the quartz grain itself, and not the space between grains.

Appendix B Calcite

Calcite ($CaCO_3$) is a common mineral, which, even in weak acidic environments, is soluble. Thus, calcite can react with water and dissolve. The relation governing this chemical reaction is described below in Equation 4 (Cai et al., 2019):

$$CaCO_3 + CO_2 + H_2O \rightarrow 2HCO^-_3 + Ca^{2+} \qquad (5)$$

Generally, calcite dissolution with water is a slow process. Ciantia et al. (2015) performed tests on calcarenites (consisting of 97% calcite) and reported that, in order to dissolve 2.9% of rock mass in water, the time needed was approximately one year, while in acidic liquid, no more than an hour was needed. Thus, according to authors, calcite dissolution should be connected with long-term phenomena, rather than short-term ones. However, Morrow et al. (2000) found that friction coefficient of calcite reduced by approximately 12% after a short-term saturation.

After part of calcite content dissolves, the structure of a rock is affected in many ways. Grains mass in the rock body is reduced in the presence of water, since part of it dissolves (Ciantia, Castellanza, & di Prisco, 2015; Gutierrez et al., 2000). Furthermore, if dissolution occurs at grain contact areas or at asperity contacts in fractures, friction is reduced, resulting in a general reduction in rock strength (Gutierrez et al., 2000). Figure 69 represents the effect of long-term dissolution of calcite in a calcarenite.

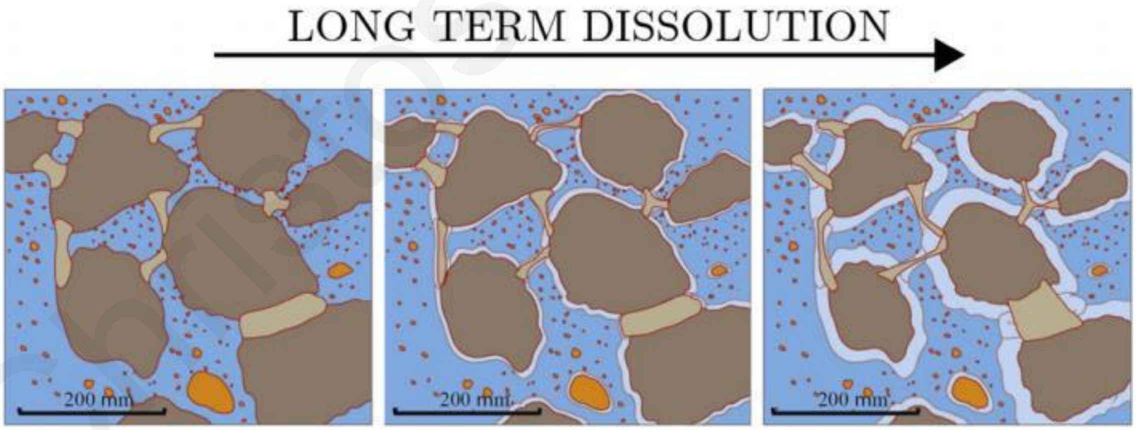


Figure 69 - Long term dissolution of calcarenite (from Ciantia, Castellanza, Crosta, et al. (2015))

The microstructure of rocks may also change in the presence of water. Changes in the surface of rock grains can lead to changes in their surface properties, such as the ability to adsorb water or other active agents (Gutierrez et al., 2000), which in turn, can lead to an indirect reduction in rock strength (for instance due to larger water content). Gutierrez et al. (2000) found that chalk samples (calcite content 98-99%) present lower values of friction angle when saturated with water than with oil. This supports the idea that calcite dissolution, affects the microstructure of the rock, leading to direct strength reduction.

Another result of calcite dissolution, which can be easily observed, is the increase in the porosity and permeability of a rock. Increase in porosity has even been observed in coals (Figure 70) where calcite is found in small quantities (Ahamed et al., 2019), and since porosity of a rock is strongly related with its strength, an increase in the porosity will certainly lead to a decrease in rock strength. Furthermore, as also explained in 3.1, pore expansion will give access to water in previously “close” pores, providing new areas for chemical reactions to occur.

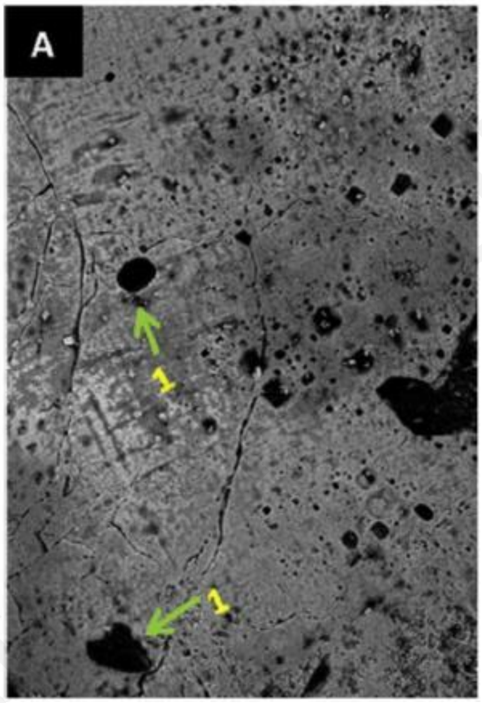


Figure 70 - SEM image of coal after saturation, showing (1) "holes" induced by the calcite dissolution (from Ahamed et al. (2019) quoting Zhang et al., 2018)

In every rock, the effect of calcite reactivity is relevant not only on the percentage of it, but also regarding the role calcite has in the structure of rock. In rocks where calcite is a cementation material between grains, its dissolution will greatly affect the wet UCS, since friction resistance and cohesion will be reduced. So, even in small portions, calcite content is something worth noted when investigating the UCS reduction. Figure 71 illustrates how the calcitic cementation dissolution affects coal UCS, but this effect can apply in other rocks where calcitic cementation can be found, such as sandstones and limestones. Indeed, Lebedev et al. (2014) examined the water weakening effect in Savonnieres limestone with calcitic cementing material, during saturation. According to their findings, the weakening of the cementing material due to its dissolution had a big impact on the decrease in strength of this limestone after saturation.

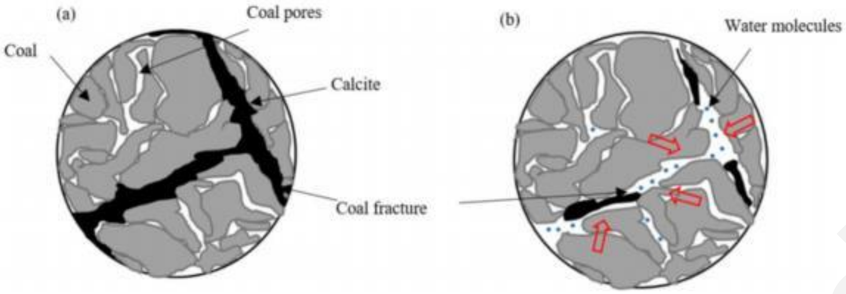


Figure 71 - Calcite dissolution in coal fractures. (a) Calcite as cementation between coal grains and (b) cementation after calcite dissolution (from Ahamed et al. (2019))

Appendix C Water distribution in cylindrical rock sample

Figure 72 shows real images of a cylindrical sandstone sample during saturation and drying process (Z. Zhou et al., 2016). As it can be seen, during saturation, the outer parts of the sample are firstly saturated and the inner follows, while during drying, the outer parts dry faster than the inner parts. So, it can be stated safely that, in a cylindrical rock sample immersed in water, when saturation degree is less than 100%, water is not equally distributed from the surface to the core.

Instead, during saturation, parts of the rock sample near the surface are fully saturated, parts near the core are dry, and in between, parts are partially saturated, with their saturation degree varying from 99%, near the surface, to 1% near the core, as it is illustrated in Figure 73 (D. Li & Wang, 2019). During drying, the opposite is valid; the core of the sample is fully saturated, while the parts near surface are dry and the saturation degree varies from 99% to 1% from the core to the surface. During saturation and drying, in the partially saturated parts, smaller pores are filled with water faster and lose their water slower than the bigger ones. Thus, in these parts, the unsaturated pores are expected to be the bigger ones.

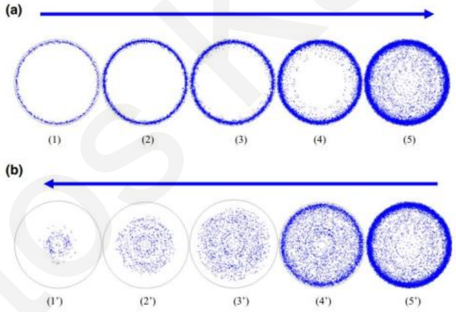


Figure 72 - Water distribution in cylindrical sandstone sample during (a) saturation and (b) drying (from Z. Zhou et al. (2016))

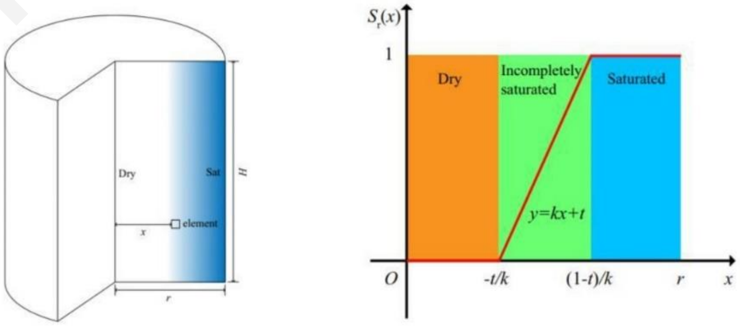


Figure 73 - Water distribution in a rock sample. Parts of the sample near surface are saturated, parts near the core are dry and parts in between are partially saturated. k stand for the variation of saturating degree per unit length and t is a parameter about the position of incompletely saturated elements (from D. Li & Wang (2019))

Appendix D Pore water pressure

Throughout the years, many relations from different researchers have been proposed, in order to estimate effective stress in porous media (i.e. the stress acting on the solid skeleton of a material), especially in soils and rocks (Guerriero & Mazzoli, 2021). No matter which relation best fits the nature of the examined material, all these relations agree that pore water pressure (from now on, “pore pressure”) plays an irreplaceable role.

Therefore, variation in pore pressure is a factor affecting the strength characteristics and the behaviour of a rock during loading. Firstly, an increase in pore pressure decreases the effective stress in the rock structure, leading to a decrease in shear stress. Furthermore, an increase in water pressure, leads to a rise on the total tensile stress acting on fracture tips (Cai et al., 2019; Zhong et al., 2019), thus contributing to the propagation of cracks (Figure 74). In addition, local stresses could corrode the internal structure of a rock (Figure 75). So, when does the pore pressure increase?

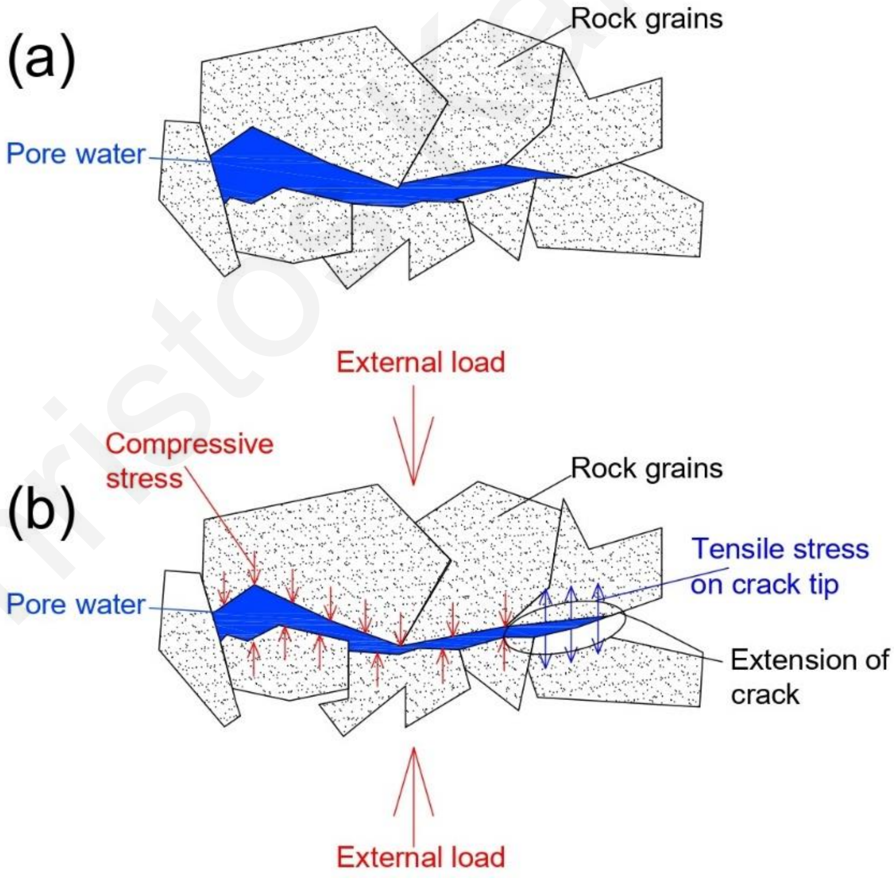


Figure 74 - (a) Pore water in crack and (b) pore water pressure increases due to compression, acting on crack tip helping the propagation of cracks

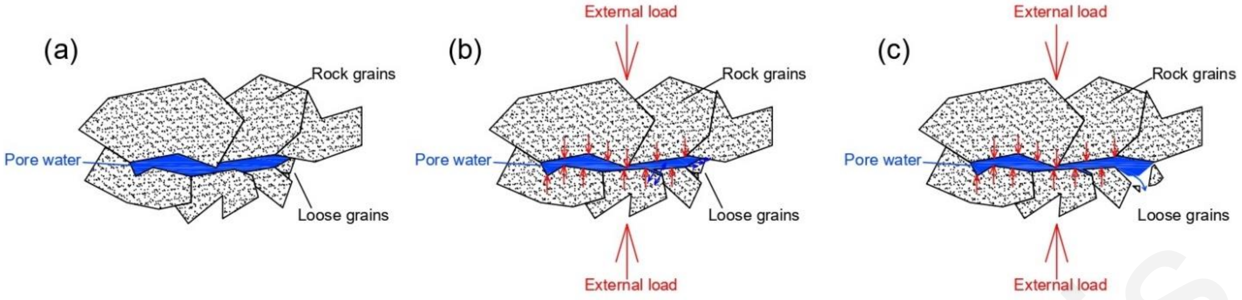


Figure 75 - (a) Pore water in crack, (b) increase of pore water pressure due to compression and (c) breakage of loose grains due to pore pressure

The major condition for pore pressure to increase inside the rock body, is for it to be undrained. In undrained conditions, the loading rate is fast enough according to the time the water needs in order to flow through the pores of the rock; thus, water pressure increases. In drained conditions, such mechanism does not occur, since water flow through the voids of the rock is fast, and pore pressure remains constant. Whether the conditions are drained or undrained, is a matter of the loading rate and of the characteristics of the rock sample. In Nicolas et al. (2016), a proposed way to check if an experiment is performed in drained or undrained conditions is given, estimating the time water needs to flow to a free surface, and comparing it with loading time. Additionally, Duda & Renner (2013) proposed another way for such a check, calculating the critical strain rate ("boundary" between drained and undrained conditions), and comparing it with the strain rate of the test. Note that, the presence of clay minerals in the rock matrix can have a reducing effect on rock permeability (Aksu et al., 2015; Duda & Renner, 2013), and it should be taken into account.

Pore pressure variation greatly depends on the stress-strain behaviour of a rock, as well. During the first two stages of loading (closure of cracks and elastic behaviour), rock samples exhibit a decrease in volume. This decrease is mostly happening due to the decrease in voids. Increase in pore pressure is caused by a reduction in pore volume (Duda & Renner, 2013). During the fourth stage (fast propagation stage), the sample exhibits dilation (i.e. the volume of the sample increases), new cracks form and connect with existing ones (Wasantha & Ranjith, 2014; Zhang et al., 2017). During this stage, pore pressure decreases (Duda & Renner, 2013; Hawkins & Mcconnell, 1992) and can even reach negative values (Wasantha & Ranjith, 2014). Observations from (Anagnostou et al., 2004) in a weak rock, after monitoring the pore pressure, confirm the above claims.

Hawkins & Mcconnell (1992) stated that for pore pressure to increase, the saturation degree must be close to, or equal to, 100%. This statement has also been supported by Zhang et al. (2017), while explaining the results of axial stiffness between samples of siltstones with different saturation degrees (Figure 76). The aforementioned authors observed that, as the saturation degree on samples increased, axial stiffness in the first stage (crack closure stage) of the strain curve exhibited lower values. However, fully saturated samples did not follow the trend and showed greater axial stiffness, almost equal to those with 10% saturation degree. The authors claimed that, since axial deformation comes mostly from large pores and fractures, this rapid increase in axial stiffness at fully saturated samples is caused by the extra stiffness provided by pore pressure. According to authors, the fact that only fully saturated samples showed such an increase in axial stiffness, testifies that for bigger pores to get fully filled with water, the saturation degree must be 100%.

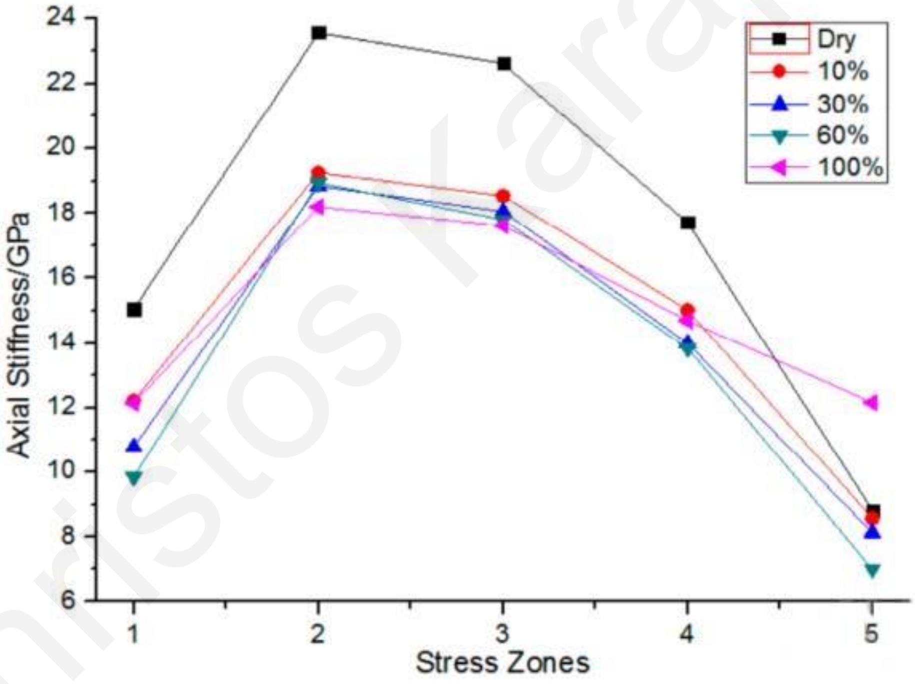


Figure 76 - Axial stiffness of a siltstone vs stress zone, at different saturation degrees (from Zhang et al. (2017))

However, such interpretations do not take under consideration the water distribution of water in a sample, and does not seem accurate. According to Appendix C, parts of the samples can be fully saturated at saturation degree way lower than 100%. Thus, pore pressure in the sample can increase even at lower saturation degrees. Of course, its influence in rock strength will be proportional to saturation degree, since the quantity of wet parts, depend on the saturation degree.

On the other hand, the increase in pore pressure can have a positive effect in wet UCS. In some of the sandstones tested by Hawkins & Mcconnell (1992), the UCS at 100% saturation showed a slight increase, compared to samples with a lower saturation degree (Figure 77). This increase happens in the fourth stage of strain curve, where dilatancy occurs. As noted, during this stage the pore pressure decreases and effective stress increases, resulting in a UCS slight increase.

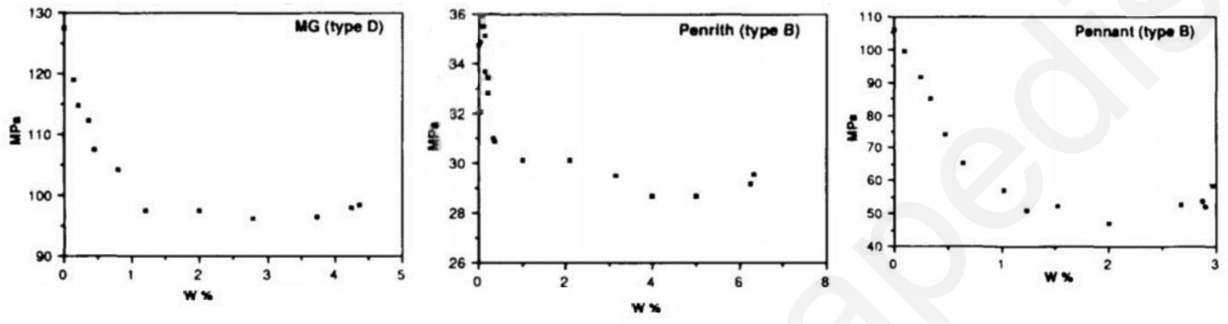


Figure 77 - Increase in UCS at higher water content in sandstones (from Hawkins & Mcconnell (1992))

What is more, (Brace & Martin, 1968), performed tests in confined conditions in different strain rates, at low porosity rocks. The samples were placed in water, with no membrane or other material separating external and pore water. The confining pressure was achieved by increasing the external water pressure. They observed, that rock samples under high strain rates showed higher compressive strength than those under lower strain rates. Their explanation was that during dilatancy (when failure happens), water into the pores of the samples (internal) at low strain rates connected with external water which produced the confining pressure, resulting in the equilibrium of internal and external water pressures, and thus, effective stress decreased. In the case of high strain rates, such an equilibrium could not be achieved, resulting in pore pressure decrease due to dilatancy, and thus, effective strength increased. However, the trend for samples to exhibit greater wet UCS in higher strain rate, has been observed by ZHOU et al. (2016) as well, in samples not immersed in water. They attributed this behavior mostly on capillary and viscosity related phenomena, rather than the increase in pore pressure.

Appendix E Capillary tension

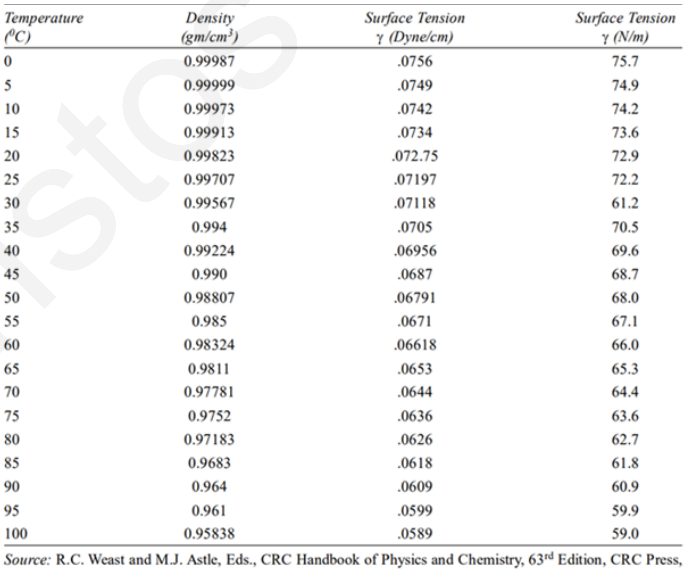
Capillary (from the Latin word *capillus*, meaning “hair”) is the phenomenon of water rise in pores until a specific height, when water comes in contact with a porous solid, such as a rock. This phenomenon contributes to the fact that, in nature, rock masses present saturation degrees between 0 and 1 above the free-water level.

During the known experiment with capillary tube, water rises inside the tube until a certain height, depending on the radius of the tube; the bigger the radius, the smaller the height of rise and vice versa. Following this experiment, capillary pressure Pc (i.e. the difference between air pressure, or any other non-wetting fluid like oil, and the pressure of water on the inter-face of the two fluids) is given by Equation 5:

$$P_c = \frac{2 \cdot \gamma_{wa} \cdot \cos\theta}{r} \qquad (6)$$

where γ_{wa} is the surface tension between water and air (or any other non-wetting liquid), θ is the contact angle on the point where the two fluids and the solid meet, and r is the radius of the tube. Table 8 presents the values of surface tension between water and air, for different temperatures.

Table 8 - Density and surface tension of water against air at different temperatures (from Srivastava et al. (2012), quoting from Weast and Astle)



Temperature (⁰C)	*Density (gm/cm³)*	*Surface Tension γ (Dyne/cm)*	*Surface Tension γ (N/m)*
0	0.99987	.0756	75.7
5	0.99999	.0749	74.9
10	0.99973	.0742	74.2
15	0.99913	.0734	73.6
20	0.99823	.072.75	72.9
25	0.99707	.07197	72.2
30	0.99567	.07118	61.2
35	0.994	.0705	70.5
40	0.99224	.06956	69.6
45	0.990	.0687	68.7
50	0.98807	.06791	68.0
55	0.985	.0671	67.1
60	0.98324	.06618	66.0
65	0.9811	.0653	65.3
70	0.97781	.0644	64.4
75	0.9752	.0636	63.6
80	0.97183	.0626	62.7
85	0.9683	.0618	61.8
90	0.964	.0609	60.9
95	0.961	.0599	59.9
100	0.95838	.0589	59.0

Source: R.C. Weast and M.J. Astle, Eds., CRC Handbook of Physics and Chemistry, 63rd Edition, CRC Press,

From Equation 5, it can be seen that P_c depends not only on the radius of the tube, but also on other material properties. If we assume that a rock consists of numerous tubes, we could suggest that the capillary pressure in a rock depends on the same parameters (Alyafei, 2021).

In soil and rock mechanics, it is generally accepted that under certain conditions, the capillary pressure between particles can increase the total cohesion of the soil or rock, as grains are pressed between each other. Han et al. (2002) modelled the capillary pressure between grains in four different cases: (i) unequal grain size, (ii) equal grain size, (iii) squeezed grains, and (iv) detached grains. The last two cases are very common in rock mechanics. Their findings showed that for the case of water, the capillary pressure between grains depends on the saturation degree (Figure 78 and Figure 79), the contact angle (Figure 78 a), the particle size (Figure 78 b), the fabric structure (Figure 78 c-d) and the loading type (compressive or tensile). Their claim, that capillary pressure is greater in grains of the same size (Figure 79), seems controversial. It is known from soil mechanics, that a soil (or any other granular material) consisting of equal size grains, presents higher pore size than one consisting of equally distributed grain sizes. If we assume the pore of a soil to correspond to the tube in the aforementioned experiment, and the pore size to correspond the radius of the tube, we could conclude that the higher the pore size, the lower the capillary pressure (Equation 5).

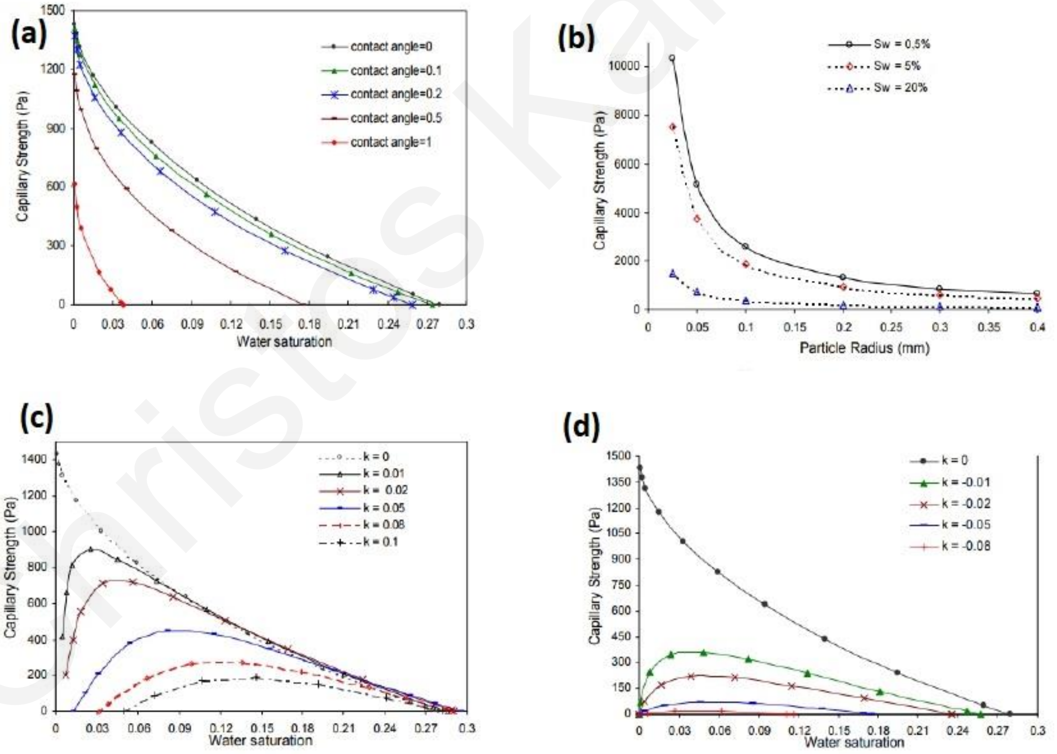


Figure 78 - Capillary strength vs (a) water saturation for different contact angles, (b) particle radius for different saturation degrees, and water saturation for different center-to-center distances in (c) detached particles and (d) squeezed particles. Note that k in (c) and (d) stands for particles distance, where k=0 for tangential attached particles) (from Han et al. (2002))

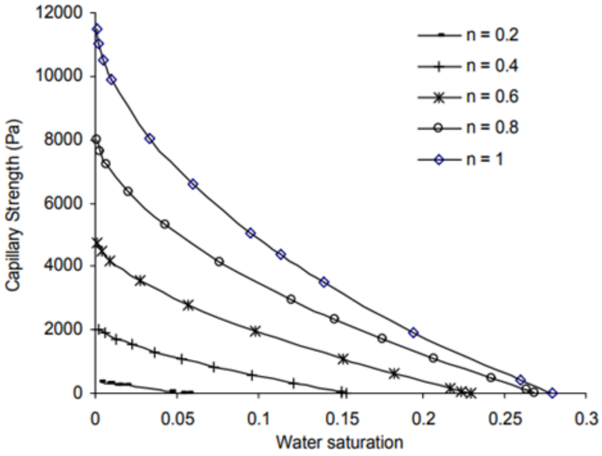


Figure 79 - Capillary strength between particles of different sizes vs water saturation. Note that, n stands for the ratio between the size of the two particles, where n=1 for particles of the same size (from Han et al. (2002))

Figure 80, presents a typical capillary pressure-water saturation curve of a rock. The y and x axis values may vary depending on each rock properties, as explained before, but the trend is similar in every rock. At lower saturation degrees, capillary pressure is highest, meaning that capillary bridges have formed in every possible point into the rock. Of course, capillary bridges cannot form when saturation degree is very close to - or equal to - zero, because the water content at this point is not enough to form such bridges. After the formation of capillary bridges, increase in saturation degree means that water in every capillary bridge is more, and thus, the contact angle between water and grains increases leading in the reduction of overall capillary pressure. At some degree of saturation, water takes over most of the available pore space, and capillary bridges vanish, resulting in the nullification of capillary pressure. At this point, strength induced by capillarity is eliminated. The curve follows the aforementioned trend, during both drying and wetting. However, for the same saturation degree, capillary pressure is higher during drying compared to wetting. Following that, it is expected that for rocks in which the capillary pressure has a significant impact on strength, the elimination of capillary bridges due to saturation, will have a negative effect.

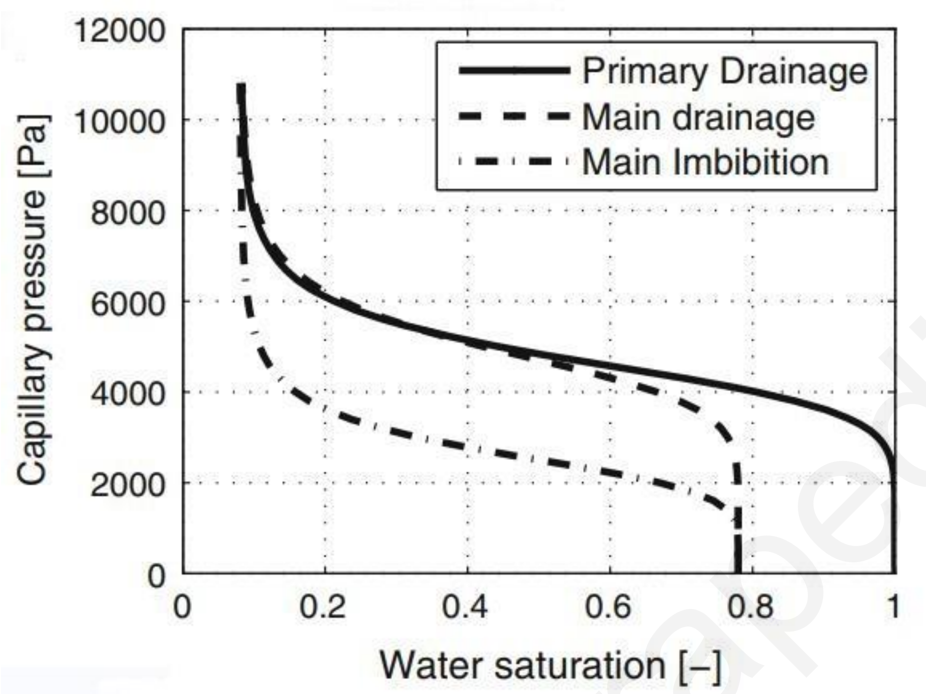


Figure 80 - Typical capillary pressure versus saturation degree curve (from Bottero et al. (2011))

An example of such rock, are chalks (Gutierrez et al., 2000; Taibi et al., 2009). It is believed that, without capillary pressure, chalks would exhibit very low strength, almost like dry sand (Gutierrez et al., 2000 quoting from Schroeder and Shao, 1996). Note that chalks are very difficult to get completely dry, so even in a theoretical degree of saturation of 0%, capillary bridges still exist. Taibi et al. (2009) examined two different chalks, and estimated that the maximum capillary cohesion was 69% and 44% of the total cohesion of the samples. Figure 81 presents the relation between suction and saturation degree for two Chalks (Taibi et al., 2009).

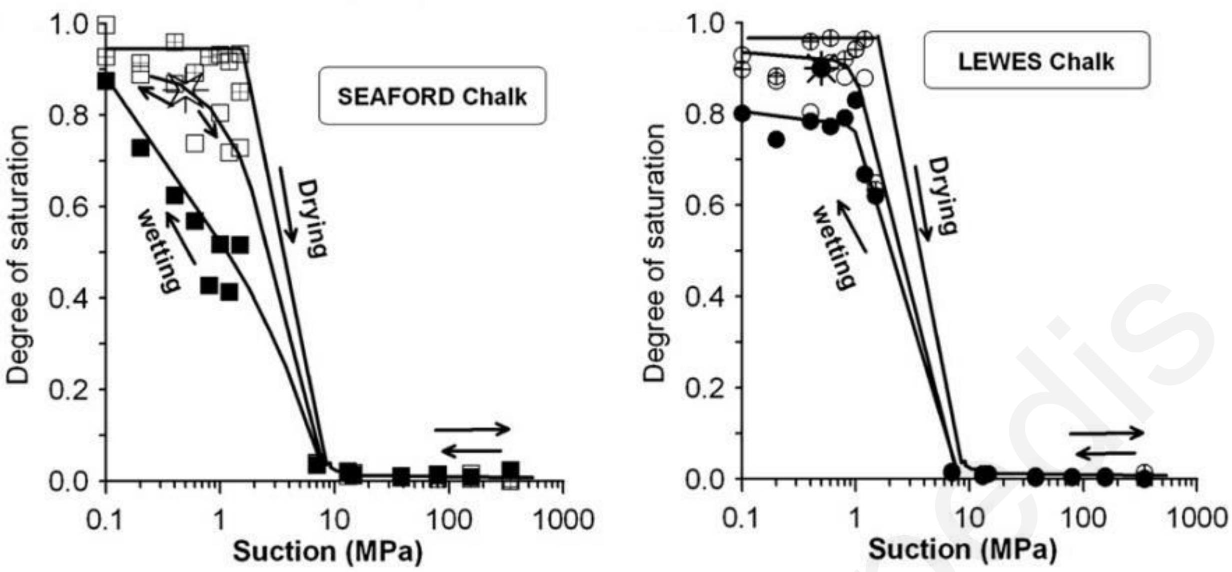


Figure 81 - Suctions vs saturation degree for two chalks (from Taibi et al. (2009))

Figure 82, presents the relation between suction and water content on some marl specimens, as found in Vergara & Triantafyllidis (2016). What is more, observing Figure 40 from the same article, one could conclude that for water content values higher than 3.5%, the UCS and suction of marl show a good relation. So, for such water content values, the UCS of marl seem to have a good dependance on capillary pressure.

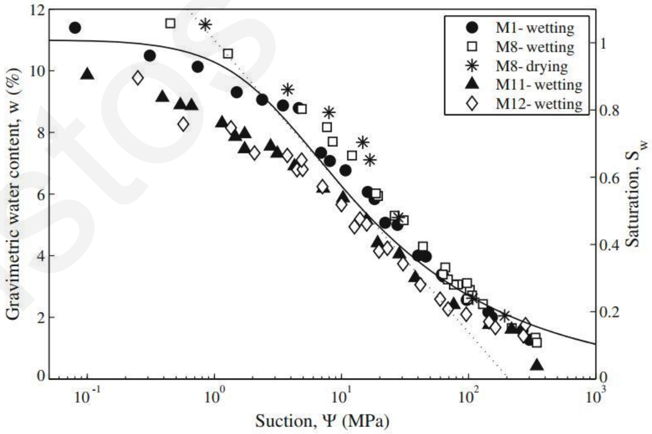
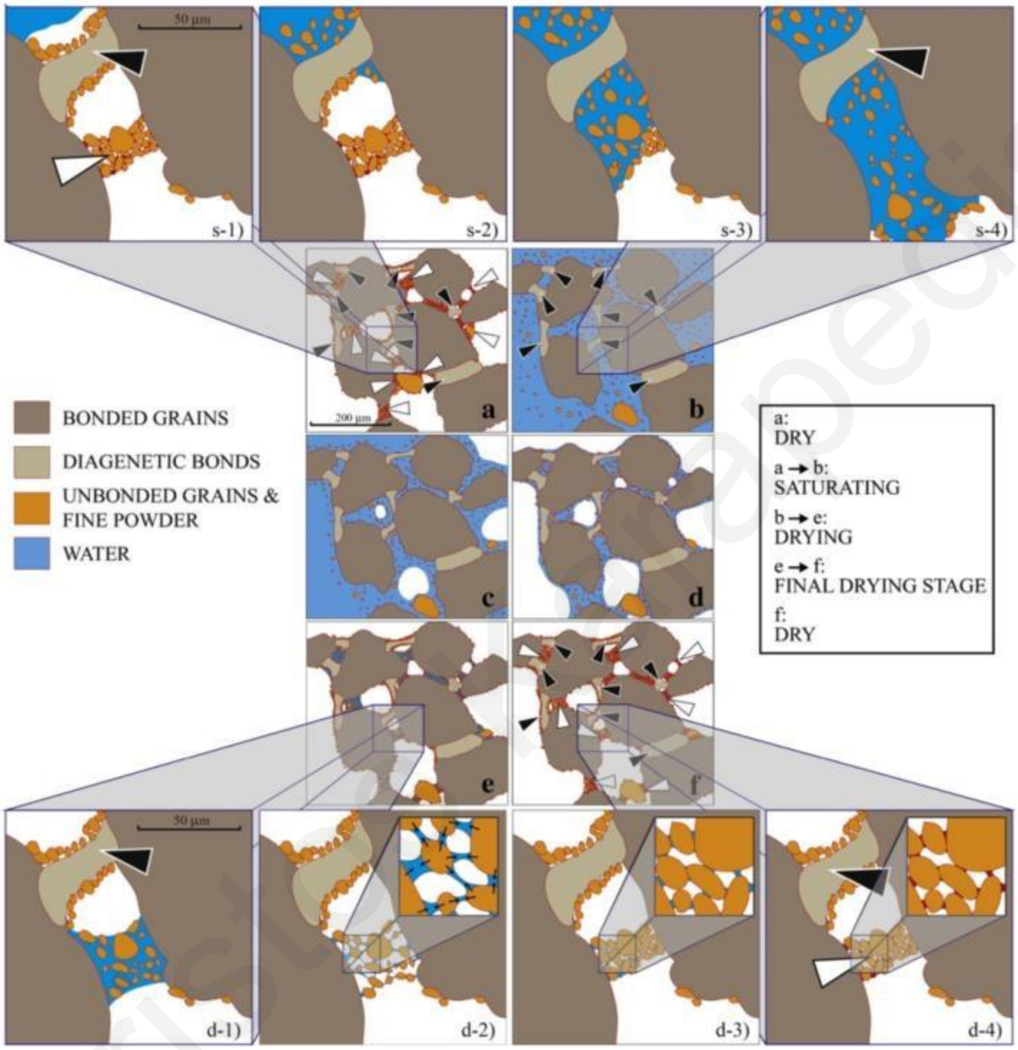


Figure 82 - Suction vs water content on marl specimens (from Vergara & Triantafyllidis (2016))

What is more, calcarenite strength is supposed to depend also from capillary phenomena. As Ciantia et al. (2015) report, calcarenites owe their strength on two different bond types; permanent (diagenetic) and temporary caused by the cohesion between unbonded grains and powder. Temporary bonds are suspended when calcarenites get saturated, as water flow scatters the unbonded grains into the pores. During drying, however, the formation of capillary bridges causes such bonds to form again, and so, calcarenite temporary strength component returns. Figure 83 is relevant (Ciantia, Castellanza, Crosta, et al., 2015). Explanation of the Figure was kept as in the referred article, for the better understanding of it.



Schematic representation of the short-term bonding–debonding process: at the initial dry state (a) the structure is held together by both diagenetic (PB, black arrows) and depositional bonds (TB, white arrows). As water fills the pores (a–b) the depositional bonds fall into suspension. This is caused by the hydro-mechanical effect of non-equilibrated tension in the depositional structures at the capillary front (s1–s4). During the drying phase (b–e), macro water menisci form between the grains. As evaporation proceeds, micro water menisci form between the smaller powder grains, and due to the capillary effect the suspended powder is pulled together until the last stages of drying (d1–d4) when, eventually, the dissolved crystals precipitate and a depositional bond is formed.

Figure 83 - Elimination and re-creation of temporary strength component in calcarenites (from Ciantia, Castellanza, Crosta, et al. (2015)). For the explanation of the representation, see in the Figure.

Bibliography

Ahamed, M. A. A., Perera, M. S. A., Matthai, S. K., Ranjith, P. G., & Dong-yin, L. (2019). Coal composition and structural variation with rank and its influence on the coal-moisture interactions under coal seam temperature conditions – A review article. In *Journal of Petroleum Science and Engineering* (Vol. 180, pp. 901–917). Elsevier B.V. https://doi.org/10.1016/j.petrol.2019.06.007

Aksu, I., Bazilevskaya, E., & Karpyn, Z. T. (2015). Swelling of clay minerals in unconsolidated porous media and its impact on permeability. *GeoResJ*, *7*, 1–13. https://doi.org/10.1016/j.grj.2015.02.003

Al Ani, T., Sarapaa, O. (2008). *Clay and clay mineralogy*. https://www.researchgate.net/publication/292706105

Alcázar-Vara, L. A., & Cortés-Monroy, I. R. (2018). Drilling Fluids for Deepwater Fields: An Overview. In *Recent Insights in Petroleum Science and Engineering*. InTech. https://doi.org/10.5772/intechopen.70093

Alyafei, N. (2021). *Fundamentals of Reservoir Rock Properties Experimental Studies of Multiphase Flow in Porous Media Using In-Situ Imaging View project Development of Virtual Field Trips Using the Augmented and Virtual Reality Tools to Enhance the Student Learning Experience in PETE Courses View project*. https://www.researchgate.net/publication/335866693

Anagnostou, G., Zurich, E., Vogelhuber, M., Anagnostou, G., & Kovári, K. (2004). *The influence of pore water pressure on the mechanical behavior of squeezing rock Analysis of large deformation problems in tunnelling View project Face stability View project The influence of pore water pressure on the mechanical behavior of squeezing rock*. https://www.researchgate.net/publication/237736309

Anderson, R. L., Ratcliffe, I., Greenwell, H. C., Williams, P. A., Cliffe, S., & Coveney, P. V. (2010). Clay swelling - A challenge in the oilfield. In *Earth-Science Reviews* (Vol. 98, Issues 3–4, pp. 201–216). https://doi.org/10.1016/j.earscirev.2009.11.003

Anne Perera, M. S., Suresh Madushan Sampath, K. H., Ranjith, P. G., & Rathnaweera, T. D. (2018). Effects of pore fluid chemistry and saturation degree on the fracability of Australian warwick siltstone. *Energies*, *11*(10). https://doi.org/10.3390/en11102795

Atkinson, B. K., & Meredith, P. G. (1981). Stress corrosion cracking of quartz: A note on the influence of chemical environment. *Tectonophysics*, *77*(1–2). https://doi.org/10.1016/0040-1951(81)90157-8

Brace, W. F., & Martin, R. J. (1968). A TEST OF THE LAW OF EFFECTIVE STRESS FOR CRYSTALLINE ROCKS OF LOW POROSITY. In *Rock Mech. Min. $ci* (Vol. 5).

Babets, D. V., Kovrov, O. S., Moldabayev, S. K., Tereschuk, R. M., & Sosna, D. O. (2020). IMPAcT of wATeR SATURATION effecT on SeDIMeNTARY ROcKS STReNGTH PROPeRTIeS. *Naukovyi Visnyk Natsionalnoho Hirnychoho Universytetu*, *2020*(4), 76–81. https://doi.org/10.33271/nvngu/2020-4/076

Barton, C. D., & Karathanasis, A. D. (2002). *CLAY MINERALS*.

Behnsen, J., & Faulkner, D. R. (2012). The effect of mineralogy and effective normal stress on frictional strength of sheet silicates. *Journal of Structural Geology*, *42*, 49–61. https://doi.org/10.1016/j.jsg.2012.06.015

Behnsen, J., & Faulkner, D. R. (2013). Permeability and frictional strength of cation-exchanged montmorillonite. *Journal of Geophysical Research: Solid Earth*, *118*(6), 2788–2798. https://doi.org/10.1002/jgrb.50226

Bird, P. (1984). *HYDRATION-PHASE DIAGRAMS AND FRICTION OF MONTMORILLONITE UNDER LABORATORY AND GEOLOGIC CONDITIONS, WITH IMPLICATIONS FOR SHALE COMPACTION, SLOPE STABILITY, AND STRENGTH OF FAULT GOUGE* (Vol. 107).

Bottero, S., Hassanizadeh, S. M., & Kleingeld, P. J. (2011). From Local Measurements to an Upscaled Capillary Pressure-Saturation Curve. *Transport in Porous Media*, *88*(2), 271–291. https://doi.org/10.1007/s11242-011-9739-4

Burshtein, L. S. (1969). *EFFECT OF MOISTURE ON THE STRENGTH AND DEFORMABILITY OF SANDSTONE*.

Cai, X., Zhou, Z., Liu, K., Du, X., & Zang, H. (2019). Water-weakening effects on the mechanical behavior of different rock types: Phenomena and mechanisms. *Applied Sciences (Switzerland)*, *9*(20). https://doi.org/10.3390/app9204450

Chen, F., Sun, X., & Lu, H. (2022). Influence of Water Content on the Mechanical Characteristics of Mudstone with High Smectite Content. *Geofluids*, *2022*.

https://doi.org/10.1155/2022/9855213

Chen, H., Geng, J., Zhang, Z., Jiang, R., Zhai, J., & Zhang, J. (2021). The Structure and Properties of Sepiolite with Partial Lattice Ions Substituted by Aluminum Ions. *Frontiers in Chemistry*, *9*. https://doi.org/10.3389/fchem.2021.721225

Chen, X., Yi, H., Gao, L., Shi, X., & Liu, Y. (2020). Effects of inhibitor KCl on hydration swelling and softening of a smectite-poor mudstone. *Journal of Petroleum Exploration and Production Technology*, *10*(7), 2685–2692. https://doi.org/10.1007/s13202-020-00948-8

Christidis, G. E. (2011). Industrial clays. *European Mineralogical Union Notes in Mineralogy*, *9*(1), 341–414. https://doi.org/10.1180/EMU-notes.9.9

Ciantia, M. O., Castellanza, R., Crosta, G. B., & Hueckel, T. (2015). Efects of mineral suspension and dissolution on strength and compressibility of soft carbonate rocks. *Engineering Geology*, *184*, 1–18. https://doi.org/10.1016/j.enggeo.2014.10.024

Ciantia, M. O., Castellanza, R., & di Prisco, C. (2015). Experimental Study on the Water-Induced Weakening of Calcarenites. *Rock Mechanics and Rock Engineering*, *48*(2), 441–461. https://doi.org/10.1007/s00603-014-0603-z

Doostmohammadi, R., Moosavi, M., Mutschler, T., & Osan, C. (2009). Influence of cyclic wetting and drying on swelling behavior of mudstone in south west of Iran. *Environmental Geology*, *58*(5), 999–1009. https://doi.org/10.1007/s00254-008-1579-3

Drits, V. A., Guggenheim, S., Zviagina, B., & Kogure, T. (2012). Structures of the 2:1 layers of pyrophyllite and talc. *Clays and Clay Minerals*, *60*(6), 574–587. https://doi.org/10.1346/CCMN.2012.0600603

Duda, M., & Renner, J. (2013). The weakening effect of water on the brittle failure strength of sandstone. *Geophysical Journal International*, *192*(3), 1091–1108. https://doi.org/10.1093/gji/ggs090

Dyke, C. G., & Dobereiner, & L. (1991). Evaluating the strength and deformability of sandstones. In *Quarterly Journal of Engineering Geology* (Vol. 24).

Eggleston, R. A., Bailey, S. W. (1967). STRUCTURAL ASPECTS OF DIOCTAHEDRAL CHLORITE. In *THE AMERICAN MINERALOGIST* (Vol. 52).

Elbrahmi, J., & Zoukaghe, M. (2016). Evolution of expansive soils structure with different solicitations and effect of some parameters on swelling properties – review article. *Theoretical & Applied Science*, *37*(05), 68–77. https://doi.org/10.15863/TAS.2016.05.37.14

Eppes, M. C., & Keanini, R. (2017). Mechanical weathering and rock erosion by climate-dependent subcritical cracking. *Reviews of Geophysics*, *55*(2), 470–508. https://doi.org/10.1002/2017RG000557

Erguler, Z. A., & Ulusay, R. (2009). Water-induced variations in mechanical properties of clay-bearing rocks. *International Journal of Rock Mechanics and Mining Sciences*, *46*(2), 355–370. https://doi.org/10.1016/j.ijrmms.2008.07.002

Firoozi, A. A., Firoozi, A. A., & Baghini, M. S. (2016). A Review of Clayey Soils. In *Asian Journal of Applied Sciences*. www.ajouronline.com

Ghasemi, M., & Sharifi, M. (2021). Effects of layer-charge distribution on swelling behavior of mixed-layer illite-montmorillonite clays: A molecular dynamics simulation study. *Journal of Molecular Liquids*, *335*. https://doi.org/10.1016/j.molliq.2021.116188

Guerriero, V., & Mazzoli, S. (2021). Theory of effective stress in soil and rock and implications for fracturing processes: A review. In *Geosciences (Switzerland)* (Vol. 11, Issue 3, pp. 1–35). MDPI AG. https://doi.org/10.3390/geosciences11030119

Guggenheim, S. (n.d.). *INTRODUCTION TO THE PROPERTIES OF CLAY MINERALS*.

Guggenheim, S., Martin, R. T. (1995). Definition of clay and clay mineral: Joint report of the AIPEA nomenclature and CMS nomenclature committees. In *Clays and Clay Minerals* (Vol. 43, Issue 2, pp. 255–256). Clay Minerals Society. https://doi.org/10.1346/CCMN.1995.0430213

Guo, P., Gu, J., Su, Y., Wang, J., & Ding, Z. (2021). Effect of cyclic wetting–drying on tensile mechanical behavior and microstructure of clay-bearing sandstone. *International Journal of Coal Science and Technology*, *8*(5), 956–968. https://doi.org/10.1007/s40789-020-00403-3

Gutierrez, M., Øino, L. E., & Høeg, K. (2000). The effect of fluid content on the mechanical behaviour of fractures in chalk. *Rock Mechanics and Rock Engineering*, *33*(2), 93–117. https://doi.org/10.1007/s006030050037

Hadizadeht, J., & Lawi, R. D. (1991). Water-weakening of Sandstone and Quartzite Deformed at Various Stress and Strain Rates. In *Lt. J. Rock Mech. Min. Sci. & Geomech. Abstr* (Vol. 28, Issue 5).

Han, G., Dusseault, M. B., & Cook, J. (2002). Quantifying Rock Capillary Strength Behavior in Unconsolidated Sandstones. *Proceedings of the SPE/ISRM Rock Mechanics in Petroleum Engineering Conference*, 171–180. https://doi.org/10.2118/78170-ms

Haq, A., Iqbal, Y., Khan, M. R., & Haq, A. (2008). HISTORICAL DEVELOPMENT IN THE CLASSIFICATION OF KAOLIN SUBGROUP. In *J Pak Mater Soc* (Vol. 2, Issue 1).

Hatch, C. D., Wiese, J. S., Crane, C. C., Harris, K. J., Kloss, H. G., & Baltrusaitis, J. (2012). Water adsorption on clay minerals as a function of relative humidity: Application of BET and Freundlich adsorption models. *Langmuir*, *28*(3), 1790–1803. https://doi.org/10.1021/la2042873

Hawkins, A. B., & Mcconnell, B. J. (1992). Sensitivity of sandstone strength and deformability to changes in moisture content. In *Quarterly Journal of Engineering Geology* (Vol. 25).

Hillier, S. (n.d.). Clay mineralogy. In *Sedimentology* (pp. 223–228). Springer Berlin Heidelberg. https://doi.org/10.1007/3-540-31079-7_47

Huang, S., Wang, J., Qiu, Z., & Kang, K. (2018). Effects of cyclic wetting-drying conditions on elastic modulus and compressive strength of sandstone and mudstone. *Processes*, *6*(12). https://doi.org/10.3390/pr6120234

Huang, S., Xia, K., Yan, F., & Feng, X. (2010). An experimental study of the rate dependence of tensile strength softening of longyou sandstone. *Rock Mechanics and Rock Engineering*, *43*(6), 677–683. https://doi.org/10.1007/s00603-010-0083-8

Huang, Y. H., Yang, S. Q., Hall, M. R., & Zhang, Y. C. (2018). The effects of NaCl concentration and confining pressure on mechanical and acoustic behaviors of brine-saturated sandstone. *Energies*, *11*(2). https://doi.org/10.3390/en11020385

Ikari, M. J., Saffer, D. M., & Marone, C. (2007). Effect of hydration state on the frictional properties of montmorillonite-based fault gouge. *Journal of Geophysical Research: Solid Earth*, *112*(6). https://doi.org/10.1029/2006JB004748

Jiang, Q., Cui, J., Feng, X., & Jiang, Y. (2014). Application of computerized tomographic scanning to the study of water-induced weakening of mudstone. *Bulletin of Engineering Geology and the Environment*, *73*(4), 1293–1301. https://doi.org/10.1007/s10064-014-0597-5

Jiménez-González, I., Rodríguez-Navarro, C., & Scherer, G. W. (2008). Role of clay minerals in the physicomechanical deterioration of sandstone. *Journal of Geophysical Research: Earth Surface*, *113*(2). https://doi.org/10.1029/2007JF000845

José Pejon, O., & Valentin Zuquette, L. (2002). *Analysis of cyclic swelling of mudrocks*. www.elsevier.com/locate/enggeo

Kawai, K., Sakuma, H., Katayama, I., & Tamura, K. (2015). Frictional characteristics of single and polycrystalline muscovite and influence of fluid chemistry. *Journal of Geophysical Research: Solid Earth*, *120*(9), 6209–6218. https://doi.org/10.1002/2015JB012286

Kean Atkinson, B. (1982). Subcriticai crack propagation in rocks: theory, experimental results and applications. In *Journal of Structural Geolo,03* (Vol. 4, Issue 1).

Kohno, M. (2020). Effects of hydraulic gradient and clay type on permeability of clay mineral materials. *Minerals*, *10*(12), 1–17. https://doi.org/10.3390/min10121064

Kohno, M. (2021). Swelling-pressure and hydraulic conductivity of compacted clays focusing on the clay-mineral type. *Materials Transactions*, *62*(8), 1203–1209. https://doi.org/10.2320/matertrans.Z-M2021833

Kohno, M., Nara, Y., Kato, M., & Nishimura, T. (2018). Effects of clay-mineral type and content on the hydraulic conductivity of bentonite–sand mixtures made of Kunigel bentonite from Japan. *Clay Minerals*, *53*(4), 721–732. https://doi.org/10.1180/clm.2018.52

Kumari, N., & Mohan, C. (n.d.). *Basics of Clay Minerals and Their Characteristic Properties*. www.intechopen.com

Lashkaripour, G. R., & Ajalloeian, R. (n.d.). *THE EFFECT OF WATER CONTENT ON THE MECHANICAL BEHAVIOUR OF FINE-GRAINED SEDIMENTARY ROCKS*.

Lebedev, M., Wilson, M. E. J., & Mikhaltsevitch, V. (2014). An experimental study of solid matrix weakening in water-saturated Savonnières limestone. *Geophysical Prospecting*, *62*(6), 1253–1265. https://doi.org/10.1111/1365-2478.12168

Li, B., Liu, J., Bian, K., Ai, F., Hu, X., Chen, M., & Liu, Z. (2019). Experimental study on the mechanical properties weakening mechanism of siltstone with different water content. *Arabian Journal of Geosciences*, *12*(21). https://doi.org/10.1007/s12517-019-4852-8

Li, D., & Wang, W. (2019). Quantitative analysis of the influence of saturation on rock strength reduction considering the distribution of water. *Geomechanics and Geophysics for Geo-*

Energy and Geo-Resources, *5*(2), 197–207. https://doi.org/10.1007/s40948-019-00106-3

Li, H., Shen, R., Li, D., Jia, H., Li, T., Chen, T., & Hou, Z. (2019). Acoustic emission multi-parameter analysis of dry and saturated sandstone with cracks under uniaxial compression. *Energies*, *12*(10). https://doi.org/10.3390/en12101959

Li, X., Peng, K., Peng, J., & Hou, D. (2021). Experimental investigation of cyclic wetting-drying effect on mechanical behavior of a medium-grained sandstone. *Engineering Geology*, *293*, 106335. https://doi.org/10.1016/j.enggeo.2021.106335

Lin, M. L., Jeng, F. S., Tsai, L. S., & Huang, T. H. (2005). Wetting weakening of tertiary sandstones - Microscopic mechanism. *Environmental Geology*, *48*(2), 265–275. https://doi.org/10.1007/s00254-005-1318-y

Liu, X., Jin, M., Li, D., & Zhang, L. (2018). Strength deterioration of a Shaly sandstone under dry–wet cycles: a case study from the Three Gorges Reservoir in China. *Bulletin of Engineering Geology and the Environment*, *77*(4), 1607–1621. https://doi.org/10.1007/s10064-017-1107-3

Liu, X., Liu, Q., Huang, S., Liu, B., & Liu, J. (2021). Effects of cyclic wetting-drying on the mechanical behavior and improved damage model for sandstone. *Marine Georesources and Geotechnology*, *39*(10), 1244–1254. https://doi.org/10.1080/1064119X.2020.1823535

Lu, Y., Wang, L., Sun, X., & Wang, J. (2017). Experimental study of the influence of water and temperature on the mechanical behavior of mudstone and sandstone. *Bulletin of Engineering Geology and the Environment*, *76*(2), 645–660. https://doi.org/10.1007/s10064-016-0851-0

Machida, S., Katsumata, K. I., & Yasumori, A. (2021). Effects of kaolinite layer expansion and impurities on the solid-state reaction of kaolinite. *RSC Advances*, *11*(61), 38473–38477. https://doi.org/10.1039/d1ra07762g

Madsen, F. T., & Muller-Vonmoos, M. (1989). The Swelling Behaviour of Clays. In *Applied Clay Science* (Vol. 4).

Masoumi, H., Horne, J., & Timms, W. (2017). Establishing Empirical Relationships for the Effects of Water Content on the Mechanical Behavior of Gosford Sandstone. *Rock Mechanics and Rock Engineering*, *50*(8), 2235–2242. https://doi.org/10.1007/s00603-017-1243-x

Mellini, M. (1981). Crystal structure of lizardite 1T. *Acta Crystallographica Section A Foundations of Crystallography*, *37*(a1), C189–C189. https://doi.org/10.1107/s0108767381093902

Min, Y., Kim, D., & Jun, Y. S. (2018). Effects of Na+ and K+ Exchange in Interlayers on Biotite Dissolution under High-Temperature and High-CO2-Pressure Conditions. *Environmental Science and Technology*, *52*(22), 13638–13646. https://doi.org/10.1021/acs.est.8b04623

Molaei, F., Kemeny, J. (2021). *Investigation of Stress Corrosion Cracking in rocks, a reactive molecular dynamic simulation Development of a Digital Twin for San Xavier Mine View project 2D material hybrid View project*. https://www.researchgate.net/publication/352569662

Moore, D. E., Lockner, D. A. (2004). Crystallographic controls on the frictional behavior of dry and water-saturated sheet structure minerals. *Journal of Geophysical Research*, *109*(B3). https://doi.org/10.1029/2003jb002582

Moore, D. E., & Lockner, D. A. (2016). 11. Friction of the Smectite Clay Montmorillonite. In *The Seismogenic Zone of Subduction Thrust Faults* (pp. 317–345). Columbia University Press. https://doi.org/10.7312/dixo13866-011

Morodome, S., & Kawamura, K. (2009). Swelling behavior of Na- and Ca-montmorillonite up to 150°c by in situ x-ray diffraction experiments. *Clays and Clay Minerals*, *57*(2), 150–160. https://doi.org/10.1346/CCMN.2009.0570202

Morodome, S., & Kawamura, K. (2011). In situ X-ray diffraction study of the swelling of montmorillonite as affected by exchangeable cations and temperature. *Clays and Clay Minerals*, *59*(2), 165–175. https://doi.org/10.1346/CCMN.2011.0590205

Morrow, C. A., Moore, D. E., & Lockner, D. A. (2000). *The effect of mineral bond strength and adsorbed water on fault gouge frictional strength* (Vol. 27, Issue 6).

Morrow, C. A., Moore, D. E., & Lockner, D. A. (2017). Frictional strength of wet and dry montmorillonite. *Journal of Geophysical Research: Solid Earth*, *122*(5), 3392–3409. https://doi.org/10.1002/2016JB013658

Nasvi, M. C. M., Ranjith, P. G., Sanjayan, J., Haque, A., & Li, X. (2014). Mechanical behaviour of wellbore materials saturated in brine water with different salinity levels. *Energy*, *66*, 239–249. https://doi.org/10.1016/j.energy.2013.12.003

Nicolas, A., Fortin, J., Regnet, J. B., Dimanov, A., & Gúeguen, Y. (2016). Brittle and semi-brittle behaviours of a carbonate rock: Influence of water and temperature. *Geophysical Journal International*, *206*(1), 438–456. https://doi.org/10.1093/gji/ggw154

Noël, C., Baud, P., & Violay, M. (2021). Effect of water on sandstone's fracture toughness and frictional parameters: Brittle strength constraints. *International Journal of Rock Mechanics and Mining Sciences*, *147*. https://doi.org/10.1016/j.ijrmms.2021.104916

Obert, L., Windes, S. L., Duvall, W. I. (1946). *Standarized tests for determining the physical properties of mine rocks.* United States Department of the Interion - Bureau of Mines https://books.google.com.cy/books?hl=el&lr=&id=KGA6DzFWcBkC&oi=fnd&pg=PP4&dq=obert+et+al+1946&ots=V2Hot-wob7&sig=nvRZh1fOikZevv4xtxP97mohTC8&redir_esc=y#v=onepage&q&f=false

Park, W. C. (1979). Cation Exchange Capacities, Swelling, and Solubility of Clay Minerals in Acidic Solutions: A Literature Review. https://koreascience.kr/article/JAKO197923034628894.pdf

Post, J. E., Bish, D. L., & Heaney, P. J. (2007). Synchrotron powder X-ray diffraction study of the structure and dehydration behavior of sepiolite. *American Mineralogist*, *92*(1), 91–97. https://doi.org/10.2138/am.2007.2134

Rao, S. M., Thyagaraj, T., & Raghuveer Rao, P. (2013). Crystalline and Osmotic Swelling of an Expansive Clay Inundated with Sodium Chloride Solutions. *Geotechnical and Geological Engineering*, *31*(4), 1399–1404. https://doi.org/10.1007/s10706-013-9629-3

Rathnaweera, T. D., Ranjith, P. G., & Perera, M. S. A. (2014). Salinity-dependent strength and stress-strain characteristics of reservoir rocks in deep saline aquifers: An experimental study. *Fuel*, *122*, 1–11. https://doi.org/10.1016/j.fuel.2013.11.033

Rathnaweera, T. D., Ranjith, P. G., Perera, M. S. A., Lashin, A., & Al Arifi, N. (2015). Non-linear stress-strain behaviour of reservoir rock under brine saturation: An experimental study. *Measurement: Journal of the International Measurement Confederation*, *71*, 56–72. https://doi.org/10.1016/j.measurement.2015.04.011

Sakuma, H., & Kawamura, K. (2009). Structure and dynamics of water on muscovite mica surfaces. *Geochimica et Cosmochimica Acta*, *73*(14), 4100–4110. https://doi.org/10.1016/j.gca.2009.05.029

Sakuma, H., Lockner, D. A., Solum, J., & Davatzes, N. C. (2022). Friction in clay-bearing faults increases with the ionic radius of interlayer cations. *Communications Earth and Environment*, *3*(1). https://doi.org/10.1038/s43247-022-00444-3

Sakuma, H., & Suehara, S. (2015). Interlayer bonding energy of layered minerals: Implication for the relationship with friction coefficient. *Journal of Geophysical Research: Solid Earth*, *120*(4), 2212–2219. https://doi.org/10.1002/2015JB011900

Sato, T., Watanabe, T., & Otsuka, R. (1992). Effects of layer charge, charge location, and energy change on expansion properties of dioctahedral smectites. *Clays and Clay Minerals*, *40*(1), 103–113. https://doi.org/10.1346/CCMN.1992.0400111

Shakoor, A., & Barefield, E. (2006). *The effect of degree of saturation on the unconfined compressive strength of selected sandstones*. https://www.researchgate.net/publication/268008400

Shukla, R., Ranjith, P. G., Choi, S. K., Haque, A., Yellishetty, M., & Hong, L. (2013). Mechanical behaviour of reservoir rock under brine saturation. *Rock Mechanics and Rock Engineering*, *46*(1), 83–93. https://doi.org/10.1007/s00603-012-0246-x

Srivastava, A. K., Prajapati, M. K., & Raha, S. (2012). A COMPARATIVE ESTIMATION OF SURFACE TENSION FOR DIFFERENT PRECIPITATED LIQUID. *IJP*, *5*(1), 11–14. https://doi.org/10.13140/RG.2.1.2634.3441

Taibi, S., Duperret, A., & Fleureau, J. M. (2009). The effect of suction on the hydro-mechanical behaviour of chalk rocks. *Engineering Geology*, *106*(1–2), 40–50. https://doi.org/10.1016/j.enggeo.2009.02.012

Tang, S. (2018). The effects of water on the strength of black sandstone in a brittle regime. *Engineering Geology*, *239*, 167–178. https://doi.org/10.1016/j.enggeo.2018.03.025

Teich-McGoldrick, S. L., Greathouse, J. A., Jové-Colón, C. F., & Cygan, R. T. (2015). Swelling Properties of Montmorillonite and Beidellite Clay Minerals from Molecular Simulation: Comparison of Temperature, Interlayer Cation, and Charge Location Effects. *Journal of Physical Chemistry C*, *119*(36), 20880–20891. https://doi.org/10.1021/acs.jpcc.5b03253

Tetsuka, H., Katayama, I., Sakuma, H., & Tamura, K. (2018). Effects of humidity and interlayer cations on the frictional strength of montmorillonite. *Earth, Planets and Space*, *70*(1). https://doi.org/10.1186/s40623-018-0829-1

Theo Kloprogge, J., & Ponce, C. P. (2021). Spectroscopic studies of synthetic and natural saponites: A review. In *Minerals* (Vol. 11, Issue 2, pp. 1–28). MDPI AG. https://doi.org/10.3390/min11020112

Tiennot, M., Mertz, J. D., & Bourgès, A. (2019). Influence of Clay Minerals Nature on the Hydromechanical and Fracture Behaviour of Stones. *Rock Mechanics and Rock Engineering*, *52*(6), 1599–1611. https://doi.org/10.1007/s00603-018-1672-1

Tu, H. (2015). *PREDICTION OF THE VARIATION OF SWELLING PRESSURE AND 1-D HEAVE OF EXPANSIVE SOILS WITH RESPECT TO SUCTION*. https://www.researchgate.net/publication/285311039

Uddin, F. (2018). Montmorillonite: An Introduction to Properties and Utilization. In *Current Topics in the Utilization of Clay in Industrial and Medical Applications*. InTech. https://doi.org/10.5772/intechopen.77987

Valaskova, M., Martynkova, G. S. (2012). Vermiculite: Structural Properties and Examples of the Use. In Clay Minerals in Nature - Their Characterization, Modification and Application. InTech. https://doi.org/10.5772/51237

Van Eeckhout, E. M. (1976). The Mechanisms of Strength Reduction due to Moisture in Coal Mine Shales. In *Int. J. Rock Mech. Min. Sci. & Geomech. Abstr* (Vol. 13). Pergamon Press.

Vergara, M. R., & Triantafyllidis, T. (2016). Influence of Water Content on the Mechanical Properties of an Argillaceous Swelling Rock. *Rock Mechanics and Rock Engineering*, *49*(7), 2555–2568. https://doi.org/10.1007/s00603-016-0938-8

Verstrynge, E., Adriaens, R., Elsen, J., & Van Balen, K. (2014). Multi-scale analysis on the influence of moisture on the mechanical behavior of ferruginous sandstone. *Construction and Building Materials*, *54*, 78–90. https://doi.org/10.1016/j.conbuildmat.2013.12.024

Wanniarachchi, A., Gamage, R. P., Lyu, Q., Perera, S., Wickramarathne, H., & Rathnaweera, T. (2019). Mechanical characterization of low permeable siltstone under different reservoir saturation conditions: An experimental study. *Energies*, *12*(1). https://doi.org/10.3390/en12010014

Wasantha, P. L. P., & Ranjith, P. G. (2014). Water-weakening behavior of Hawkesbury sandstone in brittle regime. *Engineering Geology*, *178*, 91–101. https://doi.org/10.1016/j.enggeo.2014.05.015

Wasantha, P. L. P., Ranjith, P. G., Permata, G., & Bing, D. (2018). Damage evolution and deformation behaviour of dry and saturated sandstones: Insights gleaned from optical measurements. *Measurement: Journal of the International Measurement Confederation*, *130*, 8–17. https://doi.org/10.1016/j.measurement.2018.07.075

Wong, L. N. Y., Maruvanchery, V., & Liu, G. (2016). Water effects on rock strength and stiffness degradation. In *Acta Geotechnica* (Vol. 11, Issue 4, pp. 713–737). Springer Verlag. https://doi.org/10.1007/s11440-015-0407-7

Xie, K., Jiang, D., Sun, Z., Chen, J., Zhang, W., & Jiang, X. (2018). NMR, MRI and AE Statistical Study of Damage due to a Low Number of Wetting–Drying Cycles in Sandstone from the Three Gorges Reservoir Area. In *Rock Mechanics and Rock Engineering* (Vol. 51, Issue 11, pp. 3625–3634). Springer. https://doi.org/10.1007/s00603-018-1562-6

Yao, W., Li, C., Zhan, H., Zhou, J. Q., Criss, R. E., Xiong, S., & Jiang, X. (2020). Multiscale Study of Physical and Mechanical Properties of Sandstone in Three Gorges Reservoir Region Subjected to Cyclic Wetting–Drying of Yangtze River Water. *Rock Mechanics and Rock Engineering*, *53*(5), 2215–2231. https://doi.org/10.1007/s00603-019-02037-7

Yotsuji, K., Tachi, Y., Sakuma, H., & Kawamura, K. (2021). Effect of interlayer cations on montmorillonite swelling: Comparison between molecular dynamic simulations and experiments. *Applied Clay Science*, *204*. https://doi.org/10.1016/j.clay.2021.106034

Zanazzi, P. F., Montagnoli, M., Nazzareni, S., & Comodi, P. (2006). Structural effects of pressure on triclinic chlorite: A single-crystal study. *American Mineralogist*, *91*(11–12), 1871–1878. https://doi.org/10.2138/am.2006.2191

Zhang, D., Gamage, R. P., Perera, M. S. A., Zhang, C., & Wanniarachchi, W. A. M. (2017). Influence of Water Saturation on the Mechanical Behaviour of Low-Permeability Reservoir Rocks. *Energies*, *10*(2). https://doi.org/10.3390/en10020236

Zhao, B., Li, Y., Huang, W., Yang, J., Sun, J., Li, W., Zhang, L., & Zhang, L. (2021). Mechanical characteristics of red sandstone under cyclic wetting and drying. *Environmental Earth Sciences*, *80*(22). https://doi.org/10.1007/s12665-021-10067-0

Zhong, C., Zhang, Z., Ranjith, P. G., Lu, Y., & Choi, X. (2019). The role of pore water plays in coal under uniaxial cyclic loading. *Engineering Geology*, *257*. https://doi.org/10.1016/j.enggeo.2019.05.002

Zhou, M., Li, J., Luo, Z., Sun, J., Xu, F., Jiang, Q., & Deng, H. (2021). Impact of water–rock interaction on the pore structures of red-bed soft rock. *Scientific Reports*, *11*(1). https://doi.org/10.1038/s41598-021-86815-w

Zhou, Z., Cai, X., Cao, W., Li, X., & Xiong, C. (2016). Influence of Water Content on Mechanical Properties of Rock in Both Saturation and Drying Processes. *Rock Mechanics and Rock Engineering*, *49*(8), 3009–3025. https://doi.org/10.1007/s00603-016-0987-z

Zhou, Z., Cai, X., Ma, D., Chen, L., Wang, S., & Tan, L. (2018). Dynamic tensile properties of sandstone subjected to wetting and drying cycles. *Construction and Building Materials*, *182*, 215–232. https://doi.org/10.1016/j.conbuildmat.2018.06.056

ZHOU, Z. long, CAI, X., ZHAO, Y., CHEN, L., XIONG, C., & LI, X. bing. (2016). Strength characteristics of dry and saturated rock at different strain rates. *Transactions of Nonferrous Metals Society of China (English Edition)*, *26*(7), 1919–1925. https://doi.org/10.1016/S1003-6326(16)64314-5