



Influence of calcium leaching on the mechanical behavior of a rock–mortar interface: A DEM analysis

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ABSTRACT

Being a potential preferential way for water to flow, interfaces between host rock and engineered barriers are critical in the design of deep radioactive waste repositories. In case of cementitious materials, presence of water may lead to long term degradation by leaching. Such a phenomenon could impede the integrity of the confinement by its effect on the hydro-mechanical properties of the interface. Recent experimental results from Buzzi et al. [8] have evidenced some effects of leaching on the hydro-mechanical behavior of rock–concrete interfaces for one leaching time. This paper intends to investigate the influence of leaching on the mechanical behavior of rock–mortar interfaces by means of numerical simulations. These latter will be run for several leaching times to produce a better understanding of the phenomenon. For this purpose, a DEM approach has been developed to simulate the increase of the macro-porosity resulting from the leaching process. The implementation of the approach is first discussed. Then direct shear tests under constant normal stress are performed on a simple interface geometry and on a natural interface geometry. The results after Buzzi et al. [8] are corroborated by this research.

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1. Introduction

Many countries are currently facing the issue of nuclear waste storage and one possible option is underground repositories. Over the past 20 years, comprehensive studies have been undertaken to assess the feasibility of such a solution and to better understand the materials involved in the design of both engineered and natural barriers. These includes different types of rock, concrete or compacted bentonite mixtures or pellets. Not only the fundamental properties of these materials are of prime importance but also their possible interactions [13,16,34] and the issue of interfaces [11,7]. Unlike usual civil engineering structures, the service life of nuclear repositories is counted in thousands of years [37] and consequently, many studies are devoted to long term behavior of materials with an emphasis on aging or degradation. One possible degradation phenomenon is calcium leaching, which generally affects cementitious materials in presence of water. Natural leaching is a very long process and very few data are available in the literature about natural degradation of concrete [33]. In order to obtain data in an acceptable time frame, accelerated degradation protocols have been established. Leaching kinetics can be accelerated

either by use of ammonium nitrate [10] or by electrical potential [32]. Even though the acceleration factor is significant (up to 300 with ammonium nitrate [1]), the tests are still long lasting. During the process of leaching, several chemical components of the cement paste such as portlandite are dissolved, locally augmenting the macro-porosity [10]. As a result, the degraded material experiences a loss of mechanical strength and a reduction of stiffness [23,14,10,18]. Unlike for the mechanical properties, the effect of leaching on the transfer properties of leached cementitious materials (e.g. permeability) have not been widely investigated. Actually, they are usually correlated to the porosity by means of empirical formulas [27,25]; the general trend being that an increase of porosity produces an increase of permeability.

So far, most of the research on leached cementitious materials have been focused on the properties of the bulk material. In fact, little data are available on the impact of leaching on interfaces despite their importance for the efficiency of the nuclear waste repository. A recent experimental study by Buzzi et al. [8] has shown that a leached rock–concrete interface exhibits a lower shear strength than an intact interface. Also, and against expectations, the leached interfaces did not seem to conduct more water than the intact ones. The study by Buzzi et al. [8] was performed for a fixed leaching depth. The study presented in this paper was undertaken to provide a better understanding of the mechanical behavior of leached interfaces for various leaching depths. The Discrete

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Element Method code PFC3D from Itasca [31] has been used since DEM offers the possibility to capture the increase of macro-porosity resulting from leaching and the breakage of contact asperities, two key phenomena of the problem. The method to implement the leaching has firstly been validated by means of numerical unconfined compression tests. Then, two interface morphologies have been sheared under different levels of constant normal stress and for different leaching depths. The first interface is a simple saw tooth geometry in order to eliminate complications due to a complex geometry. Then, a natural morphology has been tested to confirm the results obtained on the saw tooth interface. The results tend to corroborate the conclusions obtained by Buzzi et al. [8] and to provide new insight into the effect of leaching on the mechanical behavior of interfaces. The existence of a transitional leaching depth, at which the joint exhibits neither dilation nor contraction has been evidenced. Most of the mechanical strength is lost before this leaching depth and the joint becomes fully contractant passed this same leaching depth. The implications for water flow within the interface are also briefly discussed. The preliminary results of this study were published in Lambert et al. [22] and they are partly reproduced in this paper to offer the reader a complete view of the problem.

2. Intact materials

This numerical study consists in three series of tests i.e. unconfined compression tests on intact and leached mortar, direct shear tests on intact and leached saw tooth interfaces and direct shear tests on intact and leached “natural” interfaces. The materials considered in these simulations are a mortar and a granite, the mechanical and chemical (if relevant) properties of which are given by Bernard et al. [4] and Le Bellego et al. [23] for the mortar and by Grasselli [15] for the granite.

The calibration of the contact laws and properties between particles was achieved following the procedure described in Itasca [20] in order to obtain the macro properties (unconfined compressive strength and Young’s modulus) given by Bernard et al. [4], Le Bellego et al. [23] and Grasselli [15]. The result of the calibration and the intact properties of the different materials are given in Tables 1 and 2. Note that for the mortar, two kinds of particles have been used i.e. cement and sand. The proportions of cement and sand were calculated knowing the volumetric air fraction of 3% in the mortar [4], its composition (1380 kg/m³ of sand) and assuming $G_s = 2.6$ for the sand. It can then be deduced that 45% of the particles are cement and 55% are sand. These particles are randomly distributed in the specimen. From a numerical point of view, sand and cement particles have the same density but different stiffness. As suggested by Bernard et al. [4], the stiffness of the sand–sand contact is approximately 3.5 times greater than the stiffness of

Table 1
Material parameters for intact materials.

Description	Cement	Sand	Granite
Particle properties			
Particle contact modulus (GPa)	18.0	63.0	56.1
Ratio of particle normal to shear stiffness (kn/ks)	2.5	2.5	2.5
Particle friction coefficient	0.364	0.364	0.7
Parallel-bond properties			
Parallel-bond radius multiplier	1.0	1.0	1.0
Parallel-bond modulus (GPa)	18.0	63.0	56.1
Ratio of parallel-bond normal to shear stiffness	2.5	2.5	2.5
Mean value of bond normal strength (MPa)	69.5	69.5	191
Standard deviation of bond normal strength (MPa)	5	5	19.1
Mean value of bond shear strength (MPa)	69.5	69.5	19.1
Standard deviation of bond shear strength (MPa)	5	5	19.1

Table 2
Results of the calibration for intact mortar and granite.

Properties	Mortar		Granite	
	Exp.	PFC	Exp.	PFC
UCS (MPa)	68.1	62.3	48.44	48.6
E (GPa)	44.1	44.5	172.53	170.8

the cement–cement contact. An equivalent stiffness is automatically calculated by PFC3D for the cement–sand contact. The bond strength of the sand–sand contact and cement–cement contact are the same for a matter of simplicity.

3. Implementation of leaching

Several studies on cementitious materials have shown that leaching of cement paste leads to an increase of macro-porosity [10,18,5], which has to be reproduced to simulate leaching. The main components dissolved during the leaching process are the Calcium Silicate Hydrates (CSH), the Calcium Hydroxides (CH) and the Hydrated Aluminates (denoted HA herein). It has also been evidenced that leaching is a multiple front process [9]. Bernard et al. [4] have actually identified four leached zones, each one being delimited by two successive fronts and characterized by different amounts of CSH, CH and HA. The position of each front has been observed to be a function of the square root of leaching time ($x_{dij} = \alpha_{ij} \times \sqrt{t}$). In the scope of this paper the positions of the various fronts are defined in relation to the overall leaching depth LD . The data after Bernard et al. [4] were fitted to obtain the position of each dissolution front and the amount of CSH in zones 3–5 as a function of the leaching depth LD [22]. The equations are given in Fig. 1, which is adapted from Bernard et al. [4]. For a matter of simplicity, the percentage of leached CSH is considered to be constant (average value) in zones 3 and 4.

Effect of leaching is introduced by randomly removing a given percentage of particles within the leached zones in accordance with the chemical profiles given in Fig. 1. The mechanical properties of the bonds between the particles are kept unchanged. The number of particles deleted is estimated from the proportion of chemical species dissolved at a given leaching depth (or leaching time). CSH, CH and HA are considered to have the same mechanical properties and size. No distinction is made between these three components. However, it is still required to quantify the volumet-

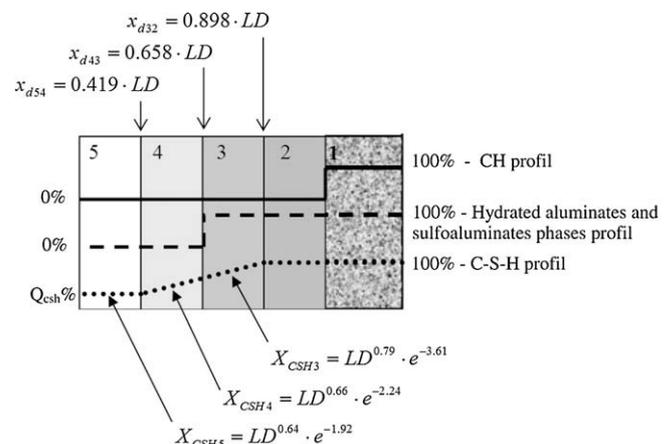


Fig. 1. Hydrated aluminates and sulfoaluminates phases profiles adapted from Bernard et al. [4] with expressions of the front positions (x_{dij} expressed in mm) and percentage of leached CSH in each zone as a function of leaching depth (LD expressed in mm).

ric fraction of CSH, CH and HA present in the mortar to properly implement the leaching. Since cement particles represent 45% in volume of the mortar (see Section 2), on the basis of experimental observation by Bernard et al. [4], the initial volumetric proportion of each chemical species (VPM^o) in the mortar can be estimated (see Table 3).

For each zone, at a given time, a proportion δ of the components is dissolved so that the percentage of particle to be removed (PPR) for each chemical species becomes:

$$PPR = \delta \cdot VPM^o \quad (1)$$

As all the particles have the same mechanical properties and no distinction is made between different components, the total percentage of cement particles randomly removed is given by:

$$PPR_{total} = \delta_{CH} \cdot VPM_{CH}^o + \delta_{CSH} \cdot VPM_{CSH}^o + \delta_{HA} \cdot VPM_{HA}^o \quad (2)$$

where the subscripts CH, CSH and HA refer to the corresponding chemical species.

The method to implement the leaching process has been validated via some unconfined compression tests on cylindrical specimens for simulated leaching times of 28, 56 and 98 days [22]. The position of the leaching fronts and the percentage of particles removed (see Table 4) have been estimated in accordance to experimental observations. Note that modeling the evolution of leaching depth in time is out of the scope of this paper. The numerical results, expressed in terms of decrease of Young's modulus and decrease of unconfined compressive strength with leaching time, have been compared to the experimental data in Table 5. A good agreement has been found for the deformation properties between experimental and numerical data. However, the strength reduction obtained numerically tends to underestimate the strength the leached material. The procedure developed in this paper reproduces the increase of macro-porosity by randomly deleting particles in the sample. Luping and Nilsson [24] showed that with the same porosity, samples with smaller pore size exhibits a greater strength. The use of a polydisperse particle size distribution and removal of the smallest particles would certainly improve model predictions and is worth further investigations.

4. Behavior of single saw tooth interface

4.1. Interface model

The mechanical response of rock joints is highly influenced by the morphology of the interface [3] so that a simple saw tooth interface has been chosen to start with. It consists of a single tooth 5 mm high and with an angle of 30° (Fig. 2). Using simple geometries

Table 3
 $VPCP^o$: initial volumetric proportion in cement paste. VPM^o : initial volumetric proportion in mortar.

	CH	CSH	HA
$VPCP^o$ (%) after Bernard et al. [4]	14.7	40.2	17.9
VPM^o (%) ($VPM^o = 0.45 \times VPCP^o$)	6.6	18.1	8.1

Table 4
Estimation of the percentage of particles removed in each leached zone to simulate the increase of macro-porosity and position of dissolution fronts for various leaching times.

Leaching duration	Percentage of particles removed				Position of fronts (mm)			
	Zone 2	Zone 3	Zone 4	Zone 5	x_{d21}	x_{d32}	x_{d43}	x_{d54}
28 days	6.6	9.4	22.9	25.6	8.9	8.0	5.9	3.7
56 days	6.6	10.2	24.9	28.3	12.6	11.3	8.3	5.3
98 days	6.6	11.1	27.1	31	16.7	15	11	7

Table 5

Validation of the implementation of leaching. (a) Evolution of ratio of Young's modulus over initial Young's modulus with respect to leaching time. (b) Evolution of ratio of unconfined compressive strength over initial unconfined compressive strength with respect to leaching time.

Leaching time (days)	0	28	56	98
E/E_0 (exp. after Bernard et al. [4])	1	0.77	0.64	0.47
E/E_0 (PFC)	1	0.67	0.55	0.43
UCS/UCS_0 (exp. after Le Bellego et al. [23])	1	0.76	0.63	0.51
UCS/UCS_0 (PFC)	1	0.64	0.51	0.34

tries is a common approach when testing rock joints and in fact, this very same geometry was used by Yang and Chiang [36]. Fig. 2 shows not only the contact morphology but also the expected water flow path and the progression of the leaching fronts with in the walls of the contact as a result of the water flow. As the study is about rock–mortar contacts, only the mortar wall is leached.

The rock–mortar specimen is built on 31,200 particles having a radius ranging from 0.4 mm (in the vicinity of the interface) to 1.6 mm. The interface is frictional (friction angle of 20°) and the recent “smooth joint model” (SJM) [26] was used to prevent the contact roughness from depending on the particle size. The specimen is firstly subjected to a compression along axis Y (Fig. 2) and then to a shearing along axis X at constant normal stress. Displacements along Z are restrained. The sum of contact forces on the periphery of the upper half are used to compute the normal stress and shear stress. In the following, the mechanical behavior of the leached contact is studied as a function of the leaching depth LD . The method described in Section 3 was used to determine the position of the leaching fronts and the amount of CSH left in zones 3–5 for various leaching depths.

4.2. Mechanical behavior of the intact interface

Numerical direct shear tests under constant normal stress ranging from 1.5 MPa to 6 MPa were performed on the intact interface. Here, the simulations aim to check that the well known qualitative response of rock joints upon shearing is reproduced. Fig. 3a and b show the evolutions of shear stress and of normal displacement with tangential displacement. It can be seen that the qualitative mechanical behavior of rock joints upon shearing is well captured [17,12]: the higher the normal stress, the higher the shear stress and the lower the dilation. Also, the peak in stress occurs in the first 2 mm of shearing. Experimentally, the dilation is preceded by a slight contraction usually due to the walls interlocking. This is not simulated here and, consequently, no contraction is observed before dilation. These numerical tests are useful to validate the qualitative behavior of the intact interface. Should any change take place for the leached interface, it will be attributed to the leaching process.

4.3. Mechanical behavior of the leached interface

Similarly, direct shear tests under constant normal stress were performed on the leached interface for various leaching depths.

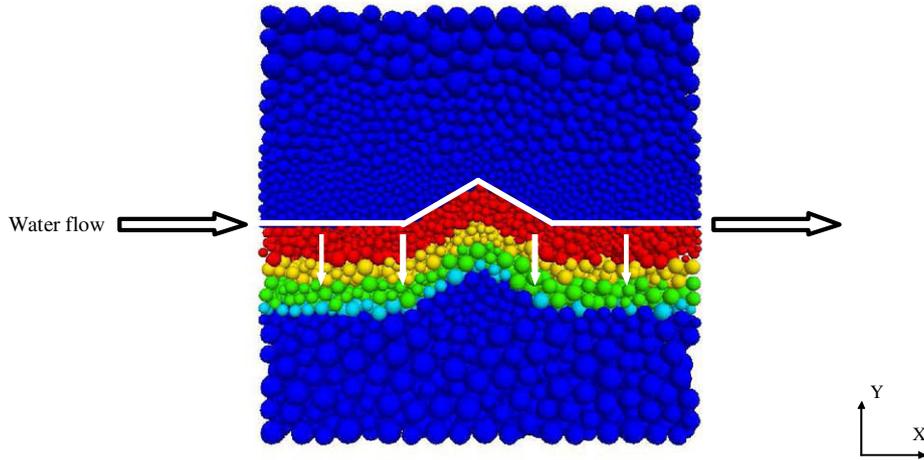


Fig. 2. View of the joint model using DEM code PFC. Dimensions of the model: 50 mm (X) × 50 mm (Y) × 40 mm (Z). White line represents the contact, the darkest material is sound, the lightest is leached.

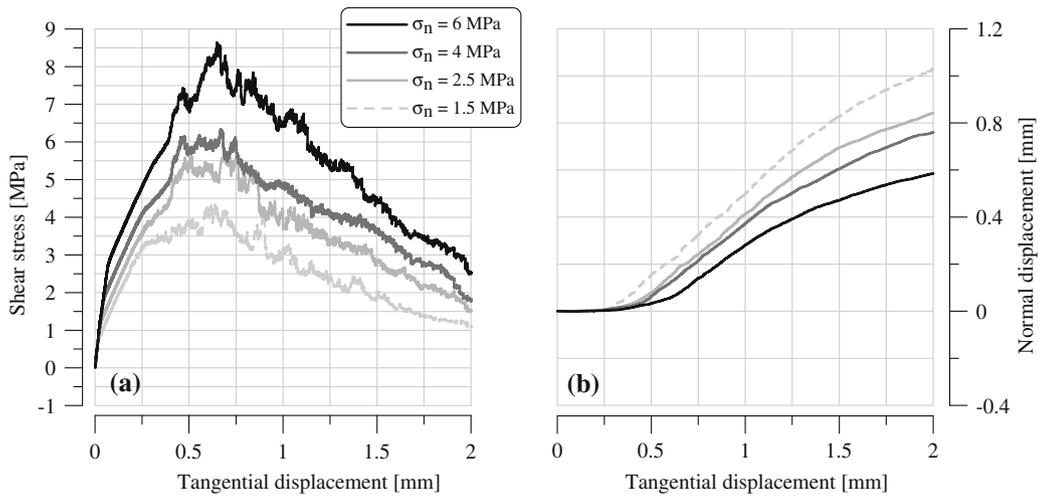


Fig. 3. Numerical results on intact interface. (a) Evolution of shear stress vs. tangential displacement. (b) Evolution of normal displacement vs. tangential displacement. Normal stress ranges from 1.5 MPa to 6 MPa.

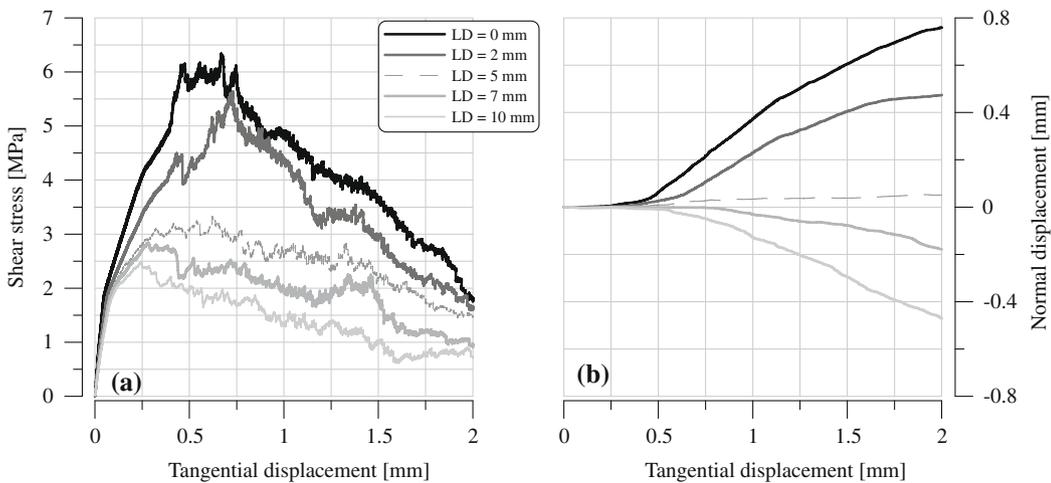


Fig. 4. Numerical shear tests at constant normal stress ($\sigma_n = 2.5$ MPa). Evolution of: (a) shear stress vs. tangential displacement and (b) normal displacement vs. tangential displacement for different leaching depths LD .

The evolutions of shear stress and of normal displacement with the tangential displacement for a normal stress of 4 MPa are shown in Fig. 4a and b. The reduction in shear strength as leaching progresses is very clear. Consistent with the results by Buzzi et al. [8], a change in behavior from brittle to ductile progressively takes place. The loss of mechanical strength of the interface is due to the weakening of the bulk material [10,23] and, in the framework of rock joints, could be seen as a weathering of the mortar wall [2]. Not only the shear strength is affected by leaching but also the joint dilation upon shearing. It is progressively reduced to a point where the joint becomes fully contractant, which is also consistent with the results obtained by Buzzi et al. [8] (see Fig. 5). The leaching depth at which no volume change occurs will be referred to as the transitional leaching depth. The interface progressively turns fully contractant as zone 5, which is the most porous (around 30% in Table 4), extends. The creation of a porous band or compaction band leads to contraction instead of dilation. Obviously, the deeper the leaching front, the wider the compaction band and consequently, the more the interface will contract.

Interestingly, the major part of shear strength is lost in the first 5 mm of leaching. This can very likely be related to the roughness of the interface and, more specifically in this case, to the height of the tooth (5 mm). Until a leaching depth of 5 mm, a part of the tooth is still intact creating a hard point in the contact, through which forces are still transmitted (see Fig. 6a and b). The most sig-

nificant loss of strength takes place during this first phase (around 50%). Passed the transitional leaching depth (5 mm), the tooth is totally weakened and there is no hard point in the contact. It can be seen from Fig. 6c that the transmission of forces within the contact is more uniform through the contact area. For more extended leaching, the shear strength keeps decreasing at a very slow rate and would eventually stabilize.

Numerical shear tests under increasing normal stress were also performed to identify the effect of leaching on the failure criterion of the contact. Mohr Coulomb criterion is not exactly the most appropriate failure criterion for rock joints due to a nonlinearity under low normal stress [3]. As a result of employing such criterion, very high values of cohesion can be found which is not realistic and surely not a conservative engineering approach. Patton [30] proposed a bilinear model to account for the low experimental cohesion, later improved by Jaeger [21] in an exponential continuous form (Eq. (3)).

$$\tau_p = (1 - e^{-\sigma/\sigma^*}) \cdot C_J + \sigma \cdot \tan(\phi_r) \tag{3}$$

where τ_p is the peak shear stress, σ is the normal stress, σ^* is the exponential decay parameter delimiting the low stress zone from high stress zone, C_J is the equivalent Mohr Coulomb cohesion and ϕ_r is the equivalent Mohr Coulomb friction angle. Barton [3] proposed another formulation based on the Joint Roughness Coefficient (JRC) and on the Joint Compressive Strength (JCS) (Eq. (4)).

$$\tau = \sigma \cdot \tan \left(JRC \cdot \log_{10} \left(\frac{JCS}{\sigma} \right) + \phi_b \right) \tag{4}$$

where τ is the peak shear stress, σ is the normal stress, and ϕ_b is the base friction angle.

Jaeger and Barton failure criteria have been estimated in Fig. 7a and b, respectively and the loss of mechanical strength clearly appears on the failure criteria. Fig. 7c shows the loss of friction angle and cohesion C_J for the Jaeger criterion. The two stage loss of mechanical strength is visible with most of the drop in equivalent friction angle and equivalent cohesion occurring in the first 5 mm of leaching. Regarding the Barton criterion, Fig. 7d shows the loss of JCS for the Barton criterion. Interestingly, in this criterion, a value of JCS lower than UCS indicates weathering of the wall. The lowest value of JCS considered by Barton [3] is a quarter of UCS. Here with leaching, JCS drops to around 1/70 of the UCS (see Table 2) suggesting that leaching is an extreme case of weathering. JRC and ϕ_b do not vary with leaching as they represents the roughness of the interface and the angle of frictional sliding resistance between particles, respectively.

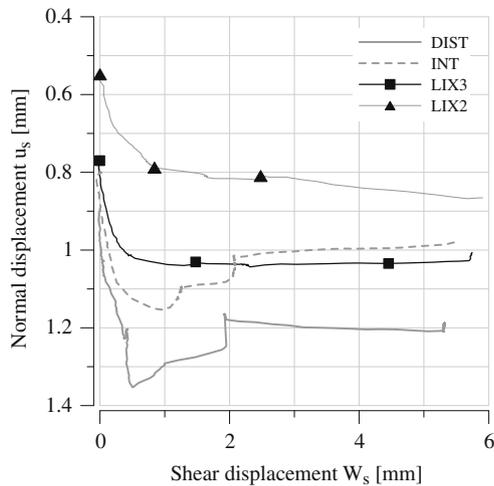


Fig. 5. Evolution of normal displacement u_s with respect to shear displacement W_s after Buzzi et al. [8].

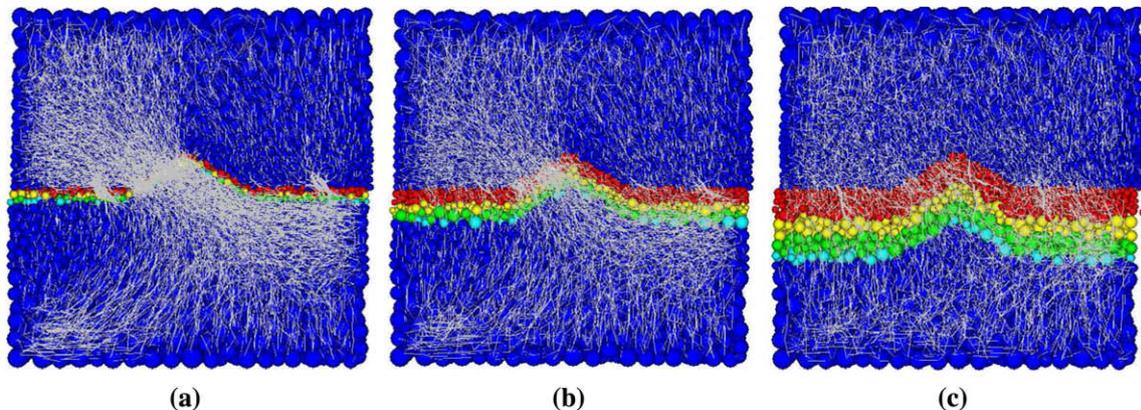


Fig. 6. View of the chain forces after 0.6 mm shear displacement for different leaching depths: 2 mm (a), 5 mm (b) and 10 mm (c). Thickness of the line represents the intensity of the contact force.

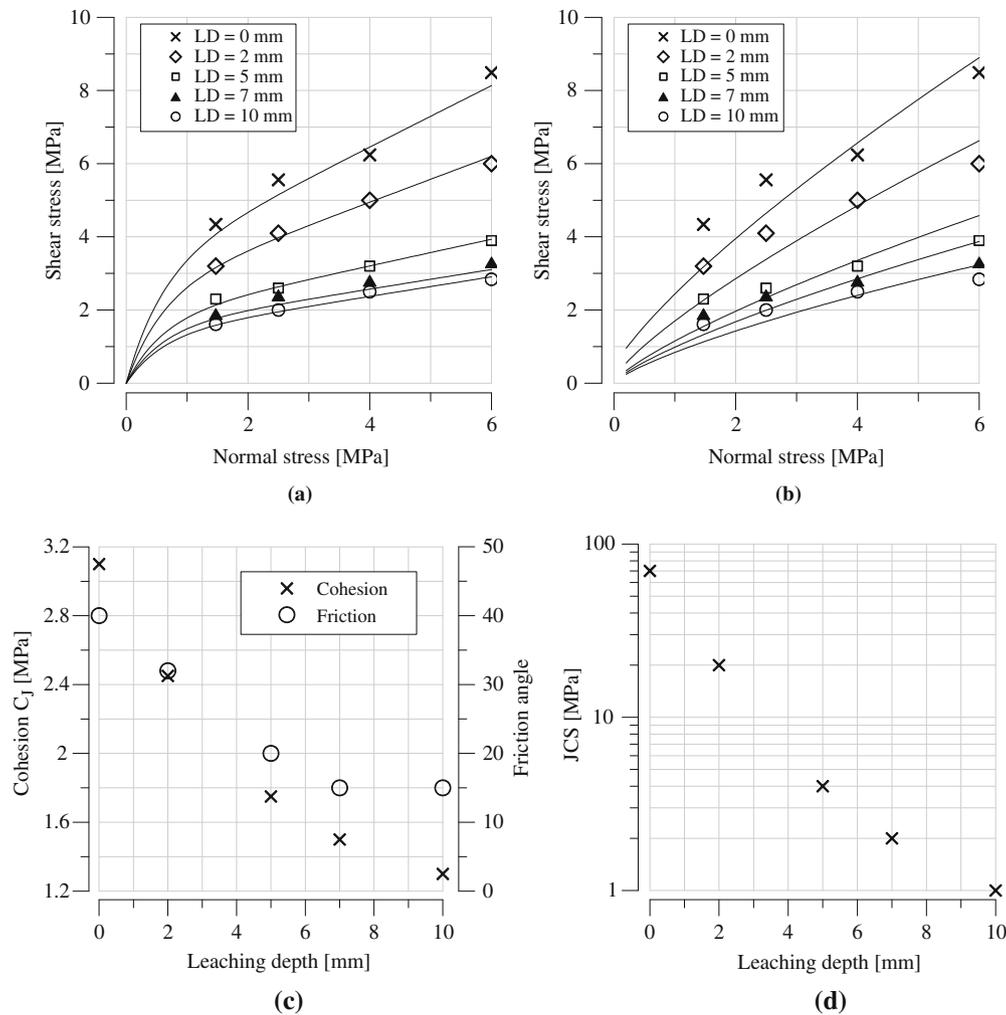


Fig. 7. Failure criterion of intact and leached interfaces: (a) Jaeger's criterion (Eq. (3) with $\sigma^* = 0.6$ MPa). (b) Barton criterion (Eq. (4) with $JRC = 15$ and $\phi_b = 40^\circ$, values selected for a best fit). (c) Evolution of equivalent friction angle and equivalent cohesion with leaching depth (Jaeger criterion). (d) Evolution of JCS with leaching depth (Barton criterion).

5. Application to a natural joint morphology

5.1. The interface model

The saw tooth interface being somehow an extreme morphology, simulations were also conducted on a smoother natural surface (see Fig. 8a). This surface comes from a natural discontinuity in granite studied by Grasselli [15]. The surface is around 70×70 mm² and the amplitude of the asperities is around 7.5 mm. Fig. 8b shows a view of the DEM model for this interface. The boundary conditions described in Section 4.1 were applied and tangential displacement was imposed along axis X.

5.2. Mechanical behavior of the intact natural interface

The mechanical response of the intact interface is plotted in Fig. 9 in terms of shear stress and normal displacement and it is consistent with the general mechanical behavior of rock joints available in the literature. The slight plateau before the peak in Fig. 9a is due to the interaction of the multiple asperities of the surface. It is here exacerbated by the scale of the horizontal axis and can actually be observed on experimental results, see for example in Hans and Boulon [17].

5.3. Mechanical behavior of the leached natural interface

The qualitative mechanical response of the leached saw tooth interface and the leached natural interface are very similar. A change from brittle to ductile behavior is observed with a significant loss of shear strength (from 5 MPa initially to 1.7 MPa under 15 mm of leaching) and the interface progressively turns fully contractant as leaching progresses (Fig. 10a). The transitional leaching depth i.e. that producing no volume change, appears to be around 7.5 mm here, which is the amplitude of the asperities height (Fig. 10b). The results obtained by Indraratna et al. [19] on rock joints with infill material suggest that the transition takes place for an infill thickness (or here a leaching depth) close to the asperity amplitude. This was effectively observed here on the saw tooth interfaces, on the natural interfaces and in the results by Buzzi et al. [8] where the asperity amplitude and the transitional leaching depth are around 5 mm.

When estimating the possible failure criteria of the interface (Jaeger and Barton criteria in Fig. 11), the two stage loss of mechanical strength appears clearly. The JCS drops from 70 MPa down to 0.5 MPa, the equivalent friction angle from 38° down to 20° and the equivalent cohesion from 1.46 MPa to 0.3 MPa. The JRC of the joint has been estimated to 12 visually using Barton's profiles and ϕ_b of 40° is slightly high but consistent for a granite [3]. From

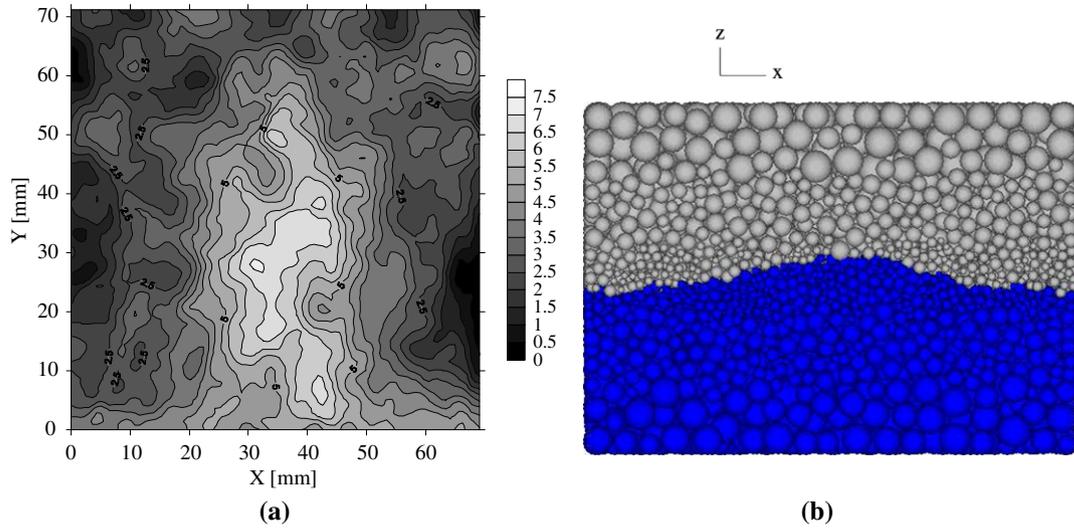


Fig. 8. (a) Representation of the granite surface. All dimensions in mm. (b) Cross section located at $Y = 35$ mm. Interface sheared along axis X .

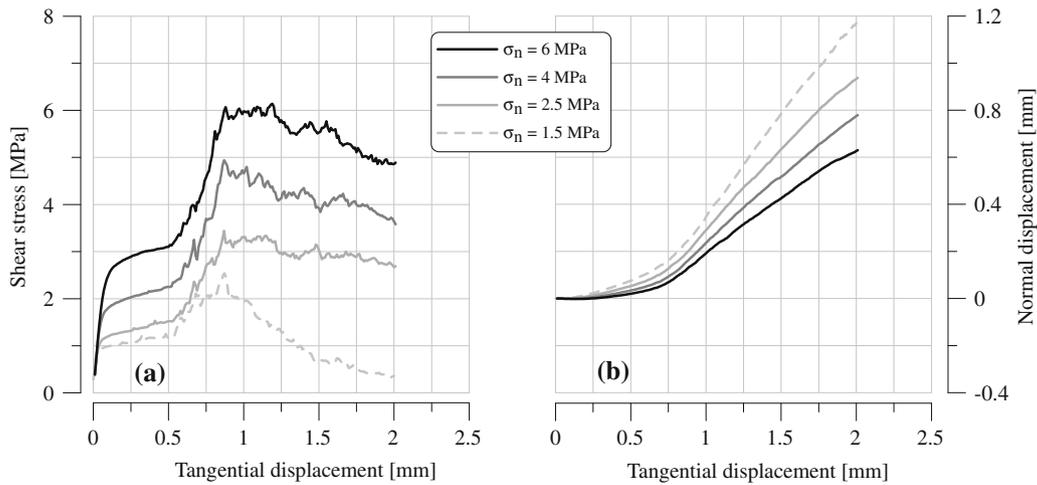


Fig. 9. (a) Evolution of shear stress versus tangential displacement for the intact natural interface. (b) Evolution of normal displacement versus tangential displacement. Shear test under constant normal stress of 4 MPa.

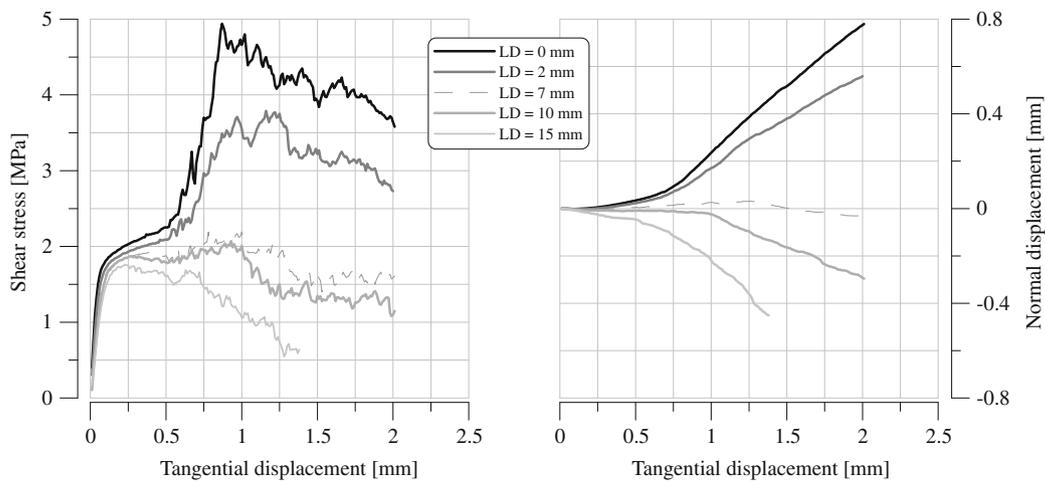


Fig. 10. (a) Evolution of shear stress versus tangential displacement. (b) Evolution of normal displacement versus tangential displacement. Shear test under constant normal stress of 4 MPa. Leaching depth ranges from 0 mm to 15 mm.

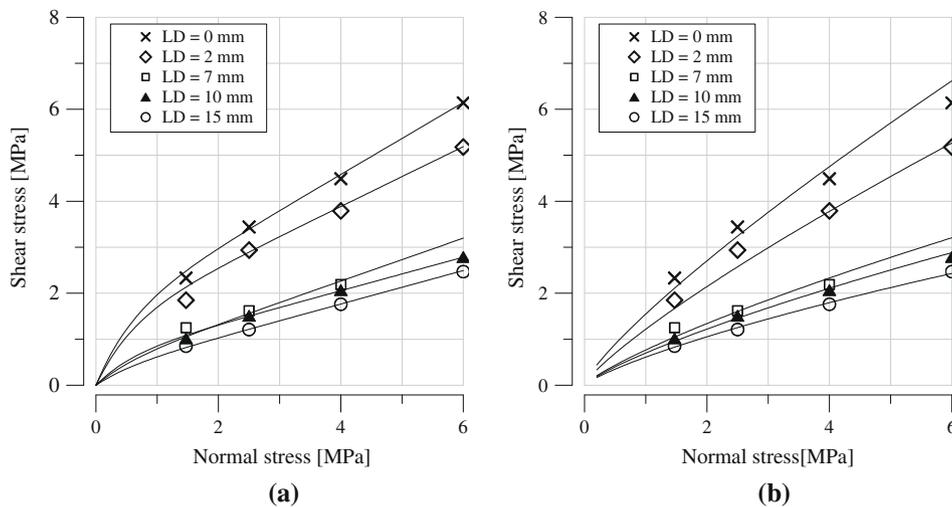


Fig. 11. Failure criterion of intact and leached interfaces: (a) Jaeger's criterion (Eq. (3) with $\sigma^* = 0.6$ MPa). (b) Barton criterion (Eq. (4) with $JRC = 12$ and $\phi_b = 40^\circ$).

an engineering point of view, the issue of leaching should be accounted for, if relevant, when initially defining the failure criterion for the interface. This could be done by determining the failure criterion at the transitional leaching depth. Indeed, most of the mechanical loss occurs before reaching this depth.

6. Significance for water flow

Transmissivity is a crucial parameter of the interface in the context of nuclear waste storage [8,7,11]. The leaching of the interface walls leads to a local increase of porosity in the bulk material and an augmentation of the interface transmissivity could logically be expected. On the contrary, Buzzi et al. [8] observed that the leached interface would not conduct water upon shearing before physical breakage of the interface. The results presented herein tend to corroborate the fact that, passed the transitional leaching depth, the leached interface does not necessarily conduct water. Indeed, it has been shown that passed the transitional leaching depth, the interface contracts upon shearing. As a result, the mechanical and hydraulic aperture also decrease and the overall transmissivity of the interface will decrease. The relationship between mechanical aperture, hydraulic aperture and transmissivity are well known [17,29,28] and also captured by the Reynolds lubrication equation [6] or by the cubic law [35].

7. Conclusions

Calcium leaching is a well known long term degradation phenomenon of cementitious materials in presence of water. The result of leaching is a local increase of porosity and a loss of mechanical strength and stiffness. However, most of the studies so far have been conducted on bulk materials and the impact of leaching on the behavior of interfaces have been somehow neglected despite its relevance in the context of nuclear waste repositories. The recent study by Buzzi et al. [8] showed that a leached rock–concrete interface exhibits a lower shear strength than an intact interface. Also, and against expectations, the leached interfaces did not seem to conduct more water than the intact ones. A numerical study was under taken here to extend the experimental study by Buzzi et al. [8] and to provide some new insight into the effect of leaching on the mechanical behavior of rock–mortar interfaces. The existence of a transitional leaching depth, at which the joint exhibits neither dilation nor contraction has been evidenced. This depth depends on the roughness of the interface and is roughly

equal to the amplitude of the asperities as shown by Indraratna et al. [19] for infill joints. It has been observed that most of the mechanical strength is lost before the transitional leaching depth. This translates clearly to the failure criterion of the interface, which does not vary much once the transitional leaching depth has been reached. Another remarkable feature is that the joint becomes fully contractant passed the transitional leaching depth, which has significant implications for water flow within the interface. Indeed, contraction of the interface means reduction in its transmissivity. This conclusion is very specific to the interfaces since, for bulk materials, leaching means increase of porosity and thus increase of permeability.

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