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EFECTUL LEVIGĂRII ASUPRA BETONULUI – PARTEA A II-A: EVOLUȚIA COMPORTĂRII MECANICE A ITZ ÎN TIMPUL LEVIGĂRII LA SCARĂ LOCALĂ

LEACHING EFFECT ON CONCRETE - PART II: MECHANICAL BEHAVIOUR EVOLUTION OF ITZ DURING LEACHING AT THE LOCAL SCALE

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Cement paste/aggregate bond influences durability of concrete subjected to leaching, following the existence of a particular, more vulnerable zone in cement paste, adjacent to aggregates - the interfacial transition zone (ITZ). In order to assess the mechanical behaviour of concrete at local scale of cement paste/aggregate bond, tensile tests were carried out on cement paste and cement paste/aggregate composite samples. The relationship between chemical degradation and mechanical properties (Young's modulus and tensile strength) was expressed through the notion of chemical degradation rate. Other aspects discussed, such as the leaching kinetics, chemical dissolution and cracking were used to highlight the origin of the loss of mechanical properties of the cement paste and of the cement paste/aggregate bond. Following an important dissolution within ITZ, a total loss of adhesion between cement paste and aggregate following leaching occurs gradually. Concerning the cement paste, it undergoes a smaller decrease of Young's modulus and tensile strength compared to the composites, being also less affected by cracking. Overall, this study highlights the mechanism by which the mechanical behaviours of cement paste and cement paste/aggregate bond are affected by leaching.

Legătura dintre pasta de ciment și agregat influențează durabilitatea betonului supus levigării, din cauza existenței unei anumite zone mai vulnerabile în pasta de ciment, adiacentă agregatelor - Zona de Tranziție Interfacială (ITZ). Pentru a evalua comportamentul mecanic al betonului la scara locală a legăturii pastă de ciment/agregat, s-au efectuat încercări de întindere pe eșantioane din pastă de ciment și compozite pastă/agregat. Relația dintre degradarea chimică și proprietățile mecanice (modulul lui Young și rezistența la întindere) a fost exprimată prin noțiunea de degradare chimică. Alte aspecte discutate, cum ar fi cinetica de levigare, dizolvarea chimică și fisurarea au fost evocate pentru a evidenția originea pierderii proprietăților mecanice ale pastei de ciment și a legăturii pastă / ciment. În ceea ce privește legătura pastă/agregat, după o dizolvare importantă în cadrul ITZ, se produce treptat o pierdere totală de aderență între pasta de ciment și agregat. În ceea ce privește pasta de ciment, aceasta suferă o scădere mai mică a modulului lui Young și a rezistenței la întindere în comparație cu compozitele, fiind de asemenea mai puțin afectată de fisurare. În general, acest studiu evidențiază mecanismul prin care comportamentele mecanice ale pastei de ciment și ale legăturii de pastă de ciment / agregate sunt afectate de levigare.

Keywords: Concrete, Leaching, Local Scale, Tensile Tests

1. Introduction

Concrete is a composite material formed by a cementitious matrix, the cement paste, which acts like a binder of the granular skeleton. Inside cement paste, a particular area forms in contact with the aggregates. This Interfacial Transition zone (ITZ) extends over a thickness of several tens of micrometres from the surface of the aggregate and has microstructural properties which influence the mechanical properties of cement paste/aggregate bond [1]. The higher porosity and higher content of chemical species with weak cohesive properties (portlandite) within ITZ make the cement paste/aggregate bond less resistant and less rigid [2, 3] than cement paste. These characteristics generates a significant effect at macroscopic scale, where the strength and rigidity of the concrete are

lower than in the ideal case where cement paste / aggregate bond would be perfect [4,5].

Leaching is a chemical pathology that affects concrete in contact with a relatively acidic chemical solution compared to its interstitial solution. This pathology produces the dissolution of calcium in the cementitious matrix and thus, following the increase in porosity it generates a decrease in its mechanical properties.

The effect of ITZ is also visible on degraded concrete. In fact, following leaching, the portlandite which is present in excess at the level of the ITZ is completely dissolved, thus giving way to a porosity which greatly reduces the adhesion between the cement paste and the aggregate. Another effect of leaching at interfaces is cracking due to stress concentrations generated by shrinkage of the paste [6]. Consequently, following these phenomena, a

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significant drop of mechanical properties of concrete occurs. In fact, in the literature, it has been observed that for the same overall extent of the degraded zone, the loss of compressive strength of the mortar, considered a special case of concrete, is greater than that of paste, because of significant chemical degradation of the ITZs [7].

Therefore, the effect of ITZ on the mechanical behaviour of healthy and degraded concrete by leaching is obvious and has been reported in several studies, essentially at macroscopic scale. However, the mechanical behaviour of the cement paste/aggregate bond is not known in detail, because of the difficulty of isolating this area to exhaustively observe its mechanical properties. These kind of observation are useful, since concrete structures are designed for longer and longer service lives and therefore it is necessary to take account of the mechanical behaviour of ITZ in life time predictions.

In this context, we present a study on the leaching effect on the mechanical properties of cement paste and cement paste/aggregate bond in order to highlight influence of ITZ, using a specific experimental protocol. This experimental protocol was set up to measure the mechanical properties of cement paste and of cement paste / aggregate bond [8] and it was extended in the present study to allow more localized characterisations. Beforehand, in part I of this study [9], the degradation kinetics and the extent of chemical dissolution were evaluated at the ITZ level and at the core of the cement paste. The results of this analysis will be presented in section 2.2 and will be used to interpret the evolution of the mechanical behaviour (rigidity, cracking, rupture) of cement paste and cement paste/aggregate bond according to chemical degradation. These mechanical properties of the material will be evaluated following micro indentation tests at microstructure of the paste and mechanical tensile tests on cement paste and composite cement paste/aggregate samples.

2. Materials and methods

2.1. Materials

The study of the mechanical properties of cement paste and cement paste/aggregate bond was realised on samples with same geometry and materials and submitted to the same leaching scenario as those used for chemical analysis in part 1[9]. The samples are parallelepiped (Figure 1) with total dimensions of 10x10x30 mm³ and a tolerance of ± 0.1 mm. The dimensions of the aggregate used to fabricate the composite sample are 10x10x15 mm³ and the same tolerance of ± 0.1 mm. From a mechanical point of view, the main advantage is to allow determination of the average

stress in the cross section during mechanical tests.

The materials used to manufacture the samples were chosen in order to get close to the formulations of high performance concretes that are likely to be used for structures subjected to leaching. This type of concrete is generally produced with reduced water/cement ratios (less than 0.42) and limestone aggregates in order to guarantee a satisfactory durability. Consequently, in this study, the cement paste was produced with CEM I R 52.5 CE CP2 NF cement and water/cement (w/c) ratio of 0.4. The aggregates were obtained from limestone rocks extracted from the Villeneuve-lès-Maguelone quarry (Hérault, France).

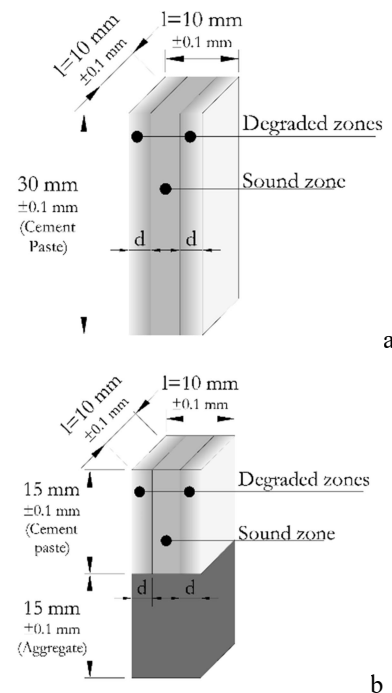


Fig. 1 - Geometries of cement paste samples (a) and composite samples (b)/ Geometria eșantioanelor din pastă de ciment (a) și compozite (b).

After casting, all the samples were kept in their mould in a 100% relative humidity atmosphere for one day and then demoulded. After demoulding, the samples were kept for 40 days in a water solution saturated with lime in order to prevent cracking due to drying shrinkage.

An accelerated degradation method was chosen in order to obtain a similar chemical degradation to the baseline scenario, but in a shorter time, immersing the samples in ammonium nitrate solution (NH_4NO_3). By isolating two longitudinal opposite faces, we obtained unidirectional degradation which generated two degraded zones on the sides and one sound zone in the middle (Figure 1). The samples were subjected to degradation for several leaching durations: 0.75, 2, 4, 8 and 12 days.

2.2. Chemical behaviour of the degraded samples

The main goals of the chemical characterization of the samples presented in part 1 [9] was to determine the leaching kinetics and to evaluate the dissolution state of the ITZ and of the cement paste.

In order to determine leaching kinetics, degraded depth d of the cement paste and of ITZ was measured on cross sections. Grey level contrast between sound zone (dark grey) and degraded zone (light grey) allowed delimitation of the sound and degraded zones (Figure 1). We observed that the evolution of the degraded depth of the cement paste was proportional to the square root of time. Concerning ITZ, its degraded depth evolution was similar with that of the paste at the beginning and then accelerated after 2 days to reach a 10 % maximum gap after 12 days. We mention that the acceleration of ITZ kinetics is due to the significant increase of porosity of ITZ due to leaching. Another factor that could accelerate the leaching kinetics of ITZ is cracking. In fact, pre-cracked composite samples after leaching and prior to mechanical tests are observed after 8 days of leaching (see section 3). However, no supplementary cracking effect is observed on ITZ leaching kinetics for the leaching times generated for this study.

Thus, as the degraded depths d were determined, in the present study we used the notion of chemical degradation rate δ in order to evaluate the effect of leaching on the mechanical properties of the samples. The chemical degradation rate δ is the ratio between the area of the degraded portion A_d of the cross section and the total area of the cross section A_t and was calculated with the formula :

$$\delta = \frac{A_d}{A_t} = \frac{2d \times l}{l^2} = \frac{2d}{l} \quad (1)$$

Whereas l is the width of the square cross section.

Regarding chemical dissolution, the composition of the ITZ and of the cement paste was evaluated through the Ca/Si molar ratio in the sound and degraded zones. In the sound state, the Ca/Si is significantly higher in ITZ than in cement paste, because of an important concentration of portlandite. The degraded zone was divided in an intermediate degraded zone and a highly degraded one. The fact that in the intermediate and highly degraded zones the Ca/Si ratios in ITZ and cement paste are similar testifies a greater dissolution of portlandite in ITZ. In fact, the more important dissolution in ITZ generates a superior increase of porosity which can subsequently affect the cement paste/aggregate bond.

2.3. Mechanical properties assessment

2.3.1. Micro indentation

Chemical analyses presented in part 1 [9] of the article were supplemented by micro indentation measurements supposed to reveal the effect of chemical degradation on the Young's modulus at the microstructure scale of cement paste. A single indentation supposes the insertion of a pointed and rigid tool on the surface of the material in a loading/creep/unloading sequence for a given maximum force [10]. Considering that at the beginning of the unloading phase, the material's response is elastic (Figure 2a), at this moment, the Young's modulus of the material can be calculated.

Access to the average properties of the microstructure is possible following the creation of sufficiently large indentation grids. It has been demonstrated in the literature that for indentation depths greater than 10 μm , access to the homogeneous mechanical properties of the cement paste is possible [11, 12]. Consequently, in this study, the maximum indentation force was chosen 5 N, in order to obtain an average maximum depth of 15 μm in the sound zone. Indentation grids of 23x5 indents were carried out in the degraded zone of the bulk cement paste, while in the sound zone a line of 23 indents served as a reference (Figure 2b). We thus obtained the evolution of the Young's modulus in the degraded zone and the average Young's modulus in the sound zone.

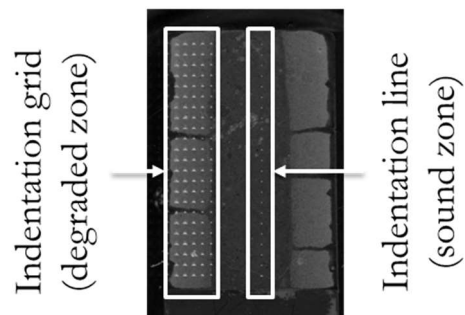
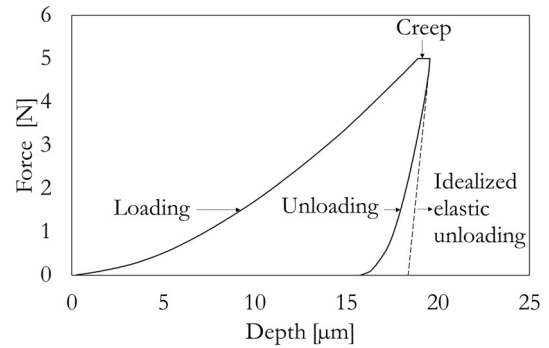


Fig. 2 - Single indentation force/depth example (a); location of the indentation grid (b) / Exemplu de curbă forță/adâncime pentru o indentare individual (a); localizarea grilei de puncte de indentare (b).

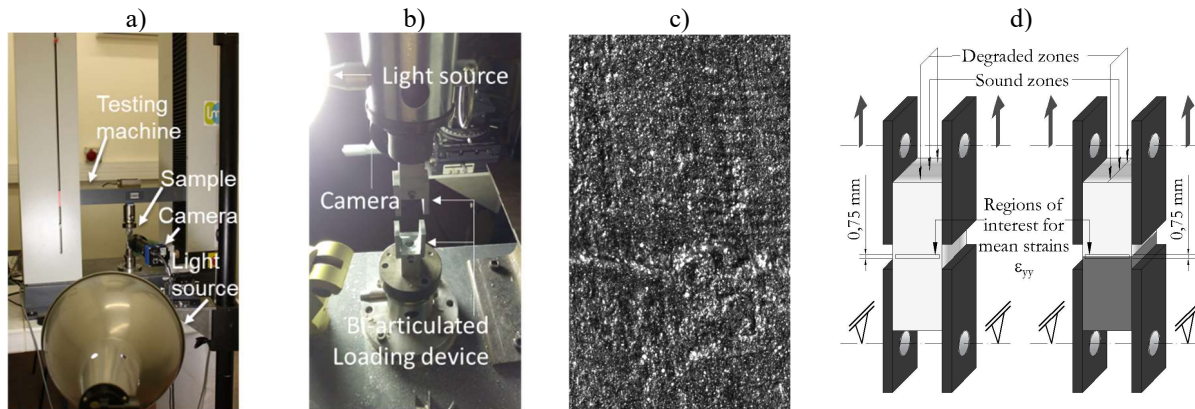


Fig.3- Global system used for tensile test (a); loading double bi-articulated system (b); random pattern applied on a composite sample in order to allow the DIC (c); location of the region of interest for mean strains (d) / Sistemul global folosit pentru încercarea de tracțiune (a), sistem de încărcare biarticulat (b); motiv aleatoriu aplicat pe suprafața unui eșantion compozit pentru a permite Corelarea Numerică de Imagini (CNI) (c); localizarea zonei de interes pentru calculul deformațiilor medii (d).

2.3.2. Tensile tests

Mechanical tensile tests were carried out on sound and degraded cement paste and composite samples in order to determine the average stress/strain curves. Testing system is composed of a MTS machine for mechanical loading and measurement of force while a high resolution camera, coupled with a light source was used for measurement of the displacement (Figure 3a). The camera used to record the images is monochrome, with a resolution of 16 megapixels, or 7.7 micrometres/pixel.

In order to obtain mechanical loading of samples, a displacement rate of 10 $\mu\text{m}/\text{sec}$ was imposed by the machine. The test device demanded gluing four loading plates to the sample and the transmission of the mechanical load to the axes which pass through the holes in the plates. This system was implemented in a previous work [13]. The load transmission part from the machine to the axes is articulated by a pivot. This gives rise to a bi-articulated loading device at both ends (top and bottom) (Figure 3b) which is supposed to apply the load uniformly on the cross section of the sample which is perpendicular to the loading axis. The double bi-articulation of the loading system was necessary because in the absence of the pivots, the geometric defects of the gluing of the plates caused uneven loading. Indeed, for this type of tensile test, Lhonneur [14] indicated that in order to have a precise estimation of the mean strength (or force at rupture), 10 tests are necessary. Therefore, for each degradation time, 10 tests were carried out on the sound and degraded samples. The tensile stress σ_T has been calculated with the formula:

$$\sigma_T = \frac{F}{A} \quad (2)$$

whereas F is the total tensile force and A is the cross' section surface area.

In order to obtain the average strain associated to the tensile stress, the displacement fields were calculated beforehand over the entire face of the sample during loading by Digital Image Correlation (DIC). By using this technique, local displacements were calculated [15, 16] following detecting spatial variations of a random pattern recorded on one face of the sample (Figure 3c). The mesh size of the displacement fields was chosen 0.375 mm. Strain fields were obtained by the derivation of the displacement fields, with the same mesh size. Mean strains were calculated within an area located in the central region of the sample (Figure 3d). They were obtained by averaging the local strains located inside this area. For the degraded samples, the measurement of the strain was carried out on an attacked face while the plates were glued on a face that is parallel to the leaching's direction of propagation (Figure 4d). One thus obtains a parallel loading of the sound zone and of the degraded zones.

3.Results

3.1. Micro indentation

The results of the micro indentation tests reveal the evolution of the Young's modulus in the degraded zone of cement paste, as well as the mean Young's modulus of the sound zone. As in the case of the chemical analysis of the Ca/Si ratio, an intermediate degraded zone and a highly degraded zone can be observed. In the intermediate degraded zone, the Young's modulus drop is pronounced, while in the highly degraded zone, the value of the Young's modulus is lower and its decrease is less pronounced (Figure 4). Indeed, by representing the Young's modulus evolution, the Ca/Si molar ratio and the grey level in the cement paste [9] after 4 days of degradation, we notice a similar evolution in the degraded zone (Figure 4). Consequently, the grey level can be seen in this case a revealing indicator of the chemical degradation state and of the loss of

mechanical properties. Furthermore, from part 1 [9] observations, it can be seen that for all leaching times, the degraded zone of cement paste and ITZ is composed of an intermediate degraded zone and of a highly degraded zone. Therefore, by extrapolating the results obtained at 4 days of leaching, it can be assumed that mechanical properties follow the same trend as grey level and Ca / Si ratios. In fact, this observation confirms others in literature who have noticed a correlation between the decalcification and the micro hardness decrease, to estimate thus the value of the Young's modulus in the degraded area [17].

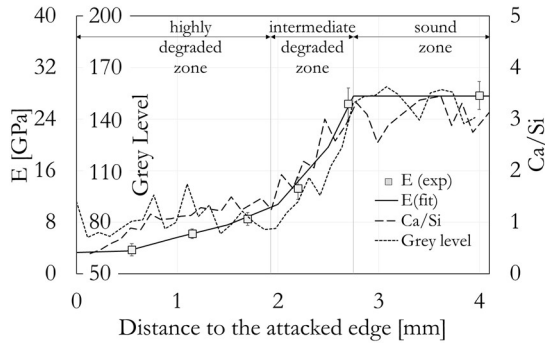


Fig. 4- Evolution of Young's modulus E as a function of the distance from the attacked edge and comparison between the evolution of the Young's modulus, of Ca / Si molar ratio and of grey level in the degraded zone *Evoluția modulului lui Young ca funcție de distanța de la fața atacată și comparație între evoluția modulului lui Young E , a raportului molar Ca/Si și a nivelului de gri în zona degradată.*

3.1.1. Mechanical tests

After evaluating the chemical degradation effect at microstructure scale, overall mechanical properties of samples were obtained from the stress/strain curves. These curves were obtained following fitting of the raw stress/strain curves which were affected by measurement noise. Due to the very small measured strains, which approach the limits of the method, this noise originates from the difficulty of carrying out precise local measurements and has no proper physical sense. In addition, the use of a high-resolution camera necessary to detect local displacements and strains, did not make it possible to capture the post-peak behaviour, because of the low recording rate (4 Hz). The post-peak part of the curves has been approximated from the force/displacement curves of the loading machine which indicate a fragile rupture and therefore has been represented in the graphs by a thinner vertical line. These stress/strain curves are presented in Figure 5.

In this figure, it can be observed that following chemical degradation, cement paste and composite samples undergo a loss of tensile strength σ_{Tr} and of Young's modulus. In addition, with degradation, an increasingly non-linear portion appears. This non-linear portion occurs following cracking onset. The onset of a crack on the

observed face results in a change of the stress/strain curve slope. The stress/strain curves were represented up to a degradation rate of around 60%, corresponding to 4 days of leaching, because after this point, the samples were pre-cracked before loading and the linear part no longer existed.

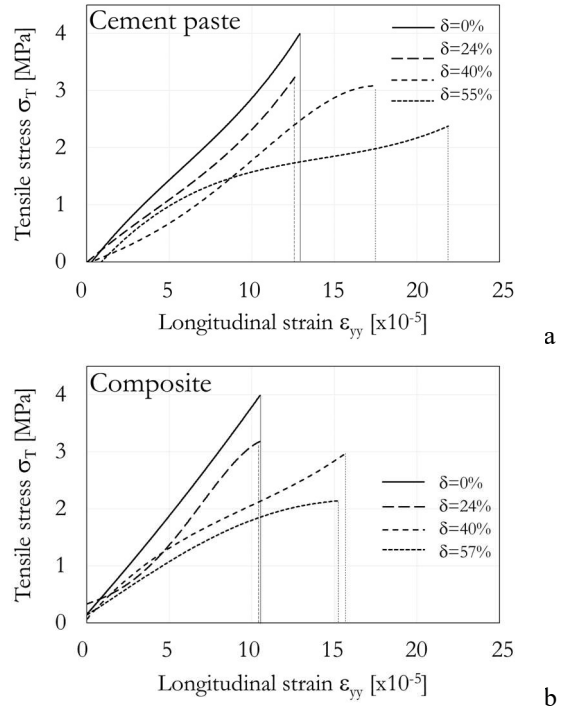


Fig. 5 - Stress/strain curves of sound and degraded samples in cement paste (a) and composite (b) during tensile test *Curbele efort unitar/deformație a eșantioanelor nedegradate și degradate din pasta de ciment (a) și compozite (b) la încercarea de întindere.*

From the linear part of the curves, within the material's behaviour is assumed elastic, the Young's moduli of the cement paste and of the composites were calculated, their evolution as a function of the chemical degradation rate being represented in Figure 6a. Following these results, the evolutions of the Young's moduli as a function of the degradation rate of cement paste and composites were modelled by polynomial curves. A priori, there is a more pronounced decrease in the Young's modulus of the composite compared to the cement paste. However, the direct comparison of cement paste and composite modulus is not relevant, because composite contains a half aggregate whose properties do not vary. Under these conditions, in order to take into consideration, the effect of ITZ, an ideal elastic modulus corresponding to perfect bond was calculated for each chemical degradation rate as a reference. Considering the loading configuration, the cement paste and the aggregate can be modelled as two springs linked in series, whose equivalent module corresponding to a perfect bond E_{cp} can be calculated with the formula:

$$E_{cp} = \frac{2}{\frac{1}{E_p(\delta)} + \frac{1}{E_g}} \quad (3)$$

Whereas $E_p(\delta)$ represents the Young's modulus of cement paste for the degradation rate δ and E_g represents the Young's modulus of limestone aggregate used in the present study which is equal to 60 GPa. Subsequently, the Young's modulus of composite corresponding to a perfect bond is proportional to that of cement paste.

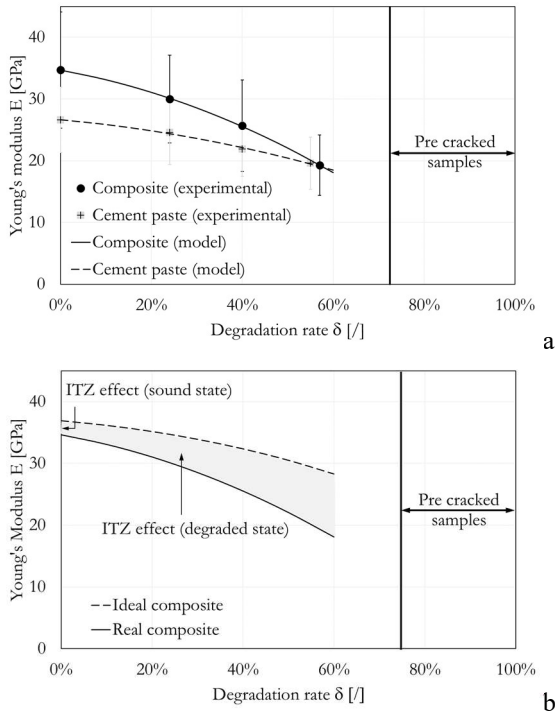


Fig. 6 - Evolution of the Young's modulus of the cement paste and the composite as a function of the degradation rate(a) and evolution of the Young's modulus of the real and ideal composites as a function of the degradation rate(b) / *Evoluția modulului lui Young al pastei de ciment și a compozitului ca funcție de rata de degradare (a) și evoluția modulului lui Young al compozitelor reale și ideale în funcție de rata de degradare (b).*

By comparing the evolution of Young's modulus of the real composite with that of the ideal composite corresponding to the perfect bond, we note that the value for the real one is lower and that of the ideal one. Besides that, the difference increases while degradation rate increase (Figure 6b). This is due to the existence of ITZ, which has been previously proven (part 1 [9]). On the one hand, the fact that its existence makes the contact imperfect in the sound state is manifested by the difference between the two Young moduli, that of the real composite and of the ideal composite. On the other hand, the higher chemical degradation of the ITZ compared to the bulk cement paste generates a greater loss of Young's modulus of the real composite compared to the ideal composite

which does not contain ITZ.

Concerning the tensile strength of the cement paste and of the composites, its evolution as a function of the chemical degradation rate is presented in Figure 7a). As in the case of the Young's modulus, the evolutions of the tensile strengths were approximated by polynomial functions. By comparing the tensile strengths of the cement paste and of the composite, it is observed that for chemical degradation rates lower than 60%, they are almost equal, but slightly lower for the composites. For these chemical degradation rates, the majority of composite samples undergo ruptures in the bulk cement paste, while ruptures at the interface occur in maximum 30% of cases. On the other hand, for chemical degradation rates greater than 70%, the tensile strength of the composites exhibits an accentuated decrease, since rupture occurs at the interface (Figure 7b). Indeed, for a chemical degradation rate of 100%, the tensile strength of the composites is almost zero.

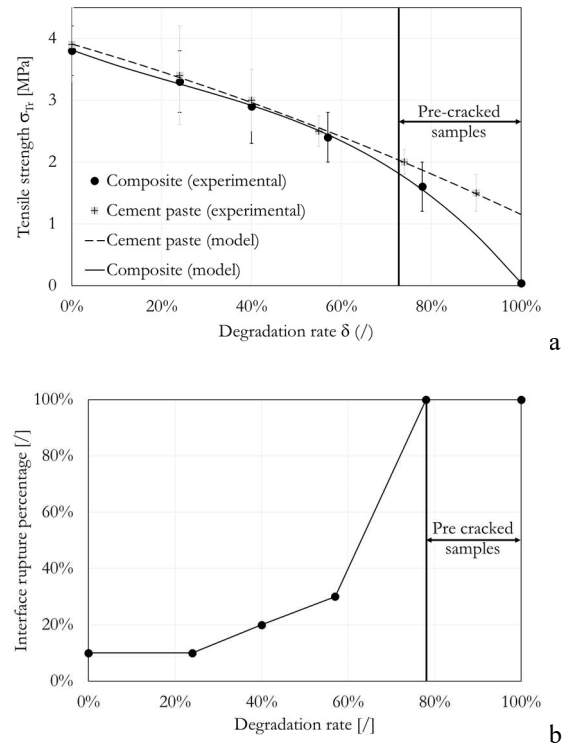


Fig. 7- Evolution of the tensile strengths of the cement paste and of the composite samples as a function of the chemical degradation rate(a) and evolution of the percentages of composite samples which present ruptures at the interface as a function of the chemical degradation rate (b) / *Evoluția rezistenței la întindere a eșantioanelor din pasta de ciment și compozitelor pasta de ciment/agregat ca funcție de rata de degradare (a) și evoluția procentajului de eșantioane compozite care prezintă ruperi la interfață ca funcție de rata de degradare (b).*

By comparing the evolution of the tensile strength of the cement paste to that of composite, we found appropriate to observe the influence of cracking on the two types of samples. An obvious difference appears between the cement paste and

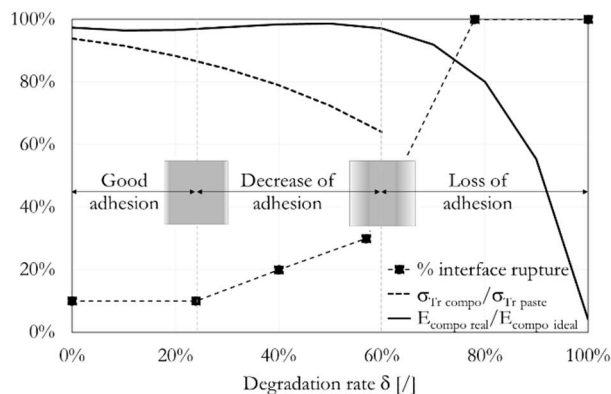


Fig. 8 - Evolution of the quality of the cement paste/aggregate adhesion as a function of the chemical degradation rate
Evoluția calității adeziunii dintre pasta de ciment și agregat ca funcție de rata de degradare.

the composite for chemical degradation rates greater than 70% (Figure 8a). Indeed, after this threshold, the cement paste and the composites samples are pre-cracked before loading. The pre-cracking of the cement paste and of the interface is generated by stress concentrations which accumulates during the leaching following the cement paste's shrinkage. These stress concentrations appear in the degraded zone of the cement paste, but especially at the cement paste/aggregate interface. This phenomenon has been described and modelled in detail by Rougelot et al. [6].

However, by comparing the consequences of pre-cracking of the cement paste and of the cement paste/aggregate bond, the difference is important. On cement paste samples, the effect is barely visible, since the decrease in strength follows the initial trend. On the other hand, on the composites, the ruptures begin to occur at the interface (Figure 7b) and the decrease of tensile strength visibly accelerates (Figure 7a). Indeed, the cracks observed on the samples are located in the observed face, that attacked by the aggressive solution (Figure 3c), therefore belonging to the highly degraded zone. Considering that the degradation of ITZ is more pronounced, deeper cracking could be facilitated, while in cement paste, it could be limited to a more superficial area. Overall, the more pronounced loss of mechanical properties of ITZ compared to cement paste confirms an aspect which is known in the literature, but mainly indirectly deduced through macroscopic studies on cement mortar and cement paste[7].

After the observations made on the evolution of mechanical properties as a function of the chemical degradation rate, we globally observed three phases in the mechanical behaviour of the composite. Regarding the quality of the cement paste/aggregate bond, there is a phase characterized by good adhesion of the degraded zone, a "decrease of adhesion" phase and a "loss of adhesion" phase (Figure 8). In the "good adhesion phase", the loss of Young's

modulus of the composite compared to that of the cement paste as well as the percentage of composite which present ruptures at the interfaces are reduced, while the rupture occurs for tensile strengths almost equal to those of the cement paste. In the "decrease of adhesion phase", the decrease of the Young's modulus of the composite accelerates and the number of composites which exhibit ruptures at the interface increases, but the tensile strengths of the composite remain close to that of the cement paste. In the "loss of adhesion phase", the composite samples are pre-cracked before the tensile test, exhibit interface ruptures and the tensile strength's drop is remarkable.

Subsequently, a comparison can be made between the results obtained in the present study and those obtained by Jebli [13] who carried out a test campaign on cement paste and composite samples at the same scale. The difference between the materials used in this study and the present one consisted in the fact that Jebli used a specific formulation to ordinary concretes: CEM II LL 32.5 cement, w/c of 0.5 and limestone aggregates. In that study, the tensile strengths of the cement paste (2.4 MPa at sound state), but mainly of the composite samples (1.8 MPa at sound state) were visibly lower compared to those obtained in the present study, while all of the composites had ruptures at the interface. Another important difference was noticed in the evolution of the mechanical properties. After the start of chemical degradation, the composite samples underwent a rapid drop of strength. After that, the decrease of tensile strength as a function of the chemical degradation rate has diminished, but the strengths were significantly lower compared to those obtained in the present study. So, by comparing the results of the two studies, we notice that the use of a cement with a higher resistance class and the reduction of the lower w/c ratio had a beneficial effect on the quality of the adhesion between the cement paste and aggregate subject to leaching.

4. Conclusion

This study globally highlighted the process by which the chemical degradation produced by leaching affect the mechanical properties of concrete at local scale (cement paste and cement/aggregate bond), by revealing the ITZ role. Also, the beneficial effect of the use of a cement with a higher strength class and lower w/c ratio was pointed.

Overall, leaching generates a decrease of Young's modulus and tensile strength of the cement paste and of the cement paste/aggregate composites samples used in the present study. From a chemical and mechanical point of view, the degraded zone of cement paste and ITZ is composed of an intermediate degraded zone and

a highly degraded zone.

Differences that have been noticed between the chemical degradation of cement paste and that of ITZ had consequences in the mechanical behaviour of cement paste and cement paste/aggregate bond. Important chemical dissolution within ITZ generates a significant increase of porosity and subsequently a more important loss of Young modulus of the cement paste/aggregate bond with respect to bulk cement paste.

Furthermore, the presence of ITZ and its pronounced chemical degradation results, coupled probably with shrinkage effect results in a progressive loss of adhesion of the cement/aggregate bond. The decrease of tensile strength of the composite is greater than that of the cement paste and culminates with a complete loss of adhesion of the cement/aggregate bond in the degraded area.

In perspective, the approach presented in this study could be continued in order to determine the constitutive laws mainly of the cement paste/aggregate bond (by considering the ITZ), but also of cement paste at the local scale. The use of specific geometries could guide the rupture to a known location (e.g. the interface) in order to determine the resistance and the post-peak behaviour of the material. At the same time, the adaptation of strain measurement DIC techniques to the specificity of the material (very low strains and fragile behaviour) is supposed to allow the determination of localized strains of the ITZ (or of the interface) as well as the consideration of shrinkage effects.

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REFERENCES

- [1] J. P. Ollivier, J. C. Maso, and B. Bourdette, "Interfacial Transition Zone," *Adv. Cem. Based Mater.*, 1995, **2**(1), 30-38.
- [2] A. Sidorova, E. Vazquez-Ramonich, M. Barra-Bizinotto, J. J. Roa-Rovira, and E. Jimenez-Pique, "Study of the recycled aggregates nature's influence on the aggregate-cement paste interface and ITZ," *Constr. Build. Mater.*, 2014, **68**, 677-684.
- [3] P. Trtik and P. J. M. Bartos, "Micromechanical properties of cementitious composites," *Mater. Struct.*, 1999, **32** (5), 388-393.
- [4] P. K. Mehta and P. J. M. Monteiro, "Concrete Microstructure, Properties, and materials", 2006, McGraw-Hill Publishing,.
- [5] Z. Sun, E. J. Garboczi, and S. P. Shah, "Modeling the elastic properties of concrete composites: Experiment, differential effective medium theory, and numerical simulation," *Cem. Concr. Compos.*, 2007, **29**(1), 22-38.
- [6] T. Rougelot, N. Burlion, D. Bernard, and F. Skoczylas, "About microcracking due to leaching in cementitious composites: X-ray microtomography description and numerical approach," *Cem. Concr. Res.*, 2010, **40**(2), 271-283.
- [7] C. Carde and R. François, "Effect of ITZ leaching on durability of cement-based materials," *Cem. Concr. Res.*, 1997, **27**, 971-978.
- [8] M. Jebli, F. Jamin, E. Malachanne, E. Garcia-Diaz, and M. S. El Youssoufi, "Experimental characterization of mechanical properties of the cement-aggregate interface in concrete," *Constr. Build. Mater.*, 2018, **161**, 16-25.
- [9] A. Girboveanu, M. Jebli, F. Jamin, V. Huon, M. S. El Youssoufi, and D. Georgescu, "Leaching effect on concrete-part I: characterization of chemical degradation evolution of ITZ," *Rom. J. Mater.*, 2020, **50** (1), 129-137.
- [10] W. C. Oliver and G. M. Pharr, "An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments," *J. Mater. Res.*, 1992, **7**(6), 1564-1583.
- [11] S. Igarashi, A. Bentur, and S. Mindess, "Microhardness testing of cementitious materials," *Adv. Cem. Based Mater.*, 1996, **4**(2), 48-57.
- [12] Y. Wei, S. Liang, and X. Gao, "Indentation creep of cementitious materials: Experimental investigation from nano to micro length scales," *Constr. Build. Mater.*, 2017, **143**, 222-233.
- [13] M. Jebli, F. Jamin, C. Pelissou, E. Malachanne, E. Garcia-Diaz, and M. S. El Youssoufi, "Leaching effect on mechanical properties of cement-aggregate interface," *Cem. Concr. Compos.*, 2018, **87**, 10-19.
- [14] J. Lhonneur, A. Girboveanu, F. Jamin, C. Pélissou, Y. Monerie, and M. S. El Youssoufi, "Étude statistique de la réponse mécanique lors d'essais de traction directe à l'échelle locale," 2019, in 14e Colloque National en Calcul de Structures-CSMA 2019.
- [15] B. Wattrisse, A. Chrysochoos, J.-M. Muracciole, and M. Némot-Gaillard, "Analysis of strain localization during tensile tests by digital image correlation," *Exp. Mech.*, 2001, **41**(1), 29-39.
- [16] V. Huon, B. Wattrisse, M. S. El Youssoufi, and A. Chrysochoos, "Elastic behavior of anisotropic terra cotta ceramics determined by kinematic full-field measurements," *J. Eur. Ceram. Soc.*, 2007, **27**(5), 2303-2310.
- [17] B. Gerard, O. Didry, J. Marchand, D. Breyse, and H. Hornain, "Modelling the long-term durability of concrete for radioactive waste disposals", 1996, in Proceedings of MRS Fall meeting