Evolution of Air Permeability of Concrete due to Expansion Caused by Internal Swelling Reactions (ISR)

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Abstract. Predicting and evaluating the transfer properties of concrete affected by Internal Swelling Reactions (ISR) is a main challenge for experts involved in concrete durability testing. The aim of this study is to measure the evolution of air permeability with the development of ISR, for different levels of expansion and various degrees of concrete saturation. In this work, two ISR were studied: the Alkali-Silica Reaction (ASR) where the origin of the swelling is located in the aggregates, and the Delayed Ettringite Formation (DEF) where the origin is located in the cement matrix. The first results obtained show that the development of ISR and induced cracks leads to an increase in the air permeability of concrete, especially in highly saturated concrete. The data of this study make it possible to evaluate the evolution of the transfer properties according to the generated expansion and induced cracking and the degree of saturation of the concrete.

Keywords: Durability · Permeability · Expansion · AAR · DEF

1 Introduction

Internal Swelling Reactions (ISR) are endogenous and deleterious chemical reactions that cause pressure in concrete, leading to the formation of cracks in the material and in the affected structures. In the literature, most studies characterize the evolution of mechanical properties with expansion, but few are interested in the transfer properties. It has been shown that even if these reactions lead to a decrease in the mechanical properties of the concrete, they do not necessarily lead to a decrease of their mechanical safety [1]. However, the induced cracking obviously affects the durability of the material. The transport properties of concrete play a key role in predicting the durability of concrete structures [2–4]. In the case of structures with a containment function, it is necessary to quantify the changes in permeability caused by the development of ISR. Gas permeability also allows, as a characteristic indicator, the evaluation of the sustainability potential of the structures concerned, regardless of their function.

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The aim of this study is to experimentally characterize the interaction between the physico-chemical state and the transport properties of damaged cementitious materials. The measurements will be performed on concretes affected by Alkali-Silica Reaction (ASR) and Delayed Ettringite Formation (DEF) to represent different types of expansion and different cracking patterns. Permeability measurements should be taken at several well-defined times to assess the effect of crack connectivity and the state of filling by reaction products.

On site, the degree of saturation of the core of massive structures and the concrete cover exposed to rain is often high (higher than 80% [5]). With climatic variations, the saturation of structural concrete varies between 30 and 80%. For a saturation rate higher than 60%, air transport in ordinary concrete is low due to the continuity of the liquid phase [3]. Moreover, the Kelvin Laplace equation shows that a crack with an opening larger than one micrometer will be drained even at a very high saturation rate (99.99%). Knowing that the air permeability of concrete is largely influenced by its degree of saturation [3, 6, 7] it is relevant to perform measurements at different saturation rates of cementitious materials damaged by ASR and DEF.

2 Materials and Methods

2.1 Concrete Damaged by ISR

In the present study, two types of concrete were investigated:

- B11 is used as a reference concrete (curing time at 20 °C) and to obtain DEF damaged concrete (specimens cured for three days at 80 °C at young age),
- T11 is used to obtain concrete developing ASR

The water-to-cement ratio (w/c) was of 0.56. These concrete mixes were selected to induce the development of ISR as well as for their representativeness in comparison with the concretes used in nuclear civil engineering structures.

The cement adopted for the realization of two concrete mixes is CEM II A-LL 42.5 R CE PM-CP2 NF. The chemical composition of the cement is given in Table 1.

Table 1. Components and mineral composition (%) of cement

S_iO_2	Al_2O_3	Fe_2O_3	T_iO_2	MnO	CaO	MgO	SO_3	K_2O	Na_2O	P_2O_5	Loss on ignition
19.7	4.6	3.2	0.3	0.1	63.5	1.2	2.7	1.36	0.11	0.5	3.6

For B11 concrete, specimens were made using a calcareous limestone aggregate (0/4, 4/11.2 and 11.2/22.4 mm) considered as non-reactive to ASR (NR) from the Center of France, (2650, 2630 and 2630 kg/m^3 respectively; water absorption 2.8, 2.3 and 2.1% respectively).

For Alkali-Silica Reaction, concretes were produced with different types of aggregates:

- A non-reactive (NR) 0/4 mm calcareous limestone sand from France, Boulonnais quarry (2650 kg/m^3; water absorption 0.5%)
- A potentially reactive (PR) (4/14 and 14/20 mm) aggregate formed of silica and limestone from the Northern France (density 2660 kg/m^3; water absorption 0,5%; reactive silica content 7%)

A high-range water-reducing admixture was exclusively for the B11 concrete: MasterGlenium 21 (BASF) (1.06 g/cm^3; dry extract EN 480-8 12.9). The T11 concrete was produced without any admixture.

The mixing water of both mixes was doped with NaOH in order to obtain an equivalent alkali content of 5 kg/m^3.

Table 2 shows the concrete mixes used for the reference concrete and the concrete affected by DEF.

Table 2. B11 concrete mix (kg/m^3)

Ciment CEM II	Calcareous limestone aggregate			Water-reducing admixture	Water	Na_2O_{eq}
	0/4 mm	4/11.2 mm	11.2/22.4 mm			
350	772	316	784	1,225	195	5

Table 3 presents the concrete mix used for concrete affected by ASR.

Table 3. T11 concrete mix (kg/m^3)

Ciment CEM II	Calcareous aggregate	Silica and limestone aggregate		Water	Na_2O_{eq}
	0/4 mm	4/14 mm	14/20 mm		
350	772	316	784	195	5

In this study, it was decided to adopt the ISR degradation strategies proposed by IFSTTAR [8] and [9] with minor modifications to accelerate the DEF kinetics.

Table 4 shows the preservation and conditioning protocols for both types of concrete.

Table 4. Conservation and conditioning protocols

DEF	ASR
7-day heat treatment in climatic chamber	Endogenous cure
Endogenous cure from 7 to 28 days	
At the end of the cure, all the specimens were immersed in water at 38 °C. A pump was used to homogenize the solution in terms of temperature and ionic species present in the water	

2.2 Experimental Techniques

2.2.1 Expansion Monitoring

The evolution of ISR was characterized by the concrete expansion. The monitoring of these expansions was based on longitudinal deformation measurements of the specimens using an extensometer. For this purpose, 6 stainless steel studs were glued in pairs to obtain 3 expansion measurements according to the height of the specimen. The pairs of studs were spaced 10 cm apart (deformation measurement base) at equal distances from the mid-height of the specimen. The three measurement lines were separated by 120° on the lateral surface of the specimen.

2.2.2 Permeability Measurement

The air permeability measurements of the cylindrical samples were performed using a Cembureau type constant permeameter according to the standard (XP P18-463, 2011). In order to obtain a satisfactory accuracy of the intrinsic permeability, the Cembureau test includes four measurements of the apparent permeability at four injection pressures applied for each measurement ranging from the following values: 1, 2, 3 and 4 bars (0.1, 0.2, 0.3 and 0.4 MPa) for three different samples.

The apparent permeability is obtained from to the following equation [10]:

$$k_A = \frac{Q_s}{S}\frac{2\mu L P_s}{\left(P_e^2 - P_s^2\right)} \tag{1}$$

Where: k_A is the apparent permeability (m^2), Q_s is the volume flow (m^3/s), μ is the dynamic viscosity of the air (Pa.s), L is the thickness of the sample, P_s is the absolute pressure at the outlet (Pa), S is the section of the sample (m^2) and P_e is the absolute pressure at the inlet (Pa).

The degree of saturation of the concrete has a strong influence on the air permeability [6], therefore, six states of saturation were chosen (80, 60, 40, 20, 3 et 0%) to perform the permeability measurements. In our study, the choice of the drying temperature was very delicate. Since ettringite is metastable with increase temperature (especially above 60 °C), it was important to minimize the effects of temperature on the microstructure of the original and damaged concrete. Thus, for the drying method, it was proposed to apply drying by steps of increasing temperature as presented in Table 5.

Table 5. Conditioning protocol followed in this study

Saturation rate (%)	Drying temperature (°C)
From 100 to 30	40
From 30 to 20	50
From 20 to 3	80
From 3 to 0	105

This approach reduced the moisture gradients that can induce shrinkage gradients. Such gradients can be the cause of microcracking of the specimens during preconditioning. At the end of each drying and just before the permeability measurement, the concrete saturation is homogenized (for at least the same time as the drying). This step helps to unify the saturation within the sample trying to obtain a homogeneous permeability in the concrete.

The main interest in using the Cembureau apparatus was to obtain permeability measurements in a standardized framework, as has already been done for DEF damaged concrete [11, 12]. However, the Cembureau test has an important limitation for permeability measurements on cracked concrete: in order to obtain a reliable measurement of air flow, a confinement pressure is applied to ensure the tightness of the specimens on the lateral face. This confinement pressure of 0.8 MPa can lead to a certain re-closure of the cracks and reduce the real permeability of the concrete. The cracks induced by the development of ISR are diffuse, multidirectional, and result in significant relative displacements of the crack lips. In addition, they can be partially filled by the new products due to chemical reactions. Such cracks are difficult to close by a small radial load (less than 1 MPa confinement pressure for permeability measurement), as shown by the results of compression tests performed after expansion [13]. In the present experiments, the effect of the external confinement pressure on the Cembureau test measurement was evaluated, but only for the driest saturation state after drying at 105 °C. For confinement pressures between 0.5 and 0.9 MPa, the reduction in flow obtained during the tests was about 12% and 20%, respectively, on the specimens developing ASR (whatever their expansion) and DEF (for the largest expansions). However, at this level of saturation, the ASR products were probably dehydrated by the drying process and no longer played their role in reducing crack re-closure, and a smaller effect can be expected for higher levels of saturation. This point needs to be verified in future measurements. Therefore, the values presented in this paper should be considered as a minimum value of permeability increase. The real permeability increase may be higher. To avoid this problem, a permeability measurement device without confinement pressure is under development.

3 Experimental Results

3.1 ASR and DEF Expansion

Figure 1 shows the deformation of the three concretes after immersion in water at 38 °C.

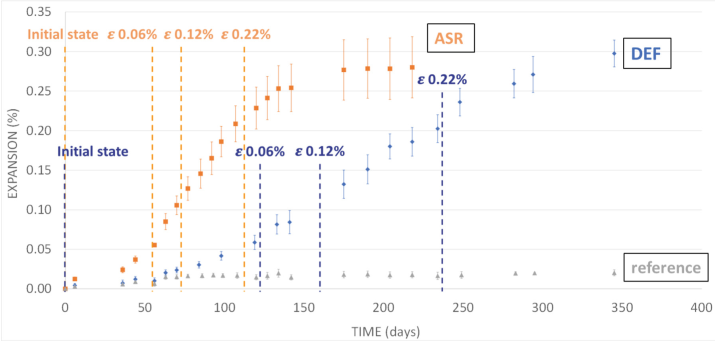

Fig. 1. Deformations of concrete as a function of time

The reference concrete showed only slight water-induced swelling (water absorption between endogenous curing state and water immersion).

After 36 days of immersion in water at 38°C, the deformation rate of the ASR-affected concrete changed from the latent period (small expansion, comparable to water absorption) to the acceleration phase (significant expansion). This acceleration phase lasted about 100 days before entering in the stabilization period. The maximum expansion achieved was about 0.27%. The characteristic expansion values used as expansion targets for permeability tests are given in Table 6.

In the case of the concrete affected by DEF, the expansion was still in the acceleration phase after 345 days. The end of the latent period is after approximately 65 days, which is longer than that observed for ASR. Table 6 groups the dates corresponding to the expansions for performing permeability measurements. The durations for DEF-affected concrete were almost twice as long as those observed for ASR. After 345 days, the DEF-affected concrete still showed significant swelling and was currently at an expansion level of 0.3%.

Table 6. Dates corresponding to the expansions for the permeability measurements

	0,06%	0,12%	0,22%
DEF	122 days	164 days	240 days
ASR	59 days	74 days	117 days

3.2 Permeability of Concrete Damaged by ASR

Air permeability was measured for all samples at six saturation levels (80, 60, 40, 20, 3 and 0%). As an example, Fig. 2 shows the apparent permeability of the concrete developing ASR as a function of the reciprocal of the mean pressure P_m for a degree of saturation of 40%. First, it is important to note that the linearity of the apparent permeability with reciprocal of the pressure (Klinkenberg's law) was not modified by the appearance of the cracks due to ASR.

Similar results were obtained for the other degrees of saturation. The apparent pressure for an inlet pressure of 2 bar is given in the following part for the discussion of the results.

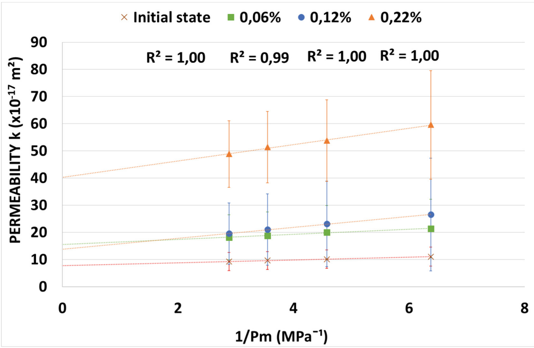

Fig. 2. Permeability measured ASR-affected concrete for a saturation degree of 40%

For this 40% saturation, the apparent permeability shows a significant increase even for small expansion. At 0.06% of expansion, the apparent permeability that of the undamaged concrete (11.1×10^{-17} m^2 at 2 bar inlet pressure). The permeability continued to increase with expansion until it reached 5.4 times the initial permeability for 0.22% of expansion (59.6×10^{-17} m^2). The intrinsic permeability for 0.06% of swelling is slightly higher than that for 0.12%. This can be explained by the increase in the measurement dispersion with the degree of expansion. This increase is due to the appearance of cracks with larger opening in some specimens. Similar behavior was observed for the other saturation degrees but with different levels of permeability modifications as discussed in the following part. The air permeability was strongly influenced by the degree of ASR-expansion and by the induced cracking.

3.3 Permeability of Concrete Damaged by DEF

Figure 3 presents the air permeability of concrete samples damaged by DEF for the 40% saturation degree as for the ASR, the air permeability of the concrete was directly related to the degree of expansion. For the same degree of saturation, the permeability of the concrete increases with the variation of the expansions. At 40% of saturation, the permeability measured in the initial state was also multiplied by 5.4 times as for ASR to go from a value of (1.3×10^{-17} m^2) to a value of (7.1×10^{-17} m^2) for an expansion of 0.22%. Despite the different initial permeability values, the ratio between the initial state and the permeability at 0.22% expansion was the same for ASR and DEF at 40% of saturation. The results for the other saturation rates are presented in the following section.

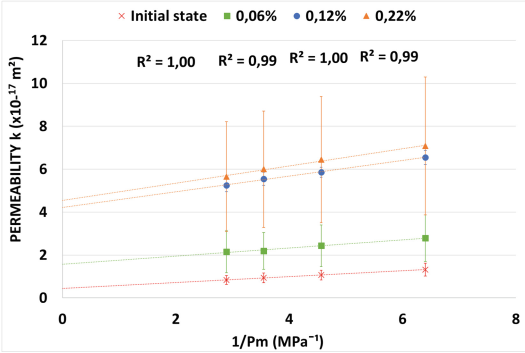

Fig. 3. Permeability measured for the DEF-affected concrete for a saturation rate of 40%.

4 Synthesis and Discussion

4.1 Apparent Permeability of Concrete Damaged by ASR

Figure 4 shows the evolution of the permeability at an inlet pressure of 2 bars (k2bars) for ASR-affected concrete as a function of the degree of saturation for the different levels of swelling.

As for the usual concrete [3], the air permeability increased nonlinearly with the decrease of the saturation level due to the opening of the pore network by the water outflow. When a low saturation level is reached (less than 3%), the strong increase in permeability indicates stronger damage due to preconditioning.

In the case of undamaged concrete, a very low permeability was found at a saturation level to or higher than 60% as usual for such concretes [3] which remain impermeable for high saturation levels. The air permeability evolves little because the liquid phase in concrete porosity is still continuous. The liquid phase becomes discontinuous around 60% of saturation, hence the sharp increase in permeability.

On the contrary, for the damaged concrete, an expansion of only 0.06% was sufficient to reach an apparent permeability of about 12×10^{-17} m^2 at 80% of saturation, similar to the permeability of the undamaged concrete at 40% saturation. This single result indicates the significant effect of ASR cracking on the air permeability of highly saturated concrete. In concrete affected by ISR, the cracking induced by the expansion leads to gas percolation networks that do not exist in concrete before expansion. The effect of expansion on permeability is a function of strain rate. This effect is exacerbated at high saturation rate and decreases as the drying process progresses.

For the 0.22% expansion, the effect is even greater. At this expansion, the decrease in saturation leads to a significant increase in permeability in the damaged concrete. In this case, the shrinkage induced by drying can increase the opening of the cracks induced by ASR and thus leading to the permeability evolution.

From the same results, Fig. 5 shows the evolution of the air permeability of ASR concrete for different levels of expansions according to the degree of saturation.

The increase in air permeability with increasing degree of saturation was almost linear for concrete with high degree of saturation (higher than 60%). The evolution of

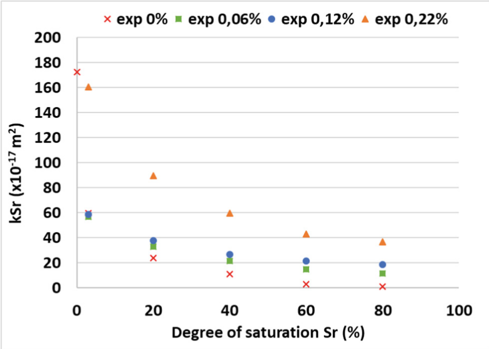

Fig. 4. Evolution of the gas permeability concrete affected by the ASR for different levels of expansion as a function of the degree of saturation

permeability with expansion becomes non-linear for drier concrete. For the degree of saturation lower than 40%, the evolution was still quite linear for expansion levels lower than 0.12%, but nonlinearity appears for the largest expansion studied in the present work (0.22%).

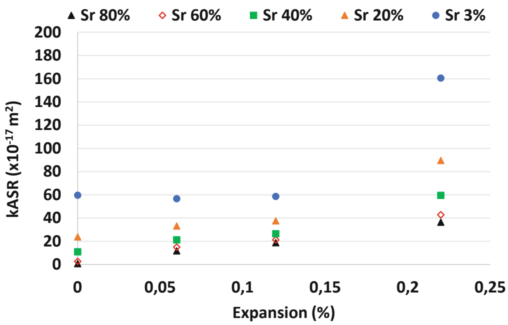

Fig. 5. Gas permeability of ASR affected concrete for different saturation rates as a function of expansions

For the permeability obtained on samples dried at 80 °C, the evolution of the permeability is zero for expansions lower than 0.12% while cracks were always visible on the external surface. This result shows the importance of the saturation degree of saturation to characterize the cracks due to ASR by air permeability measurement. If the degree of saturation is too low, the preconditioning strongly affects the concrete by thermal damage causing bridges between wide cracks. Then, the impact of the preconditioning takes too much importance compared to the effect of cracking induced by ASR, which can no more be quantified.

4.2 Apparent Permeability of Concrete Damaged by DEF

Figure 6 presents the evolution of the permeability at an inlet pressure of 2 bars (k2bars) of the concrete affected by the DEF as a function of the degree of saturation. For the initial concrete without expansion, the air permeability was low even after drying up to 40% of saturation. Due to the different nature of the aggregates, this concrete has a lower permeability than the ASR susceptible concrete prior to initial expansion.

For DEF-damaged samples, the permeability measured at all levels of expansion for a saturation degree higher than or equal to 40% remains lower than 10×10^{-17} m^2. The permeability measured for all DEF damaged samples with the different levels of expansion remains significantly lower than that of the concrete developing ASR.

However, even when permeability was low, DEF also affected this property:

- At 40% saturation, air permeability at 0.12% expansion was 5 times greater than at baseline.
- At 20%, the air permeability increased 1.5 times from the initial state to 0.06% and 3 times from the initial state to 0.12%.

As observed for the concrete affected by ASR, the permeability of the concretes developing DEF shows an increase with the level of expansion (Fig. 7).

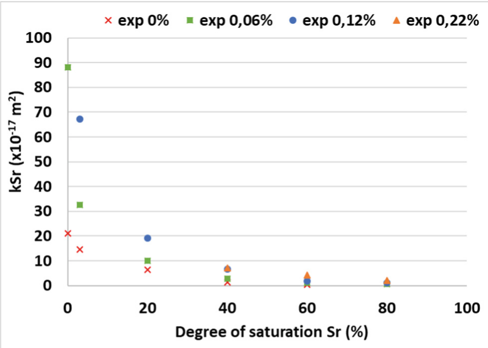

Fig. 6. Gas permeability of DEF affected concrete for different levels of expansion as a function of the degree of saturation

At high levels of saturation, the effect appears to be small. However, for this concrete without expansion, the permeability was completely zero at 80% of saturation. At 0.06%, the permeability was still small but no longer zero (0.6×10^{-17} m^2). The permeability continued to increase with the expansion, reaching 2×10^{-17} m^2 at 0.22% expansion. Although this value was smaller than for ASR, it was a significant variation. This effect is more visible as the degree of saturation decreases: at 40%, the permeability of concrete with expansion of 0.12 and 0.22% expansion is six times greater than that of concrete without expansion. As soon as the drying led to a degree of saturation lower than or equal to 20%, the evolution of the air permeability with the increase of the expansion became more important.

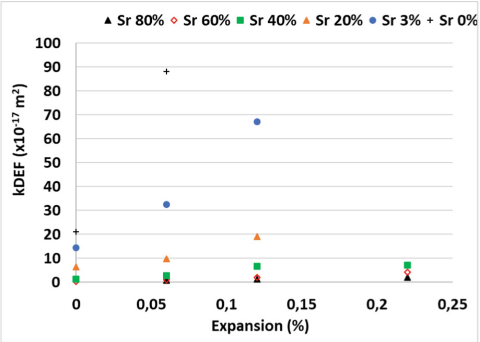

Fig. 7. Gas permeability of DEF affected concrete for different saturation rates as a function of expansions

4.3 Discussion

The permeability of the two concretes before expansion showed large differences. The permeability was about 11×10^{-17} m^2 for the concrete before ASR expansion and about 1.3×10^{-17} m^2 for the concrete before DEF expansion, for an inlet pressure of 2 bar and a saturation degree of 40%. This difference can be explained by the nature of the aggregate which modifies the transport properties of the interfacial transition zone (ITZ). Despite this difference, the impact of ASR and DEF on expansion of concrete with low saturation degrees seems quite similar. In fact, expansions of 0.06% resulted respectively to an increase of about 2 and 1.5 for a saturation degree of 40 and 20% whatever the nature of the expansion (ASR or DEF). At 60% saturation, the effect of ASR was slightly greater: the permeability at 0.06% expansion is 5.4 times greater than the initial state, while it is 3.8 times greater for DEF. For a very high degree of saturation (equal to 80%), the effect was even greater. For ASR, the permeability was 15 times greater at 0.06% expansion and 45 times greater at 0.22% expansion. For DEF, the increase cannot be quantified because the permeability was zero in the undamaged concrete. However, the increase was significant since the permeability was no longer zero at even the smallest strain. These results highlight the strong relationship between the degree of saturation and the cracking induced by ISR on the evolution of the air permeability of damaged concrete.

The physico-chemical mechanisms at the origins of the ASR and DEF are significantly different: formation of alkali-silica gels leading to cracks in aggregates then propagating in cement paste for ASR and formation of delayed ettringite in the cement paste for DEF leading to cracks in the cement paste and at the interfacial transition zone (ITZ). Expansion also leads to different types of induced cracking: microcracking usually not visible to the naked eye due to its small opening for expansion less than 0.2% for DEF and microcracking partially visible, especially for the most opened cracks, for ASR-expansion greater than 0.1%, as observed during this experimental program. Once the cracks due to ISR crossed the sample, the permeability was significantly affected with a strong increase in permeability. This explains the non-linear increase in permeability with increasing expansion. For DEF, the permeability also depends on the formation of ettringite in the cracks created. Beyond a certain drying temperature, and to reach the reference saturation rate of 0%, ettringite is partially destroyed and leaving new percolation paths with larger openings. For ASR, the gels volume is smaller and the cracks are only partially filled. The effect on permeability is not as significant.

5 Conclusion

The safety requirements for reinforced concrete structures are not limited to the mechanical stability of these structures under various stresses. In fact, the airtightness of civil engineering structures is an essential and often requested additional requirement to ensure the maintenance of containment in case of a mechanical accident or chemical attack. This experimental program is a first attempt to quantify the effect of ASR and DEF on the air permeability of concrete under saturation conditions representative of real massive structures. The effect of cracking on air permeability has been well studied in the literature, but few of these studies have evaluated the effect of the saturation degree on this effect. The permeability of concrete damaged by ASR and DEF evolves nonlinearly with the level of expansion and the saturation rate of the sample. Thus, the present work shows how the saturation degree can amplify the effect of cracking on the increase in permeability. Concrete at 80% of saturation is usually impermeable because its pore network is closed by water. However, the same concrete can become permeable when damaged by ASR or DEF, even at low expansion because the induced cracks with an opening larger than 1 μm are emptied by water even at very high saturations. Next, the results presented in this paper will be analyzed in regard to the evolution of mechanical properties to obtain correlation useful for improving the modelling of damaged structures.

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