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Influence of cracking on the diffusion properties of cement-based materials
Part I: Influence of continuous cracks on the steady-state regime

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Abstract

The influence of traversing cracks on the steady-state diffusion properties of concrete was studied. The effect of both anisotropic and isotropic crack networks was first theoretically assessed using an analytical approach. To simplify the transport equations, cracks were assumed to be of uniform size and evenly distributed on a one- or two-dimensional grid. Results of the theoretical analysis were then compared to experimental data. Both series of results indicate that cracking can markedly alter the diffusion properties of the material and favor the penetration (or the leaching) of drifting species. A simple method to predict the effect of cracking on the concrete diffusivity is proposed. Predictions are made on the basis of two parameters: the crack density and the mean crack aperture. This method can provide a first estimate of the diffusion properties of severely damaged concrete elements.

Keywords: Cracks; Diffusion; Chloride; Modeling

1. Introduction

Under natural exposure conditions, the durability of concrete is often controlled by its ability to impede ion and fluid transport. Within its service life, the transport properties of concrete are most likely to be modified. For instance, continuous hydration may contribute to improve the tightness of the material. On the other hand, reinforced and plain concrete structures are often subjected to various solicitations (thermal or moisture gradients, mechanical loads). In many cases, these solicitations tend to exceed locally the tensile strength of the material and generate cracking that may subsequently affect the transport properties of the solid.

In recent years, the effect of cracking on the permeability and the absorptivity of both loaded and unloaded concrete elements has been the subject of numerous investigations [1–4]. All of these studies have clearly indicated that the presence of cracks could contribute to increase the permeation (or absorption) coefficient of the material by several orders of magnitude. It has also been shown that parameters, such as the continuity of the crack network and the mean aperture of cracks, have a marked influence on the material permeability [3]. According to these results, any model aiming at predicting the permeability of concrete should account for local stress and strain effects since the crack aperture grows with an increase of the applied load [3].

Over the past decade, the mechanisms of molecular and ionic diffusion in cement-based materials have also been extensively studied [5,6]. Contrary to the research effort on permeability, it appears that most researchers working on the phenomenon of diffusion have curiously chosen to overlook the issue of cracking. An extensive survey of the literature has shown that the number of investigations specifically dedicated to the influence of cracking on the diffusion properties of concrete is extremely limited [5]. Furthermore, the few reports published on the subject are more or less contradictory. While in certain cases cracking has been found to affect the diffusivity of concrete [7,8], there are experimental and theoretical evidences showing that the presence of cracks has little influence on that property [4,9].

The present investigation was performed to provide more information on the influence of cracking on the diffusion properties of concrete. This study constitutes the initial phase of a more comprehensive project devoted to the transport properties of cement-based materials [10]. In this initial part of the project, we have deliberately chosen to tackle the

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problem of cracking using a theoretical approach and assuming simple crack patterns. To limit the scope of this work, only the simple case of traversing cracks (i.e., going through the entire thickness of the concrete) on the steady-state regime was considered. Predictions yielded by the models were compared to chloride (accelerated diffusion) migration test results previously obtained in our laboratory [11].

In addition, to simplify the mathematical treatment of the problem the consideration of connected crack networks allows comparison of the influence of these defects on the diffusion process to their effects on the permeability of cement-based systems. Most studies of the effect of cracks on the permeability of concrete have been carried out on continuous crack systems.

It should be emphasized that the case of evenly distributed traversing cracks of uniform size is obviously not representative of what is usually observed in service. In practice, continuous crack networks are mostly limited to heavily degraded concrete elements. Furthermore, numerous studies have clearly established that cracks are usually tortuous and not evenly distributed throughout the material [12–14]. In that respect, the development of continuous crack network can be considered as a worst-case scenario, and the results predicted by the analytical model should yield upper-bound values for the diffusivity of cracked concrete elements. The influence of discontinuous crack networks has been investigated by finite-element modeling. The results of this analysis will be discussed in forthcoming publications.

2. Theoretical background

2.1. Crack network characteristics

From the standpoint of transport, a piece of cracked concrete can be assimilated to a composite material in which the crack network is superimposed to a homogeneous reference material. A similar approach has been used by numerous authors to investigate the influence of cracking on mass transport processes (i.e., absorption, permeation, molecular diffusion, etc.) in various porous media [4,15–17]. To account for the various types of cracking that can be found in practice, two different cases were considered in this study (see Fig. 1):

- an isotropic (two-dimensional) crack network, and
- an anisotropic (one-dimensional) crack network.

The one-dimensional case corresponds to a piece of concrete in which cracking was initiated by mechanical loading. In that case, the crack network is anisotropic (i.e., all the traversing cracks are oriented in the same direction) [18]. Since it had been shown that cracks induced by freezing and thawing or by drying shrinkage generally have no special orientation [12,19], a second case of anisotropic (two-dimensional) cracking was also considered.

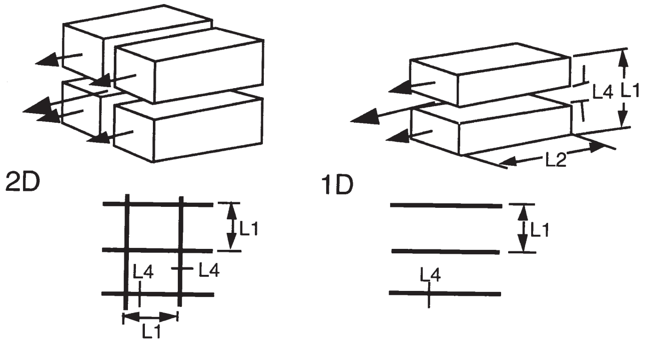

Fig. 1. Schematic representations of the two crack patterns considered in this study: (a) isotropic (2-D) cracking; (b) anisotropic (1-D) cracking.

As shown in Fig. 1, the two simple cases of crack patterns can be characterized on the basis of two parameters, namely:

- L_4: the mean aperture of the cracks, and
- L_1: the mean crack spacing.

It has been shown that the latter parameter (L_1) can be obtained simply from the determination of the crack density (expressed in m^{-1}) using an image analysis system coupled to an scanning electron microscope or to an optical microscope [12,19]. The mean crack aperture can also be estimated from image analysis measurements. However, the reliability of the results is directly linked to the type of sample preparation technique used prior to the measurements [12].

On the basis of these two parameters, one can calculate the crack spacing factor (f) as shown in Eq. (1):

$$f = \frac{L_1}{L_4} = \text{crack spacing factor} \tag{1}$$

For the sake of simplification, we have limited our analysis to a unidimensional case where a concrete sample of thickness L_2 is placed in a diffusion cell. The molecular (or ionic) concentration of the upstream compartment is kept constant throughout the experiment. Furthermore, it is believed that the volume of the downstream compartment is so large that any variation of its concentration due to the transport of particles through the sample can be neglected.

To complete the analysis of the problem, one has to define a few additional parameters:

- L_2 = the thickness of the sample under study;
- D_1 = the diffusion coefficient of a given ionic (or molecular) species in free solution;
- D_0 = the diffusion coefficient of the species in the homogeneous (uncracked) material;
- D = the apparent diffusion coefficient of the cracked material; and
- D/D_0 = the equivalent diffusivity of the material.

For most molecular (or ionic) species, D_1 (i.e., the diffusion coefficient of the species in an ideal solution) is in the order of 10^{-9} m^2/s. D_0 can be determined by means of simple diffusion experiments. It can also be estimated on the basis of migration test results [5,11].

2.2. Assumptions regarding the transport process

As emphasized in many recent publications, the mechanisms of molecular and ionic drift in cement-based materials are extremely intricate [5,6]. For instance, it has been established that ions and molecules can strongly interact with the solid in certain cases. Such an interaction tends to affect markedly the mechanisms of transport during the unsteady-state regime since only free particles are participating to the diffusion process. The phenomena of ionic diffusion and migration are further complicated by the fact that ions are charged particles. The electrical charge of the ions will affect their movement in solution. In concentrated solutions, for instance, ions will tend to interact will the solvent (this interaction is termed the "electrophoretic effect" by electrochemists) and with other ions (the latter interaction is called the "relaxation effect").

All of the interactions mentioned above have been found to have a very significant influence on the diffusion and migration processes [5]. Any reliable prediction of ionic flows should therefore account for these phenomena. However, to simplify the mathematical treatment of the problem and be able to focus on the influence of cracking on the overall process, all the equations presented in the following paragraphs were derived on the basis of the following assumptions:

- The drifting particles have no physical nor chemical interaction with the solid.
- The electrophorectic and relaxation effects can be neglected.
- The transport process is taking place under isothermal conditions.

Interaction phenomena can be neglected in the analysis of steady-state transport cases. When the steady-state regime is reached, most interaction phenomena are completed and flux conditions are stabilized. In most practical cases, the second and third assumptions are obviously not verified. It is believed, however, that these simplifications should not adversely affect the conclusions derived from our analysis, at least not from a qualitative point of view.

2.3. Influence of cracking on the steady-state regime

The main advantage of steady-state problems is that they can be treated in terms of simple flow equations. For unidimensional problems, the flow of drifting particles (F_{tot}; often expressed in mol/s) is equal to the flow of particles diffusing through the cracks (F_c) plus that diffusing through the homogeneous material (F_m), as shown in Eq. (2):

$$F_{\text{tot}} = F_c + F_m \tag{2}$$

Under steady-state conditions, the flux of particles (J_{tot}; expressed in mole/m^{2}s) going through the sample can be averaged over the entire surface of the concrete, as depicted in Eq. (3):

$$J_{\text{tot}} = \frac{J_c A_c + J_m A_m}{A_c + A_m} \tag{3}$$

where A_c and A_m (expressed in m^2) stand for the total area (perpendicular to the diffusion flux) of cracks and (uncracked) material respectively; J_c (mole/m^{2}s) the flux of ions diffusing through cracks; and J_m (mole/m^{2}s) the flux of ions going through the uncracked material.

In any diffusion (or migration) problem, the flux of particles can always be expressed as the product of the driving force (F) times the transport coefficient. In a migration test, one can write Eq. (4):

$$J_c = -D_1 F \text{ (a)} \quad \text{and} \quad J_m = -D_0 F \text{ (b)} \tag{4}$$

Knowing that [see Eq. (5)]:

$$J_{\text{tot}} = -DF \tag{5}$$

By replacing Eqs. (4a), (4b), and 5 in Eq. (3), one easily finds that [see Eq. (6)]:

$$D = \frac{A_c D_1 + A_m D_0}{A_c + A_m} \tag{6}$$

Eq. (6) can be rearranged in such a way that [see Eq. (7)]:

$$\frac{D}{D_0}(A_c + A_m) = \frac{D_1}{D_0} A_c + A_m \tag{7}$$

Noting that [see Eq. (8):

$$S = \frac{A_m}{A_c} \tag{8}$$

One can write [see Eq. (9)]:

$$A_m = A_c S \text{ (a)} \qquad A_m + A_c = A_c(S + 1) \text{ (b)} \tag{9}$$

By replacing Eqs. (9a) and (9b) in Eq. (7), one finds [see Eq. (10)]:

$$\frac{1}{n} = \frac{D}{D_0} = \frac{\frac{D_1}{D_0} + S}{1 + S} \tag{10}$$

As can be seen, on the basis of this simple analysis one can easily link the diffusivity of a cracked material to the characteristics of its crack network. The application of Eq. (10) to the two crack patterns will be discussed next.

3. Experimental procedures

In a previous series of tests carried out in our laboratory, the influence of frost-induced internal microcracking on the migration properties of one non-air-entrained concrete mixture was studied. The results of this study were recently reported [11]. The 0.45 water/cement ratio concrete was prepared at a cement content of 433 kg/m^3 with natural granitic sand and a granitic crushed aggregate. More information on the characteristics of the concrete mixture can be found in the previously published work [11].

Cylinders were moist cured for several weeks, then subjected to freezing and thawing cycles immersed in water according to the requirements of ASTM C 666—Procedure B. During the freezing and thawing cycles, the weight (in air and water) and the ultrasonic pulse velocity of the cylinders were measured regularly.

Samples were taken out of the freezing apparatus after 31, 61, and 95 cycles. These samples were then sawed and tested for chloride migration. Discs of 95-mm in diameter and 15-mm thick were used for the migration experiments. All discs were immersed in deionized water and vacuum saturated for 24 h prior to testing. The discs were then mounted on the migration cells. Both compartments of the cells were equipped with an agitating device to maintain the homogeneity of the solutions, and ruthenium-coated titanium electrodes were used. The upstream compartment of each cell was filled with a 12 g/L (0.3 M) sodium hydroxide solution containing 30 g/L of sodium chloride (0.5 M). The downstream compartment of each cell was filled with a 12 g/L (0.3 M) sodium hydroxide solution. A voltage of 10 V was applied on each cell, and the temperature of the cells was kept constant at 21°C throughout the entire duration of the tests. The chloride concentration of the solution in the downstream compartment was determined at regular intervals by means of potentiometric titration using a silver nitrate electrode. Two duplicate specimens were tested.

At the end of the migration experiments, the characteristics of the crack network of each sample were measured using an optical microscope. Prior to the observations, the samples were impregnated with a red dye and slightly polished under water. More information on the testing procedures can be found in the previously published work [11].

4. Results and discussion

4.1. Anisotropic (one-dimensional) cracking

For an anisotropic cracking pattern, one finds that [see Eq. (11)]:

$$S = \frac{L_1 - L_4}{L_4} = f - 1 \qquad (11)$$

Introducing the value of S in Eq. (10) yields Eq. (12):

$$\frac{1}{n} = \frac{D}{D_0} = \frac{\frac{D_1}{D_0} + (f - 1)}{f} \qquad (12)$$

If $L_1 >> L_4$, then $f >> 1$ and Eq. (12) can be simplified in the following way [see Eq. (13)]:

$$\frac{1}{n} = \frac{D}{D_0} = \frac{D_1}{D_0 f} + 1 \qquad (13)$$

The variation of the equivalent diffusivity of the material ($1/n$) as a function of D_1/D_0 and the mean crack spacing factor (f) is illustrated in Fig. 2. As can be seen in the figure, the presence of traversing cracks tends to markedly modify the transport coefficient of the solid.

It should be emphasized that the mean crack spacing factor (i.e., the parameter f) rarely goes below 100, even for concrete samples severely deteriorated. This point will be discussed further in the following section. Hence, in most practical cases, the increase in diffusivity is limited to a fac-

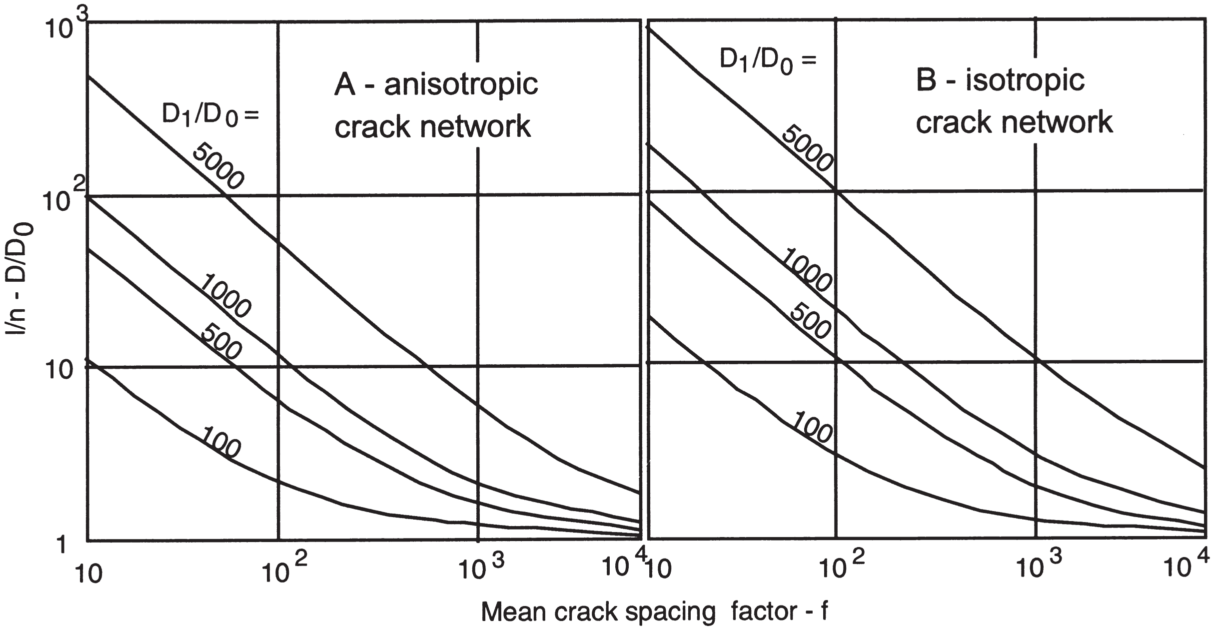

Fig. 2. Variation of the equivalent diffusivity of the material ($1/n$) as a function of D_1/D_0 and f (anisotropic and isotropic crack networks).

tor of 10. In that respect, the influence of the crack network on the material diffusivity appears to be much less significant than its effect on the solid permeability. In the case of permeability, the presence of cracks can easily increase the permeation factor by several orders of magnitude.

The curves of Fig. 2 also indicate that the influence of cracking tends to be more significant when the ratio D_1/D_0 is increased. This essentially means that the effect of cracking on the solid equivalent diffusivity will be relatively more important for dense materials (such as high-performance concrete) for which the initial (uncracked) transport coefficient is reduced. This is in good agreement with the results recently reported by Aldea et al. [20].

Finally, it should be underlined that if cracks do not markedly affect the "equivalent" diffusion coefficient of concrete, their presence may significantly alter the material durability. Cracks favor locally the penetration and/or the leaching of ions. Such an effect may in some cases be sufficient to trigger deleterious reactions that might lead to the ruin of the material [21,22]. It may also contribute to acceleration of the local dissolution of solid phases that may subsequently affect the material mechanical behavior [21].

4.2. Isotropic (2-D) cracking pattern

For an isotropic cracking pattern, one finds [see Eq. (14)]:

$$A_c = L_1^2 - (L_1 - L_4)^2 = L_1^2\left[1 - \left(\frac{f-1}{f}\right)^2\right] \tag{14}$$

and [see Eq. (15)]:

$$A_m = L_1^2\left(\frac{f-1}{f}\right)^2 \tag{15}$$

Replacing Eqs. (14) and (15) in Eq. (8), one finds [see Eq. (16)]:

$$\frac{A_m}{A_c} = S = \frac{L_1^2\left(\frac{f-1}{f}\right)^2}{L_1^2\left[1 - \left(\frac{f-1}{f}\right)^2\right]} = \frac{(f-1)^2}{2f+1} \tag{16}$$

If $f >> 1$, then, according to Eq. (16) [see Eq. (17)]:

$$S = \frac{f}{2} \tag{17}$$

Introducing Eq. (17) into Eq. (8), one can easily find that [see Eq. (18)]:

$$\frac{1}{n} = \frac{2}{f}\frac{D_1}{D_0} + 1 \tag{18}$$

The variation of the equivalent diffusivity of the material ($1/n$) as a function of D_1/D_0 and the mean crack spacing factor (f) is also shown in Fig. 2. As can be seen, the presence of an isotropic crack network has, more or less, the same effect on the material equivalent diffusivity as that of an anisotropic network. The influence of the isotropic network, however, appears to be slightly more important.

4.3. Influence of cracking on the solid volume

In most cases, the development of cracks creates an additional porosity that increases the volume of the material. This is the case for a concrete sample suffering from frost-induced microcracking. One the easiest ways of assessing the presence of cracks is to measure any change in the volume of the material. The measured value can then be used to estimate the mean crack spacing factor (the parameter f).

In the case of the development of an isotropic crack network, one can initially assume that all the cracks are geometrically oriented in the same direction as the diffusion flow. Then, one finds [see Eq. (19)]:

$$\frac{\Delta V}{V_0} = \frac{A_c}{A_m} = \frac{f^2}{(f-1)^2} - 1 \tag{19}$$

where V_0 stands for the initial volume of the uncracked solid. If the cracks do not have any preferential orientation, the crack network will tend to divide the solid into small cubes. Eq. (16) can then be modified to become Eq. (20):

$$\frac{\Delta V}{V_0} = \frac{A_c}{A_m} = \frac{f^3}{(f-1)^3} - 1 \tag{20}$$

The evolution of the solid volume change with the mean crack spacing factor is illustrated in Fig. 3 where both assumptions have been considered.

5. Validation of the theoretical analysis on the basis of experimental results

The migration coefficients of the cracked samples have been compared to that of virgin (i.e., uncracked) samples. The main results of the study are summarized in Table 1. The crack aperture (L_4) considered in the calculations is a mean value estimated on the basis of the optical microscope observations. The aperture of one single crack may vary quite significantly from one location to another. The mean crack aperture values used in the calculation should therefore be considered as rough estimates of the reality.

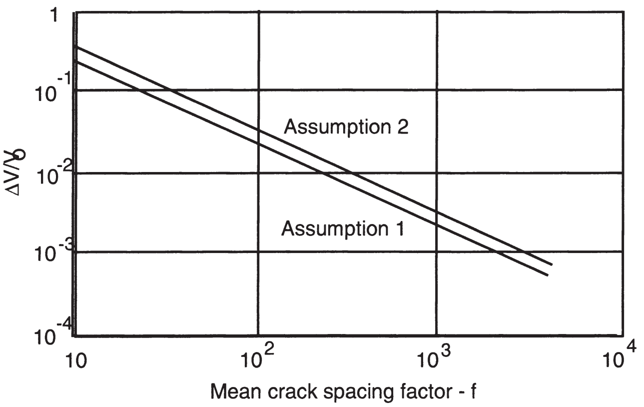

Fig. 3. Evolution of the solid volume change ($\Delta V/V_0$) with the crack spacing factor (f).

Table 1
Influence of frost-induced microcracking on chloride by migration (comparison between predicted values and experimental data)

Number of cycles	$\Delta V/V_0$[a] (%)	f	Crack aperture (μm)[b]	Diffusion coefficient[c] ($\times 10^{-9}$ cm^2/s)	$1/n = D/D_0$ Predicted	$1/n = D/D_0$ Experiment
0	–	–	–	9.70	–	–
31	0.7	440	5	24.48	5.6	2.5
61	1.7	180	9	41.64	12.0	4.3
95	2.9	105	12	76.52	20.0	7.9

[a] Established on the basis of weight measurements (air and water).
[b] Crack aperture = L_4.
[c] Established on the basis of migration test results.

As can be seen in Table 1, the mean crack spacing values calculated according to Eq. 20 (i.e., on the basis of volume change measurements) do not go under 100 even for the very degraded non-air-entrained concrete that had been subjected to 95 freezing and thawing cycles. It should be emphasized that the deterioration of that sample was quite severe and had contributed to reduce the compressive strength of the material from half of its initial value (see the previously published work [11] for more information).

The results of Table 1 also indicate that the analytical method systematically overestimates the transport coefficient values by a factor of approximately 2.5. Considering the assumptions at the basis of the analytical development and the fact that the determination of the volume change and crack aperture values are necessarily plagued by an experimental error, the precision of the model is quite acceptable.

It should also be kept in mind that one of the main assumptions at the basis of the development of the analytical model is that the cracks developed in the material are straight and connected. The intrinsic tortuosity of the crack network is not accounted for in the model. In most practical cases, the aperture along the path of one single crack can vary quite significantly.

A previous study [3] on the effect of traversing cracks on the permeability of cement systems have shown that the effective crack aperture (L_4) can be estimated on the basis of Eq. (21):

$$L_4 = \frac{1}{\tau} w \tag{21}$$

Where w is the apparent crack width of the network and τ is the tortuosity of the crack network. In most practical cases, the value of the tortuosity parameter (τ) ranges between 2 and 5. In that respect, it is interesting to note that if the presence of continuous cracks tends to have a much greater influence on the phenomenon of permeation than on the diffusion process, the tortuosity of the system appears to have more or less the same effect.

6. Conclusions

- An analytical method was developed to account for the influence of traversing cracks on the transport of ions in saturated concrete. When compared to experimental values, the model appears to yield reasonable predictions.
- The presence of continuous cracks tends to markedly modify the transport coefficient of the solid. Diffusivity of the material can be increased by a factor ranging from 2 to 10.
- Results obtained with the model indicate that the influence of cracking tends to be more significant when the ratio D_1/D_0 is increased. This essentially means that the effect of cracking is relatively more important for dense materials.

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