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Transport of fluids in cement–rubber composites

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Abstract

The transport properties of building materials exposed to an aggressive environment represent the essential parameters affecting their durability. Furthermore, the deterioration process of materials is highly related to the movement of water contained in a porous volume, whether in liquid or vapour form, and to the air permeability. The present study has been undertaken to examine the influence of rubber aggregates on the durability factors of cement–rubber composites. Results reveal the importance of the cellular character of rubber aggregates with respect to the composite's behaviour in contact with fluids.

Keywords: Composite; Cellular and compact rubber aggregates; Durability; Sorptivity; Hydraulic diffusivity; Air permeability

1. Introduction

Building materials are submitted to different types of loadings: mechanical, chemical and radiative. These loadings can create degradations as a function of their intensity and possible synergies. In almost all cases, material degradation implies, in addition to other factors, a fluid transport mechanism. It is important therefore to ascertain the ease with which fluids are going to penetrate the given material.

In order to evaluate the hydral behaviour of a material when placed in contact with water, sorptivity and hydraulic diffusivity are generally determined by use of the capillary water absorption test. The first of these two parameters translates the capacity of a material to absorb water by means of capillarity. This technique was introduced by Philip [1–4], whose terminology has been adopted by various researchers [5–12] in a number of works concerning porous building materials. Several authors have since applied this characteristic to analyse concrete quality [13,14].

Hydraulic diffusivity, on the other hand, indicates the ease of humidity transfer in liquid and vapour form and is consequently used to measure aggressive agent diffusion.

The air permeability is also important because air transports carbon dioxide and sometimes other gaseous discharges which react with cement paste components.

As regards rubber waste reuse, various techniques have been proposed. The International Symposium recently organised in Dundee [15] focused on current techniques for recycling and reusing worn tyres. Other work on the topic has also been carried out [16–18]. Nevertheless, these efforts have concentrated on the reuse of worn tyres, whereas other types of rubber waste (e.g. hoses, seals, windscreen wipers) generated from the transportation industry and household electrical appliances also exist in large quantities. These wastes may stem from either the manufacturing process or worn systems.

Previous research has allowed highlighting the value of the cellular character of rubber aggregates with respect to material deformability properties and freeze/thaw cycle behaviour [19]. The present work is intended to assess the impact of such additives on both water and air penetration, with the aim of predicting the durability of cement–rubber composites. A study of capillary water absorption and air permeability has therefore been carried out on compact and cellular rubber aggregate composites with different formulations.

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2. Materials and experimental testing

The rubber aggregates used in this study are derived from automotive industry waste, from a combination of sources, except tyres. Reduced and sieved to 1–4 mm in size, they differs from mineral aggregates in terms of the strain magnitude at ultimate tensile strength (100–200%), the non-breaking characteristic under loading, impact resisting and energy absorption properties. The rubber aggregates used have been classified into two types:

- compact rubber aggregates (CRA), and
- expanded rubber aggregates (cellular) (ERA).

The physico-mechanical properties and shape of rubber particles are shown in Fig. 1 and Table 1.

Portland cement CPA CEM II 32.5 and rubber aggregates were mixed (water–cement ratio of 0.3) in a standard mixing machine in accordance with European Standard EN 196-1. Total mixture water has been adjusted, depending on the type of rubber, so as to achieve a constant workability (i.e. a slump in the order of 9–10 cm). The volume ratio of rubber aggregates ranged from 0% to 40%.

The entrained air was first measured using the pressure method in accordance with European Standard EN 413-2. In order to ascertain the amount of air associated with the rubber particles only, a predetermined quantity of rubber on the basis of the proportion of a 30% cement–rubber mixture was placed in the air pot. The air pot was then filled with water and the mix was stirred to release any trapped air. The meter indicated an air content of between 1% and 2.5% for CRA type and of between 3% and 4% for ERA type. Because it is unlikely that the rubber would be compressed significantly by the pressure meter [20], the air may be adsorbed on the rubber particles or perhaps air gets trapped in the cellular texture of the rubber particles. In order to have a better understanding of the air content of cement–rubber mixtures, the porosity of mixtures at different volume ratio of rubber aggregates was investigated by measuring the bulk density and the absolute density using a pycnometer method. The voids ratio can be calculated from the expression:

Table 1
Properties of rubber aggregate particles

Rubber aggregates type	Unit weight (kg/m^3)	Water accessible porosity (%)	Hardness (shores)	Modulus of elasticity (MPa)
CRA	1286	0.3	85	68
ERA	1040	3	35	12

$$\text{voids ratio} = 1 - \frac{\text{bulk density}}{\text{absolute density}}$$

Prismatic specimen (40 mm × 40 mm × 160 mm) of compact rubber aggregate composites (CRAC) and expanded rubber aggregate composites (ERAC) were prepared and moist-cured for 28 days at 20 °C and 98% relative humidity. Before testing, the specimens were dried in a drying oven at 40 °C until constant mass. The physico-mechanical properties of the composites are summarized in Table 2.

2.1. Capillary water absorption

The capillary water absorption was measured by placing the sample in contact with water at a depth of approximately 5 mm and maintained at constant level, according to the device shown in Fig. 2. In order to restrict diffusion to a one-dimensional direction, the sides of the previously dried composite were sealed hermetically with a plastic film. The sorptivity S was determined by measuring i the absorbed water volume per unit area of contact as a function of the square root of time. S is calculated from the gradient of the initial linear region of the curve described by Eq. (1) below.

$$i = S\sqrt{t} + i_0 \tag{1}$$

where i (mm^3/mm^2) is the capillary absorption. It has a depth dimension and corresponds, according to Hall [5], to the cumulative water volume absorbed per absorbent

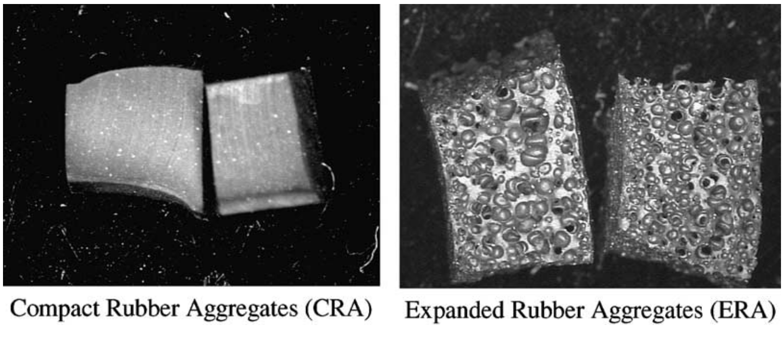

Compact Rubber Aggregates (CRA) Expanded Rubber Aggregates (ERA)

Fig. 1. Optical photo of typical rubber particles (magnification: ×20).

Table 2
Physico-mechanical properties of cement–rubber composites

Volume ratio of rubber (%)	Dry unit weight (kg/m^3)		Compressive strength (MPa)		Flexural strength (MPa)		Elasticity modulus (MPa)	
	CRAC	ERAC	CRAC	ERAC	CRAC	ERAC	CRAC	ERAC
0	1832.5		82.5		3.38		19749.65	
10	1740.5	1630.0	55.0	42.0	3.52	3.60	18698.74	16655.75
20	1672.0	1477.0	34.0	22.0	3.67	3.94	16440.13	12992.52
30	1578.0	1362.0	19.0	10.0	3.42	3.60	13570.10	10279.16
40	1522.0	1274.0	10.0	6.5	2.85	3.35	10293.55	7302.20

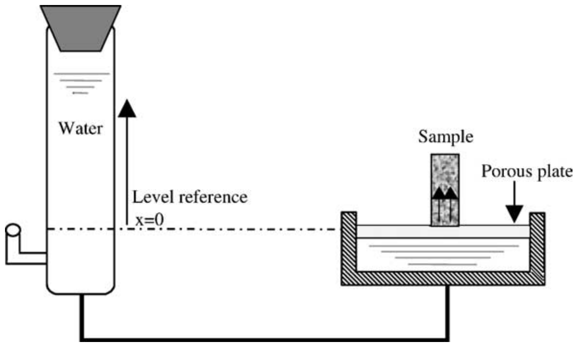


Fig. 2. Schematic diagram of capillary absorption measurement.

unit surface area, S ($mm/s^{1/2}$) is the sorptivity of the material and i_0 (mm^3/mm^2) is an empirical coefficient which depends on the contact of the sample's area surface with water. It corresponds to the instantaneous pore filling when in contact with water.

2.2. Hydraulic diffusivity measurement

The hydraulic diffusivity was estimated by means of a gravimetric technique [21]. According to the case of one-dimensional flow into an unsaturated homogeneous porous medium, the humidity transfer under isothermal conditions is, with respect to the water volume content θ, described by following partial differential equation.

$$\frac{\partial\theta}{\partial x}=\frac{\partial}{\partial x}\left(D_\theta\,\frac{\partial\theta}{\partial x}\right) \tag{2}$$

where t (s) is the time, x (mm) represents the space coordinate and D_θ (m^2/s) is the hydraulic diffusivity coefficient for a water volume content of θ which corresponds to the cumulative water volume absorbed per total volume of the material.

With the following initial and boundary conditions being applied:

$\theta=\theta_s$, for $x=0,\ t\geqslant 0$;

$\theta=\theta_0$, for $x>0,\ t=0$.

The hydraulic diffusivity was estimated by establishing the hydraulic profile that characterizes the change in water content as a function of time at the various levels. By using the Boltzmann transform ($b=x\sqrt{t}$), Eq. (2) is reduced to:

$$-\frac{b}{2}\left(\frac{\partial\theta}{\partial b}\right)=\frac{\mathrm{d}}{\mathrm{d}\theta}\left(D_\theta\,\frac{\mathrm{d}\theta}{\mathrm{d}b}\right) \tag{3}$$

By integration, the hydraulic diffusivity coefficient, at a water content of θ, can be obtained as follows:

$$D_\theta=-\frac{1}{2}\,\frac{1}{\left(\frac{\mathrm{d}\theta}{\mathrm{d}b}\right)}\int_0^\theta b\,\mathrm{d}\theta \tag{4}$$

The slope $(\mathrm{d}\theta/\mathrm{d}b)$ and the area $\int_0^\theta b\,\mathrm{d}\theta$ delineated by the curve $\theta(b)$, are displayed in Fig. 3.

The principle behind the gravimetric technique is to determine the change in the water volume being absorbed by the material as a function of time, for test specimens of various heights all cut from the same specimen. The elements were then sealed laterally so as to ensure a one-dimensional transfer direction, as indicated in Fig. 3. The change of the water volume content in the local volume, which corresponds to the difference between a successive element's height, has been determined with respect to various abscissas of $x=10$, 20, 30, 40 mm, by applying the following formula.

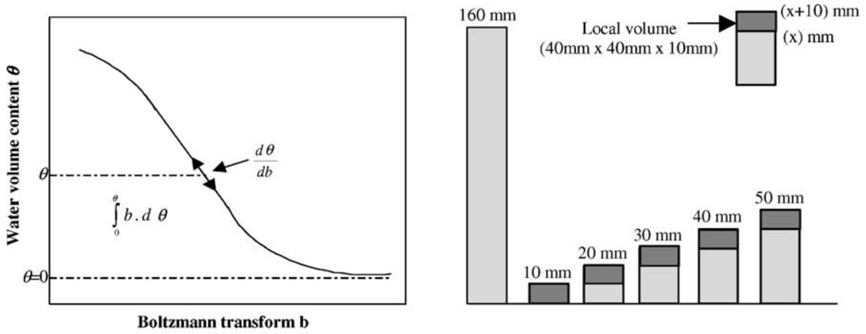
$$\theta(x,t)=\frac{V_\theta(x+10,t)-V_\theta(x,t)}{V_{\mathrm{Local}}} \tag{5}$$

where $V_\theta(x,t)$ is the volume of water absorbed by an element with a height of x (mm) at time t (s), and V_{Local} is the local volume under study.

The typical profile deduced from the hydraulic profile according to the expression (4), was established through deriving the relation between the water volume content $\theta(x,t)$ and the corresponding water volume content $\theta(b=x\sqrt{t})$.

2.3. Air permeability

A permeable device with variable pressure was designed to measure air permeability in cement–rubber composite specimens. This device was then adapted to measure gas and water permeability as well. The test was conducted on 80 mm × 30 mm dry disc samples submitted to atmospheric pressure. Under each specimen, the low air pressure was created by a capillary tube



Curve of the evolution in volumic water content as a function of the Boltzmann transform

Cutting of 160 mm long specimen into several elements

Fig. 3. Experimental determination of composite's hydraulic diffusivity by means of a gravimetric technique.

containing a manometer liquid. The low air pressure was obtained by connecting the permeable set-up to a water pump (see Fig. 4). Opening The stopcock (R) raised the liquid in the capillary tube. When the level (h_0) had been reached, the stopcock was closed and air at atmospheric pressure could pass through the material. The increase in air flow, through the specimen, caused the liquid level to decrease. The air permeability coefficient was then determined by applying Darcy's law. In accordance with simplified theory, in order to eliminate the effect of air volume expansion with a rise in temperature, the air permeability coefficient was calculated by recording the water flow rate as a function of time between two levels (h_0) and (h) from the following expression:

$$K = \frac{\mu s l}{\rho A g t} \ln\left(\frac{h_0}{h}\right) \tag{6}$$

where s (m^2) is the cross-sectional area of the capillary tube, l (m) the specimen thickness, ρ (kg/m^3) the unit weight of water, A (m^2) is the exposed specimen surface area and μ (1.8×10^{-5} N s m^{-2}) the air dynamic viscosity at 20 °C.

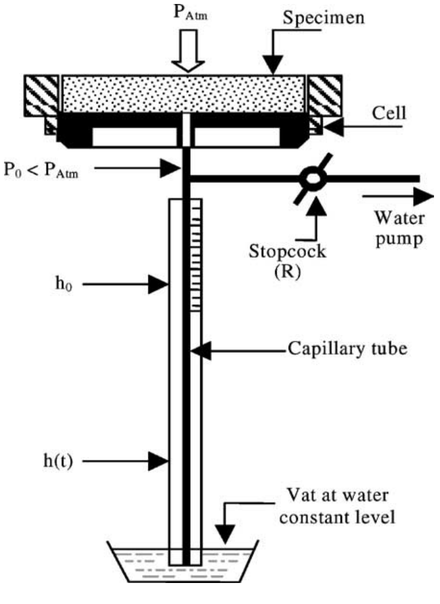


Fig. 4. Permeability test set-up.

3. Results and discussion

3.1. Capillary water absorption (sorptivity) measurement

The change in the composite's water absorption as a function of the square root of time, for different volume contents of both compact and expanded rubber aggregates, is displayed in Fig. 5a. The curves show an initial linear region, (see Fig. 5b), in accordance with the expression $i = S\sqrt{t}$. Table 3 lists sorptivity values for the various samples, obtained from the slopes in the linear region. It should be noted that a mortar (cement/sand 1:3) sorptivity value is 0.108 mm/s$^{1/2}$ for the same cement and the same curing process.

Fig. 6 displays the relative sorptivity as a function of the volume ratio of rubber aggregates. The inclusion of compact and expanded rubber aggregates reduces water sorptivity respectively by approximately: $S/S' = 1 - 1.15\alpha - 0.01\alpha^2$ and $S/S' = 1 - 2.24\alpha + 0.02\alpha^2$ (which yields correlation coefficients of $r = 0.992$ and 0.998), where S, S' are the composite and cement paste sorptivity in mm/s$^{1/2}$ respectively, and α is the volume fraction of rubber aggregates. These relationships are similar to that proposed by Hall et al. [22] in their study on the transport properties of plaster containing sand additives. They attribute this behaviour to the non-sorptive characteristic of aggregates. The absorbed water flow bypasses these particles to propagate within the cement matrix.

At saturation, it can be observed that the addition of rubber decreases water absorption by the cement paste, with this decrease varying from 17% (cement paste) to 12.5% and 8% for a composite containing 40% compact and expanded rubber, respectively. Rubber aggregates which are non-sorptive additives serve to decrease the

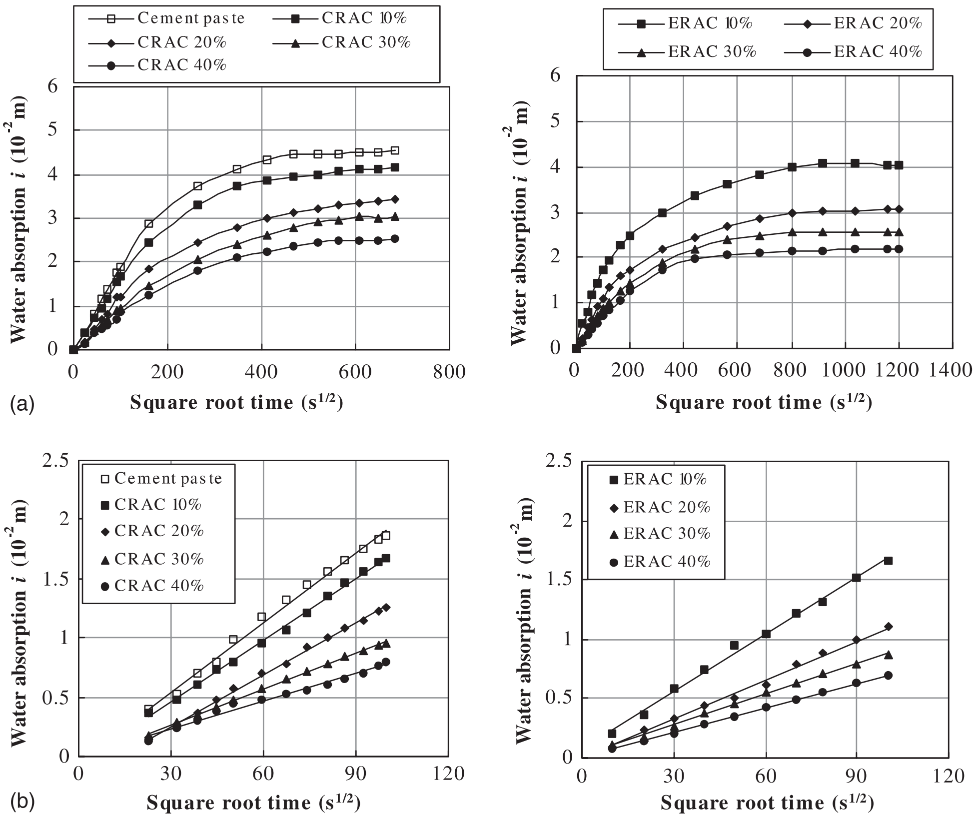


Fig. 5. Evolution of water absorption rate as a function of the square root of time for all compositions.

Table 3
Sorptivity values for all compositions

Volume of rubber aggregates (%)	Sorptivity values (mm/s$^{1/2}$)	
	CRAC	ERAC
0	0.193	
10	0.174	0.161
20	0.145	0.112
30	0.104	0.081
40	0.077	0.063

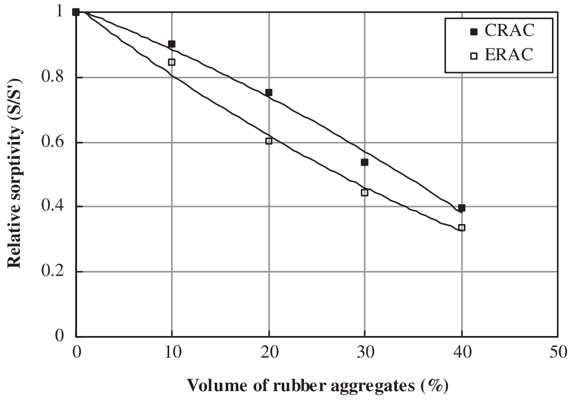


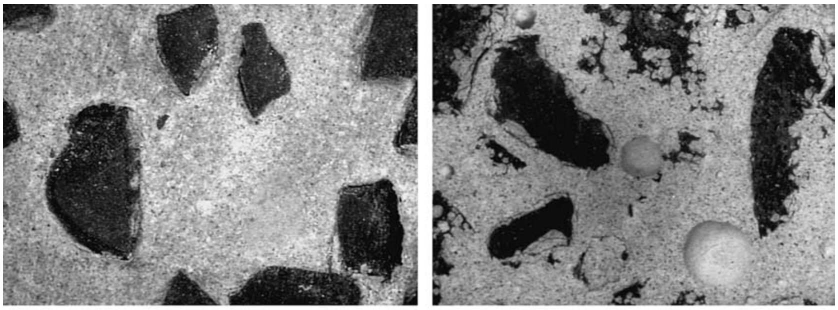
Fig. 6. Relationship between sorptivity and volume of rubber aggregates.

volume accessible to water and hence capillary porosity. But the increase of rubber particles in cement paste tends also to increase the level of air-entrainment during mixing. Table 4, which provides a list of these values, measured by the pressure method compared to the voids ratio measured by the pycnometer method, reveals that air-entrainment is more significant with expanded rubber than with compact rubber. The higher air content of mixtures may be due to the capability of rubber aggregates to entrap air in their surface.

Fig. 7 shows that the distribution of discontinuous voids due to air-entrainment in the cement matrix for CRAC and ERAC containing 30% rubber aggregates is different with the two types of aggregates. This

Table 4
Air-entrainment mixtures at different volume ratio of rubber particles

Volume of rubber aggregates (%)	Air-entrainment (%) (pressure method)		Voids ratio (%) (Pycnometer method)	
	CRAC	ERAC	CRAC	ERAC
0	2.0		2.8	
10	2.5	4.0	3.2	4.6
20	3.5	6.5	4.1	7.0
30	5.0	9.0	6.5	9.6
40	6.0	11.5	8.0	12.0



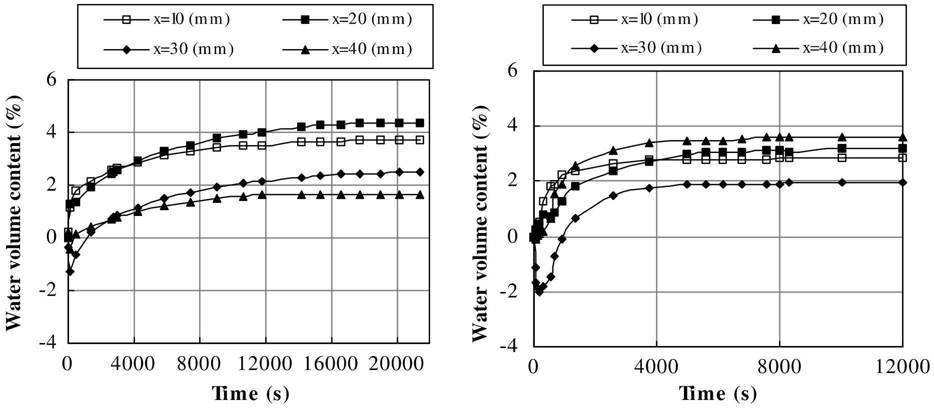
CRAC 30% ERAC 30%

Fig. 7. Optical photo of a distribution of discontinuous air-entrainment voids in a composite (30% rubber aggregates) (magnification: ×70).

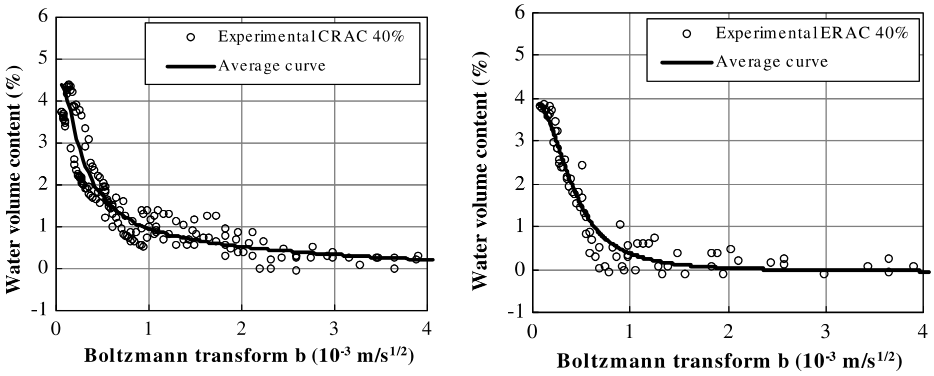
difference may be due to their morphological texture, because the volume of air normally associated with air entrainment is lower with smooth surfaces than with jagged surfaces. Consequently, we may suppose that the constriction of the water propagation in the composite is due to the presence of discontinuous air-entrainment voids, in conjunction with non-sorptive rubber particle additives, as exhibited in Tada's work [23].

3.2. Hydraulic diffusivity measurement

The composite's hydraulic diffusivity has been estimated by means of a gravimetric technique, based on a capillary suction test. Fig. 8a shows the local change in water volume content at different levels as a function of time. The corresponding single average Boltzmann profile curve, obtained with a 40% concentration of compact and expanded rubber aggregates, is displayed in Fig. 8b.



CRAC (40%) (a) ERAC (40%)



CRAC (40%) (b) ERAC (40%)

Fig. 8. Evolution in the water volume content of the local volume as a function of the Boltzmann transform (40% rubber aggregates).

The change in hydraulic diffusivity as a function of water volume content for all compositions with both types of rubber particles is shown in Fig. 9. For low water contents ($\theta \leqslant 0.5\%$), hydraulic diffusivity increases rapidly with water content and is sensitive to the volume ratio and the type of rubber. Above a defined threshold, it decreases to reach stability at a water content level that depends on the specimen's saturation point. According to Daïan [24], this point characterises the transition between vapour phase transfer and mixed liquid–vapour diffusion.

At a low water content, the condensed phase is present in an adsorbed form or in isolated capillary islets. A rise in water content serves to increase both the size and number of capillary islets. The vapour flow-passing section becomes increasingly reduced. Diffusion basically operates by means of condensation–evaporation mechanisms at liquid–vapour interfaces. Once continuity of the liquid phase has been established, the liquid phase transfer becomes more prevalent, to the detriment of the vapour phase transfer. Ultimately, at a high water content, the liquid phase transfer becomes predominant. In accordance with Crausse's findings [25] (except for the low water content zone, in which it is difficult to measure transport properties), we have observed that the increase in rubber content reduces the material's water sensitivity by virtue of limiting the diffusion process (see Fig. 10). Hall [22] explains this phenomenon by the necessity for the fluid to bypass the non-sorptive additives. This property is more significant with the expanded type of rubber aggregates (which probably results from their cellular surface) and further reduces capillary pressure in the cement paste and increases tortuosity.

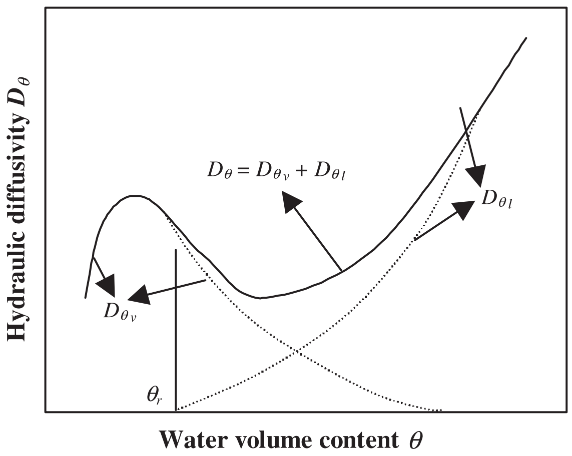


Fig. 10. Evolution of hydraulic diffusivity coefficient as a function of water volume content.

3.3. Air permeability measurement

The average air permeability coefficient values of CRAC and ERAC for different volumes of rubber particles under dry conditions are listed in Table 5. It is observed that the air permeability coefficient decreases with increasing rubber aggregate volume; values decrease from approximately 12.56×10^{-17} m^2 (cement paste) to 2.36×10^{-17} m^2 and 1.74×10^{-17} m^2 for a specimen containing 40% compact and expanded rubber, respectively. This difference between expanded and compact rubber can be attributed to the quench texture of aggregates as well as to air-entrainment, which increases the level of disconnected porosity. Such a finding

Table 5
Air permeability coefficient values for cement-rubber composites

Volume of rubber aggregates (%)	Air permeability coefficient values K (10^{-17} m^2)	
	CRAC	ERAC
0	12.56	
10	10.25	9.85
20	4.38	2.95
30	3.24	2.23
40	2.36	1.74

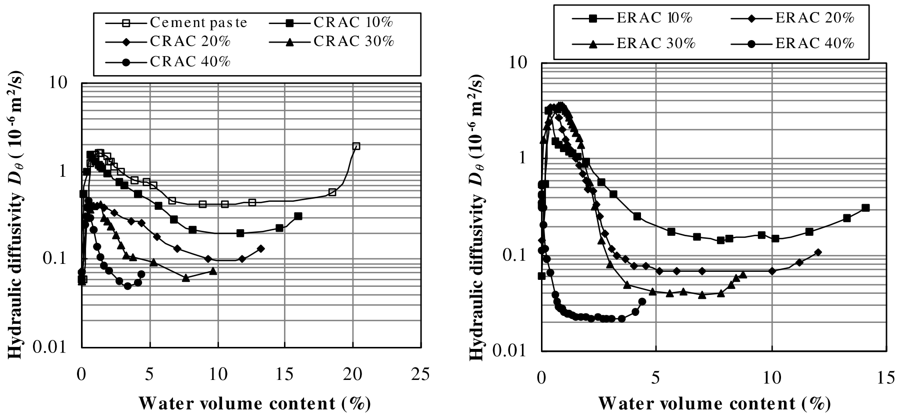
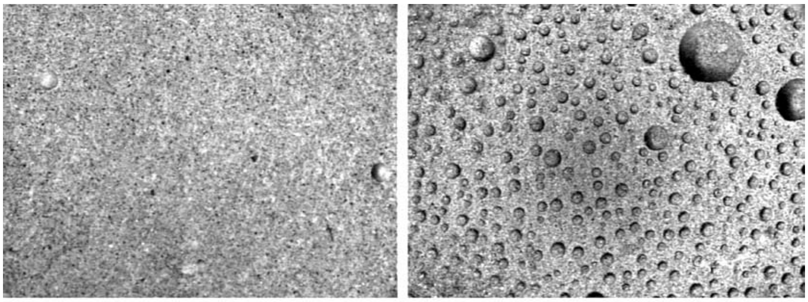


Fig. 9. Evolution in average hydraulic diffusivity as a function of water volume content for cement–rubber composites.



Cement paste (2% air-entrainment)

Cement paste (25% air-entrainment)

Fig. 11. Optical photo of cement paste specimens containing different amounts of discontinuous voids due to air-entrainment (magnification: ×70).

Table 6
Air permeability coefficient values for different amounts of air-entrainment in a cement paste

Air-entrainment (%)	Air permeability coefficient values K (10^{-17} m^2)
2.0	12.56
9.5	6.92
25.3	1.98

may serve to increase the tortuosity, thereby limiting air diffusion in the matrix. To ascertain these results, test specimens containing different amount of air-entrainment were prepared by mixing an air-entraining agent into cement paste (water/cement ratio = 0.3), during the plastic stage, at different times. The samples were cured, after hardening, for 28 days after demolding, at 20 °C and 98% relative humidity. The air permeability of hardened specimens was then determined. Fig. 11 shows an example of the distribution of discontinuous voids due to air-entrainment in the cement matrix. Table 6 which gives the corresponding values shows that increasing air entrainment corresponds with decreasing air permeability of specimens. These results are similar to that obtained by Kearsley and Wainwright [26] in their study on the effect of fly ash on the permeability of foamed concrete. They showed that if the porosity is high and the pores are interconnected the permeability is also high, but if the pores are discontinuous the permeability of the concrete is low although the porosity is high.

This property is of critical importance, particularly in light of carbonation and corrosion phenomena, which are responsible for the potential deterioration undergone by the material structure.

4. Conclusion

The work presented has allowed comparison of the transport properties of cement–rubber composites through the use of two types of rubber aggregates with different morphologies. An examination of hydraulic properties has revealed that the presence of rubber particles reduces sorptivity and hydraulic diffusivity by decreasing water absorption. Similarly, air permeability was considerably reduced due to the presence of these additives. The cellular character of rubber further improves behaviour when in contact with fluid. These results demonstrate the value of rubber additives for the durability of materials in aggressive environments.

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