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Isothermal water vapour permeability of concrete with different supplementary cementitious materials

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ABSTRACT: Water vapour permeability (WVP) is a key parameter for the sustainable thermal conditioning of buildings. The study of the WVP in concrete with supplementary cementitious materials (SCMs) allows for the design of structures with improved durability and sustainability. To our knowledge, there is insufficient experimental data in the literature regarding the WVP of concrete with SCMs.

WVP tests were made on concrete mixes containing ground granulated blast-furnace slag (GGBFS) and limestone powder (LP) as a partial replacement for ordinary Portland cement, and of concrete mixes containing pozzolanic cement (NP). Results from three moisture gradients show that GGBFS induces the greatest reduction in WVP, followed by the NP. LP shows a diluting effect of the binder, which could be compensated by GGBFS in the ternary blend. From the comparison between the WVP and the capillary sorption rate, the influence of the SCMs on the connectivity of the smallest pores is assessed.

KEYWORDS: Permeability; Durability; Blast furnace slag; Limestone; Pozzolane.

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RESUMEN: *Permeabilidad de vapor agua isotérmica de hormigones con diferentes adiciones minerales*. La permeabilidad al vapor de agua (PVA) es un parámetro fundamental para el acondicionamiento térmico sustentable de edificios. El estudio de PVA en hormigón con AM permite el diseño de estructuras con durabilidad y sustentabilidad mejoradas. A nuestro conocimiento, hay insuficiente información experimental en la literatura sobre PVA en hormigón con AM.

Se hicieron ensayos de PVA en hormigones con escoria granulada de alto horno (EGAH) y polvo calizo (PC) en reemplazos parciales de cemento normal, y de hormigones con cemento puzolánico. Resultados de tres gradientes de humedad muestran que EGHA induce a la mayor reducción de PVA, seguida para por la puzolana natural. PC muestra un efecto de dilución del ligante, el cual pudo ser compensado por la EGHA en las mezclas ternarias. De la comparación entre PVA y velocidad de succión capilar, la influencia de las AM en la conectividad de los poros más pequeños es evaluada.

PALABRAS CLAVE: Permeabilidad; Durabilidad; Escoria granulada de alto horno; Caliza; Puzolana.

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1. INTRODUCTION

Durability and sustainability have become the most important objectives when designing concrete mixes. In this respect, moisture transport mechanisms are particularly significant since they are responsible for the ingress of external agents and intimately related to the durability of concrete structures. Moisture diffusion under isothermal conditions is a transport mechanism that is driven by the existence of a concentration gradient, and it is highly influenced by the relative humidity (RH) (1). The flow of water vapour includes both gas and liquid phases of water, and hence, the pore radius involved in the process can affect the rate of transport in a particular manner. Therefore, test methods for the evaluation of the isothermal water vapour permeability (WVP) can be used to describe the pore structure of concrete and are helpful for predicting its durability when exposed to various environmental conditions. Moreover, the design of sustainable buildings requires the assessment of moisture transfer. The evaluation of the WVP is a tool to describe the ability of the material to be used as a vapour barrier that allows a more efficient conditioning of buildings.

Concerning sustainability, the increasing worldwide tendency towards the use of supplementary cementitious materials (SCMs) in concrete as a partial replacement for cement, one of the major sources of CO_2 emissions, is an excellent approach to responsible use of energy and raw materials. The conservation of natural resources by making materials last longer is, after all, a strategy for sustainability (2). The use of supplementary cementitious materials (SCMs) helps to reduce the clinker content per m^3 of concrete and hence to decrease the carbon footprint of concrete.

Thus, the study of SCMs and their effect on WVP is related to two main design features of concrete: durability and sustainability. This subject is covered in the literature to a very limited extent in comparison with the well-documented effects of SCMs on pore structure and other transport properties.

When ground granulated blast-furnace slag (GGBFS), a by-product of the steel industry, or natural pozzolan (NP) is added to concrete, there is normally an improvement in durability-related properties (3-5). This enhancement is mainly because GGBFS and NP naturally present delayed hydraulicity (very slow formation of C-S-H) and pozzolanic activity (reaction with portlandite to form additional C-S-H), which usually leads to pore refinement with time (6-8).

In contrast, Limestone Powder (LP) is an addition that does not show significant reactivity with hydration products from cement. It has been commonly considered an inert addition, and despite some reactivity to form carboaluminates as demonstrated in (9), this reaction has no significant impact on transport properties. Dilution and filler effects are the main consequences of replacing cement with LP. A high replacement ratio may therefore result in a reduction in strength and durability properties (10), due to a dilution effect of hydraulic components. Moreover, the effective water to cementitious materials ratio (w/cm) and permeability of concrete are increased with the replacement of clinker by LP (11). A consequential reduction in later strength may be obtained, and a limited LP content is generally recommended to obtain effective protection against aggressive environments (12). Nevertheless, early strength is generally increased because of the interaction between LP and clinker (13). The simultaneous actions of LP include calcium carboaluminate formation, accelerated C_3S hydration, and modifications in the Ca/Si relationship in C-S-H gel (14-15). Another beneficial effect associated with the use of LP is the attenuation of the sensibility of cement to the curing treatment, due to a higher hydration degree at early age (16). Therefore, LP is usually used in combination with other mineral additions in ternary blended cements to improve concrete properties.

As described above, the effects of GGBFS, NP and LP are complementary. Generally, LP accelerates hydration and improves early strength, whereas GGBFS or NP improves strength and refines pores at later ages. Several studies have investigated ternary blended cement with GGBFS and LP, and concrete admixed with GGBFS and LP regarding some transport properties and porosity (16-20). Water absorption is almost the same for concrete made with ordinary Portland cement (OPC) or GGBFS plus LP blended Portland cement (20), but dissimilar results regarding the effect on other transport properties may be found in the literature. Whereas the combined action of both SCMs might not significantly affect the total pore volume of concrete, pore refinement and increased tortuosity reduce permeability and sorptivity (21). It is not clear whether or not this would directly translate into the WVP, as the pore size range involved in each of these transport processes is not the same. Moreover, water vapour transport also involves evaporation and condensation in pores, which makes it a complex process to be examined as a combination of mass and energy transport mechanisms.

There seems to be a lack of reported experimental data for the WVP of concrete containing GGBFS, LP and NP. Saeidpour and Wadsö (22) studied mortars with slag and silica fume, with 70% and 10% replacement of cement in weight respectively, and with w/cm ratios of 0.4 and 0.5. They found that the presence of slag and silica fume decreased the diffusion coefficient in comparison with mortars with OPC, and also that for samples with SCMs the dependence on RH was small. However, they only tested three samples per mix, and no further comparison with other properties of the mortars was made. Chari *et al.* (23) investigated the effect of zeolite and silica fume on mortars with w/cm between 0.35 and 0.60 at different temperatures (20, 30, 40, 50, and 60°C). They measured the drying rate in an oven by weighing the samples at regular time intervals, and also tested the water absorption under immersion. Later on, they fitted the data and modelled the moisture transfer to find the "dry and wet" diffusion coefficients. The sample conditioning and drying procedure for the test were unfortunately done at 110±5°C, and this drying technique is well-known to damage the microstructure severely (24-25). It should be mentioned that the drying rate accounts for a specific process of water vapour diffusion, and the correlation with the mass transfer under service conditions is not direct. Kearsley and Wainwright (26) studied foamed concrete mixtures of different densities: 1000, 1250 and 1500 kg/m^3, and with different replacement ratios of cement with fly ash (50%, 66.7% and 75%). The authors claim that the WVP increases with cement/fly ash ratio. However, the presented data are not statistically different enough to provide sufficient evidence to support that conclusion. Their results are based on the average of only two samples, which are not enough data to obtain reliable results for diffusion tests. Some studies on WVP of self-compacting concrete (SCC) (27-28) have also been carried out, however the effect of water to cementitious materials ratio (w/cm) and the use of different SCMs on the WVP are not fully described.

In this study, the influences of GGBFS and LP, used separately and combined as a partial replacement for Portland cement and pozzolanic cement, on the WVP in nine concrete mixes with w/cm ranging between 0.35-0.50, are investigated. In consideration of different environmental exposure classes, three moisture gradients (MGs) (defined as the difference between the external and the internal RH) are assessed. The rate of weight gain and the isothermal WVP are determined and compared with other durability properties such as the total porosity and the capillary sorption rate (CSR).

2. MATERIALS

Two types of river sand were used as fine aggregate: coarse siliceous sand (CSS) and fine siliceous sand (FSS). The coarse aggregate used was granitic crushed stone with a nominal size in the range of 6-20 mm (GCS 6-20). Two types of water-reducing admixtures (WRA) were used: high range (WRAH) and low range (WRAL), and the dosage of the admixtures was chosen to achieve similar slump level. The properties of the aggregates and the reference standard test methods applied to determine those properties are shown in Table 1. The properties of the water-reducing admixtures are shown in Table 2.

Table 1. Properties of the aggregates

Properties	FSS	SCS	GCS 6-20	ASTM standard test
Density s.s.d. (g/cm^3)	2.60	2.60	2.65	C 127 (29) / C 128 (30)
Material passing the 75µm sieve (%)	1.52	0.71	0.55	C 117 (31)
Absorption (%)	0.8	0.2	0.4	C 127 (29) / C 128 (30)
Fineness modulus	1.61	2.62	-	C 136 (32)

Table 2. Properties of the water-reducing admixtures

Properties	WRAH	WRAL	ASTM standard test
Density (g/cm^3)	1.16	1.14	
Base composition	naphthalene sulfonate	modified lignosulfonate	
Solid residue	49.3	52.7	By drying at 60°C, according to ASTM C 1017 (33)

Two types of ordinary Portland cement (OPC1 and OPC2) and a pozzolanic Portland cement (PPC) were used as the main binder constituents. LP and GGBFS were used in different proportions as partial replacements for cement. Table 3 shows the physical and chemical properties of the cements and the SCMs.

Concrete mixes with four different w/cm ratios were made: 0.35, 0.40, 0.45 and 0.50. In the labelling of the concrete mixes, the numerical nomenclature denotes the w/cm.100; therefore a w/cm ratio of 0.35 is denoted as 35. The letters correspond to the binder used, as follows: O-1 and O-2 stand for concrete made only with OPC1 or OPC2, respectively; S with OPC and GGBFS; LP with OPC and LP; SLP with OPC, LP and GGBFS; and P with PPC. The properties determined in the fresh state were air content, unit weight, slump, and bleeding. In the hardened state, water absorption after 24 hours of immersion, porosity and compressive strength at 7, 28 and 90 days were evaluated. In addition, CSR (considered as the ratio between weight gain due to water uptake and the square root of time) was determined for the same concrete mixes. The complete test procedure and computation steps can be found in (21). Table 4 shows the proportioning of the nine concrete mixes studied and the results from the mentioned tests with the corresponding applied standard.

From those concrete mixes, prisms of 7x7x30 cm^3 were moulded and kept in a humid chamber at 20 ± 2°C and 95 ± 5% RH for 28 days. After this period

Table 3. Chemical and physical properties of the cements and the SCMs

Properties	OPC1	OPC2	PPC	GGBFS	LP	ASTM standard test
Blaine specific surface (m^2/kg)	288	380	281	373	556	C 204 (34)
Material retained on sieve 75µm (%)	2.4	1.30	2.1	<0.01	1.60	C 786 (35)
Compressive strength 2d (MPa)	20.4	25.6	18.9	-	-	C349 (36)
Compressive strength 28d (MPa)	44.4	45.2	41.8	-	-	C349 (36)
Density (g/cm^3)	3.13	3.11	2.95	2.87	2.75	C 188 (37)
Slag activity index (%)		-	-	95	-	C989M (38)
Chemical analysis (%)						C 114 (39)
Loss on ignition	0.80	2.14	1.20	0.87	36.01	
Insoluble residue	1.40	2.50	21.65	3.40	6.65	
SO_3	1.74	2.41	1.40	3.58	0.21	
MgO	2.44	2.76	1.01	9.36	0.76	
SiO_2	20.57	19.93	14.79	30.49	11.58	
Fe_2O_3	4.30	4.00	3.09	0.29	0.68	
Al_2O_3	4.22	4.30	4.77	10.68	1.82	
CaO	64.55	60.38	50.32	38.17	45.82	
Na_2O	0.30	0.14	0.16	1.42	0.92	
K_2O	0.98	0.85	1.24	0.44	0.19	
Mn_2O_3	-	-	-	0.50	0.16	
Cl^-	0.023	0.012	0.03	<0.001	0.01	

Table 4. Proportioning and properties of concrete mixes

Series	O35-1	O35-2	O40	O45	O50	S40	LP40	SLP40	P35	ASTM Standard test
w/cm	0.35	0.35	0.40	0.45	0.50	0.40	0.40	0.40	0.35	
Materials (kg/m^3)										
Water	133	140	140	144	150	140	140	140	140	
OPC1	380	-	-	-	-	-	-	-	-	
OPC2	-	400	350	320	300	227	262	227	-	
PPC	-	-	-	-	-	-	-	-	400	
GGBFS	-	-	-	-	-	123	-	88	-	
LP	-	-	-	-	-	-	88	35	-	
FSS	189	188	190	193	193	190	190	190	182	
SCS	749	739	754	766	767	755	755	755	713	
GCS 6-20	980	979	980	980	980	980	980	980	979	
WRAH	6.2	3.4	5.9	6.0	3.6	4.9	4.2	4.1	4.7	
WRAL	-	2.7	-	-	-	-	-	-	2.7	
Air (%)	3.0	4.2	3.1	3.0	3.1	3.2	3.5	3.4	3.2	C231 (40)
Unit weight (kg/m^3)	2404	2392	2417	2392	2404	2392	2354	2385	2417	C138 (41)
Slump (cm)	8.0	11.0	10.0	6.0	6.0	10.0	9.0	7.0	16.0	C143 (42)
Bleeding (%)	<0.01	<0.01	<0.01	3.03	0.40	<0.01	<0.01	<0.01	<0.01	C232 (43)
Water absorption, 24 h immersion (%)	3.37	2.50	3.64	3.78	3.78	4.58	3.77	4.55	2.84	C642 (44)
Porosity at 28 days (%)	8.02	6.02	8.68	8.90	8.86	10.60	8.78	10.49	6.81	C642 (44)
Compressive strength (MPa) 7d	54.2	-	47.6	37.8	37.4	47.6	37.4	43.6	-	C39 (45)
Compressive strength (MPa) 28d	60.2	58.1	53.8	45.6	46.2	52.6	42.2	51.1	48.4	
Compressive strength (MPa) 90d	-	62.1	-		-	58.1	47.3	60.8	60.3	
CSR (g/m^2/$s^{1/2}$)	1.44	1.31	1.68	2.42	2.88	0.88	1.48	0.91	1.68	*

*Argentine standard (IRAM 1871 (46))

they were cut into slices of approximately 0.5 cm and kept in a laboratory environment for approximately 90 days until they were tested. Samples were tested at the age of over 90 days to limit the possible impact of cement or SCM hydration on the results. It is considered that no significant further hydration that could affect the results occurred after these 90 days, especially when taking into account the fact that samples were maintained in unsaturated conditions during testing. The samples were hygroscopically equilibrated with the environment defining the equilibrium condition as a variation in weight of less than 0.1% in a 7 day period.

3. METHODS

The moisture diffusion phenomenon in concrete was analysed using the cup method (47-48). The applied method is an adaptation for the particular case of concrete samples considering the guidelines of ISO 12572 (49) and ASTM E96 (50). The water vapour transmission (WVT) is defined as the steady water vapour perpendicular flux per unit area through a layer of the material, under specific conditions of temperature and humidity, in g/(h·m²). Figure 1 shows a scheme of the method, where the WVT through a concrete sample is achieved by the existence of an MG using the sample as a barrier. The MG was calculated as the difference between the RH of the outside and inside environment. Inside the cup, a constant RH (i.e. vapour water pressure) was maintained with a hygroscopic salt. The concentration of a certain saturated solution at a certain temperature is constant, and when an excess of solute is provided, the mixture is able to keep a constant RH in the surrounding environment by acting as a source or sink of moisture in case of modest changes in the water content of the environment. The concrete sample was then assembled into the cup as a completely sealed cover. The outside environment was a wet chamber with RH > 95% at 22°C±1°C (isothermal conditions throughout the whole test). The sample was prevented from being in contact with any direct source of liquid water. The three MGs were achieved by using different salts inside the cup as indicated in Table 5.

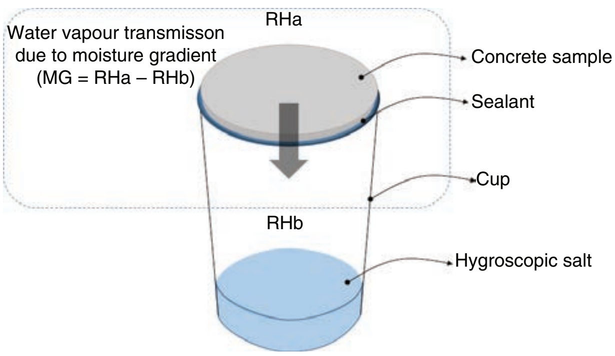

FIG. 1. Scheme of the set-up of the cup method.

TABLE 5. Salt solutions with corresponding RHs at 22°C

Saturated salt solution	Static internal RH (%)	MG (%)
NaCl	75.4 ± 0.1	19.6
K_2CO_3	43.2 ± 0.4	51.8
LiCl	11.3 ± 0.3	83.7

The samples tested were concrete slices of 7.5 cm in diameter and approximately 0.5 cm in thickness. Figure 2 shows the samples (a) and the cup (b) used in the tests. Five samples for each concrete type and each RH were tested at an advanced age (>90 days) to ensure negligible influence from changes in the pore structure with time. Those slices were then attached to the plastic cups, avoiding direct contact between the concrete slices and the solution, and completely sealed with silicone rubber. The cups were put in a humid chamber with 95% RH at 22°C±1°C. This was done to ensure an MG equal to the difference between the moisture content outside (95% RH) and inside the cup (provided by the salt). Given that the external environment was maintained at a fixed high RH, a higher MG means that concrete samples were relatively drier along their depth. The hygroscopic equilibrium reached with the surrounding environment means that with lower RH, the fraction of pore sizes at which condensation occurs is smaller and the fraction of pores available for moisture transport is larger, provided that transport of liquid does not occur.

The cups were first weighed after 24 hours of exposure, and then they were weighed at progressive time intervals until a total weight gain of approximately 10 g was registered. Since the samples did not have exactly the same thickness, for comparative purposes the influence of the thickness was removed. To do this, the thickness of each sample was measured at two points and averaged to be later considered in the calculations, which are described below.

The weight gain is a result of the flux through the sample, which in technical terms is called WVT. According to the first Fick's law, under steady conditions the WVT is computed as stated in Equation [1]:

$$[WVT = - D_w \cdot dc_w/dx], \qquad [1]$$

where D_w is the water vapour diffusion coefficient, and dc_w/dx is the gradient of the water gas phase concentration (dc_w) across the sample thickness (dx). If we express Equation [1] in terms of water vapour pressure (p_w), and assuming ideal behaviour ($p_w = c_w \cdot RT/M$; with R = g as constant

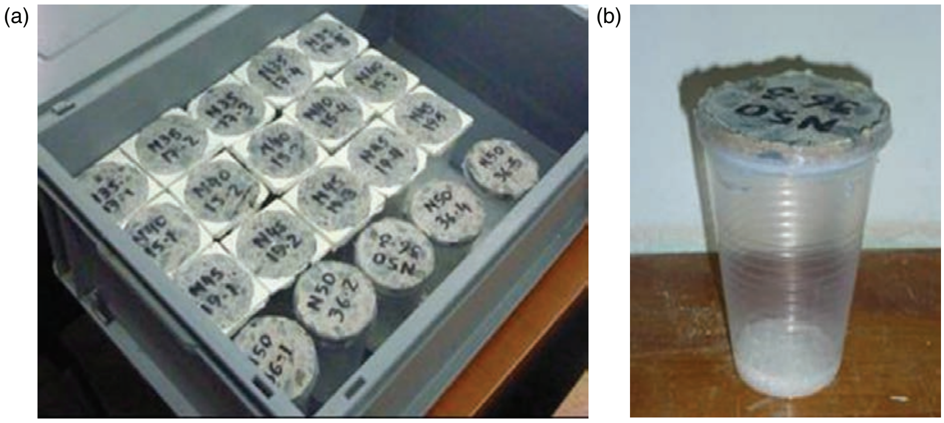

Fig. 2. Samples during the test (a), hygroscopic salt inside a cup where the concrete sample is the barrier (b)

[8.314 J/(mol K)], T = absolute temperature and M = the molar mass of water), the result is Equation [2]:

$$[WVT = - D_w \cdot M/(RT) \cdot dp_w/dx = -WVP \cdot dp_w/dx] \quad [2]$$

Here, the term - $D_w \cdot M/(RT)$ is referred to as the water vapour permeability (WVP).

A linear distribution is considered for the gradient of water vapour pressure: $dp_w/dx = S \cdot (R_a - R_b)/\Delta x$ or $dp_w/dx = S \cdot (MG)/\Delta x$, where S = saturation vapour pressure at test temperature ($46.66 \cdot 10^2$ Pa), R_a = RH at the source expressed as a fraction, R_b = RH at the vapour sink expressed as a fraction, and Δx = averaged sample thickness (m). In this study, the WVP is calculated as the product of each WVT, induced by the vapour pressure difference at isothermal conditions, and the thickness of the sample, divided by (S·MG), in g/(nPa·s·m), as shown in Equation [3]:

$$[WVP = WVT \cdot \Delta x /(S \cdot MG)] \quad [3]$$

For this computation, the weight gain (G) of each sample divided by the exposed area (G/A) and multiplied by Δx is shown for comparative purposes. The progression of $G/A \cdot \Delta x$ with time was fitted by linear regression to determine the average rate of WVT considering the thickness ($WVT \cdot \Delta x$) under steady conditions. It should be mentioned that D_w can be expected to be independent of c_w as long as water moves through the sample only by means of molecular diffusion in the gas phase. This is not absolutely true in porous materials, as condensation and evaporation take place in the pore structure as a function of the RH. Then, more condensation leads to blocking of the diffusion path, whereas evaporation liberates it. After being exposed to the MG some parts of the samples became drier and the rest became wetter, depending on their position with respect to the humidity source and sink. The only exceptions were the tests with NaCl, where the samples did not dry through their thickness, given that the highest RH was similar to the conditioning RH. The applied methodology is able to determine an average WVP in the steady regime through the sample, but condensation or evaporation that lead to this steady regime may imply hysteresis effects in the water content of the samples, and then possibly result in slightly different pore connectivity. Although this could lead to further experimental studies, dynamic measurements were not considered for the purpose of this work. Joy & Wilson (51) discussed the possible hysteresis effects that cause a dependence of the results on the initial moisture content of the sample. To address this issue, they recommended that hygroscopic materials should be preconditioned in a standard atmosphere before WVT tests are made. For this purpose, the concrete slices were hygroscopically equilibrated in the laboratory environment, with 75+/-10% RH and 20+/-5°C.

4. RESULTS AND DISCUSSION

4.1. Influence of the applied MG on the WVT·Δx for OPC concrete

Figure 3 (a, b, and c) shows the results of the cup method test performed on OPC concrete mixes with different w/cm ratios, for the three different MGs studied: 83.7%, 51.8%, and 19.6%, respectively. For all cases, $WVT \cdot \Delta x$, represented as the slope of the linear relationship between $G/A \cdot \Delta x$ and time, varies as a function of the MG. A higher $WVT \cdot \Delta x$ for a higher w/cm is found only for the highest MG. However, this trend is not observed for the lower MGs, since no distinguishable differences among the different w/cm ratios are found. However, a reduction in the value of $WVT \cdot \Delta x$ can be seen as the MG diminishes for all OPC concretes. This outcome is expected since the driving forces of diffusion decrease when the MG

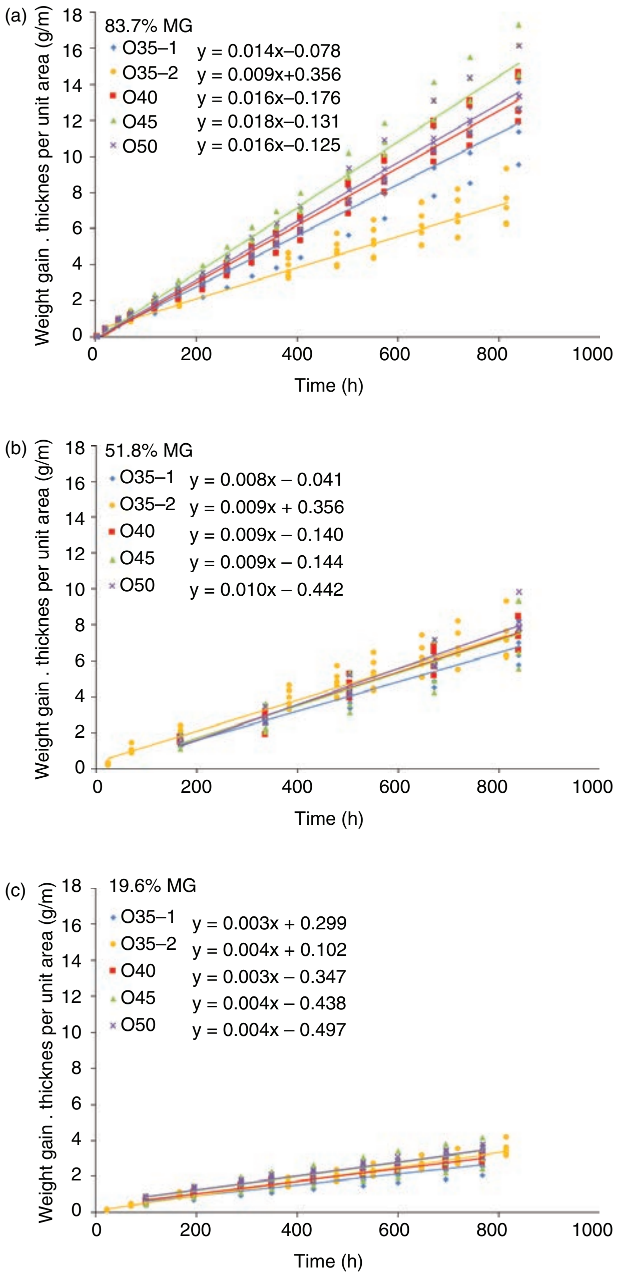

FIG. 3. WVT·Δx in OPC concrete mixes with different w/cm, MG = (a) 83.7%, (b) 51.8%, (c) 19.6%.

decreases. Particularly for the case of Figure 3 (a), a difference between the values of the WVT·Δx for O35-1 and O35-2 can be seen. Although both concrete mixes have the same w/cm, OPC1 is coarser than OPC2 as shown in the results of Blaine's specific surface area and in material retained on the sieve of 75 µm (see Table 2). The transport phenomenon is affected by this difference, as revealed by higher WVT·Δx, porosity and CSR values for O35-1 than for O35-2.

4.2. Influence of the applied MG on WVT·Δx for concrete mixes with SCMs

Figure 4 (a, b, and c) shows the results of the cup method test performed on concretes with a w/cm equal to 0.40 for the three MGs. The control concrete was made only with OPC (O40), while the other concretes were prepared by replacing part of the cement with GGBFS (S40), LP (LP40), and a combination of both GGBFS and LP (SLP40). The variation of WVT·Δx as a function of the MG is similar to that registered for OPC concretes, whereby the higher the MG, the higher the value of WVT·Δx.

As described previously, the influence of the added GGBFS is directly related to the latent hydraulic effect, which involves the generation of hydration products that reduce and obstruct the pore connectivity. This beneficial effect can be seen in S40, for which the results show a reduction in WVT·Δx in comparison with O40 for all the different studied MGs. Similarly, Vejmwlková *et al.* (27) have found a positive effect from GGBFS on vapour diffusion in SCC mixes with w/cm=0.39 and 10% replacement of the Portland cement, compared with a reference concrete mix. Saeidpour and Wadsö (22) also found a positive effect due to the presence of slag in mortars with 70% replacement, tested after 90 days and with w/cm = 0.40.

Furthermore, the results demonstrate an effect of GGBFS in relation to the MG. Figure 4 (a, b, and c) show an increasing influence of GGBFS with increasing MG. As the moisture content decreases with a larger gradient, small capillary pores are evacuated, and water vapour transport is more and more related to the smaller fraction of pore sizes. This small capillary pore fraction is influenced more by the presence of GGBFS when reacting and forming secondary hydration products. When the gradient is small, the smallest pore fraction remains saturated, and the transport process is dominated by the coarse fraction of pores. The macropores and bigger capillary pores are not significantly affected by the latent hydraulic effect of the GGBFS and therefore neither are the transport properties that are dependent on that pore size range (21), including the flux of water vapour.

The same positive effect but less marked is found for SLP40. This limited effect is due to the combination of the lower replacement percentage of GGBFS (only 25% in SLP40) and the dilution effect of the LP. Unlike the case for the concrete mixes with only GGBFS, the relative LP influence found on the WVT·Δx is similar for the three MGs. The LP incorporation causes the dilution of clinker (which leads to a higher effective water/cement ratio) and, as a positive effect, it also causes the provision of more space for hydration products

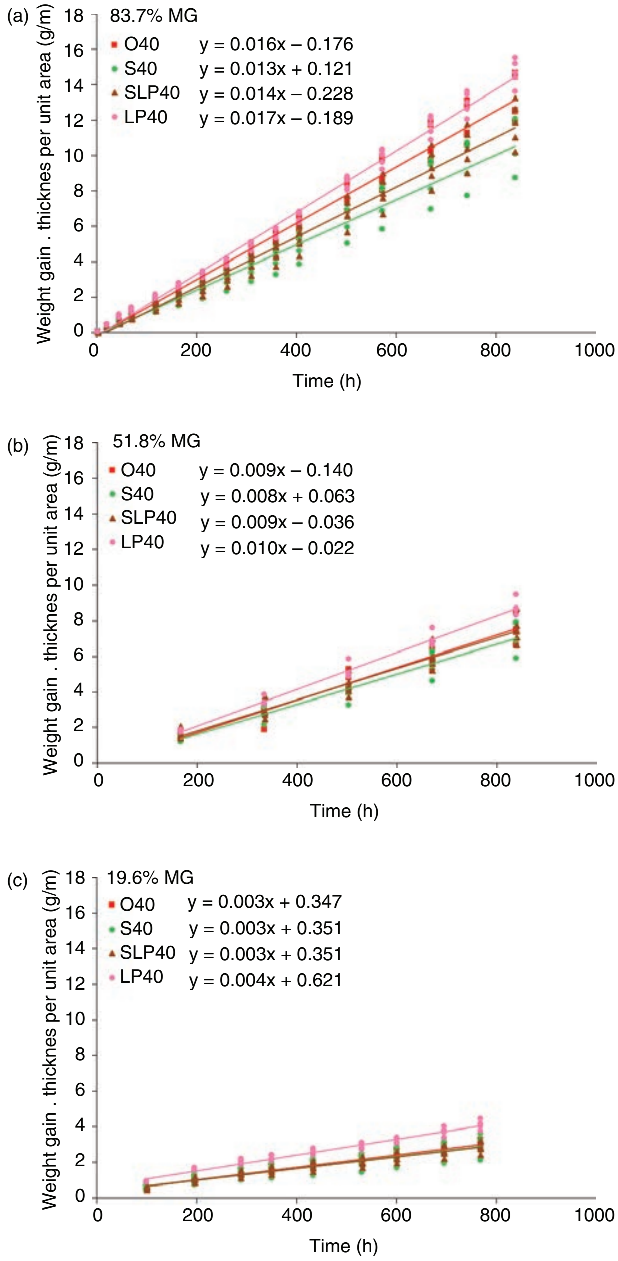

FIG. 4. WVT·Δx in concrete mixes with different SCMs and w/cm = 0.40, MG = (a) 83.7%, (b) 51.8%, (c) 19.6%.

and nucleation sites. Both dilution and enhancement of hydration are competitive effects, and the result depends on the amount of replacement (14, 17). For LP40 (25% of the cement replaced), the LP incorporation has a slightly negative effect. Here, it was found that the dominant effect of this SCM for this replacement ratio is dilution. Some effects derived from the formation of carboaluminates with the inclusion of LP could also be expected. However, this seems to be minimal in comparison with the effect of dilution, as shown by the results of LP40. Contrastingly, Mňahoncáková *et al.* (28) found a positive effect on wet and dry cup tests performed on SCC mixes with 40% cement replaced with limestone. However, the w/cm of the mix was 0.28, and the total binder content was 632 kg/m^3. The clinker content per m^3 of concrete was therefore significantly higher than the clinker content in the studied LP40 mix. This means that LP in (28) was incorporated to increase the content of fines and to obtain a stable mix, rather than to replace part of the cement. It is therefore natural that the outcome is different in both cases.

Figure 5 (a, b and c) show the results of the cup method test performed on concrete mixes with a w/cm equal to 0.35: two concrete mixes made with OPC1 and OPC2 (O35-1 and O35-2 respectively), and one concrete mix made with pozzolanic Portland cement (P35). The scattering obtained does not allow for the identification of clear differences between the P35 and O35-x concretes. Unlike the improved performance regarding transport properties of pozzolanic cements in comparison with OPC usually reported in the literature, no negative or positive effect of NP can be derived from the results obtained. However, if the results are evaluated regarding effective water to cement ratio, the reduction of clinker content in the mix was compensated by the action of the natural pozzolan in terms of water vapour permeability, which means a significant contribution of the SCM.

4.3. Comparison between WVP, porosity and CSR

As mentioned previously, the influence of the w/cm ratio on WVT·Δx in concrete is more pronounced in the case of a higher MG. Accordingly, Figure 6 compares the WVP and the total porosity for pure OPC concrete mixes, for the different w/cm ratios studied, i. e. 0.35, 0.40, 0.45, and 0.50. The values presented are the averages for the series. The general trend found is decreasing decrease in WVP with decreasing porosity. This is directly related to the increase in the matrix densification given by a low w/cm. Diffusivity is not directly dependent on pore size, but it is proportional to pore connectivity, which is increased in a paste with higher w/cm and thus larger pores (52). For transport of liquids and gases, only the part of the total porosity which is connected to the boundaries or surface is important. This is often called open porosity (53). However, the increase in the WVP with the w/cm is relatively low. The influence of the w/cm ratio is limited by the relatively low volume of paste used in the tested concretes. It is likely that the actual MG achieved in these tests is not simply the difference between the RH given by the hygroscopic salt and the RH in the outside environment. The difference in vapour pressure over the specimen will be somewhat smaller than that corresponding to ambient climates (54). In order to consider this possible MG reduction,

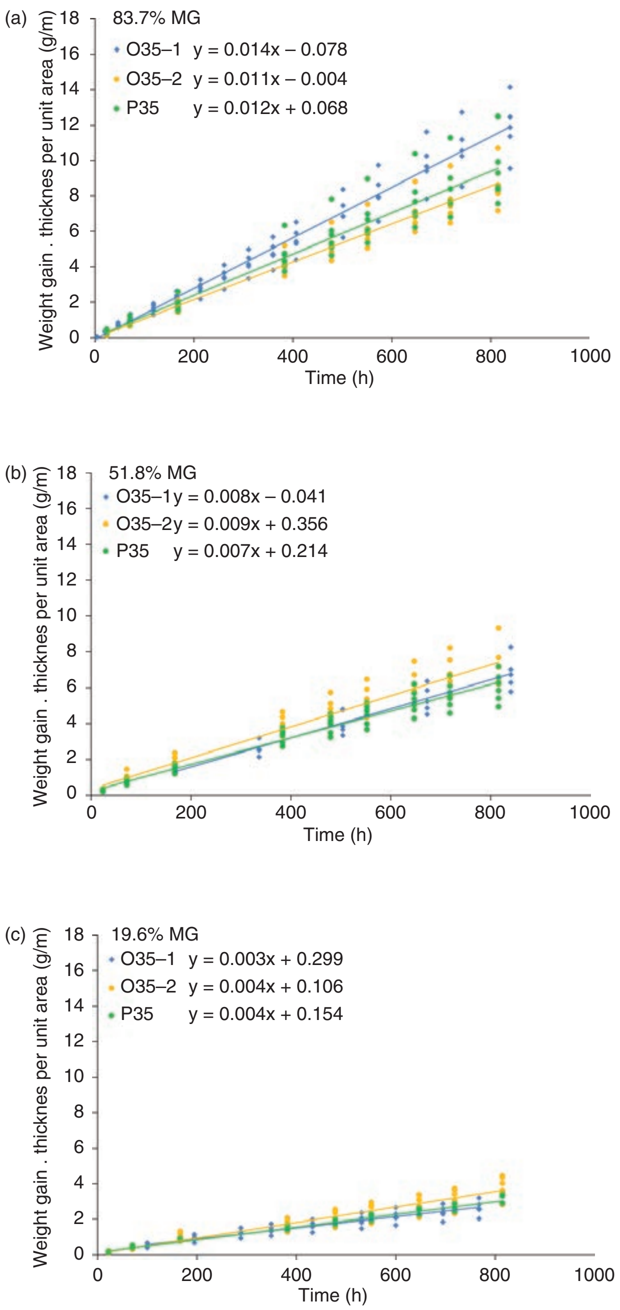

FIG. 5. WVT·Δx in concrete mixes with w/cm = 0.35, MG = (a) 83.7%, (b) 51.8%, (c) 19.6%.

measurements of RH both inside and outside of the sample, near the surface would be required, which could not be conducted in this work due to practical limitations.

Figure 7 compares WVPs of the same OPC concrete mixes and their capillary porosities (calculated as the moisture content of the sample when equilibrated at 97% RH and considering the density of each concrete mix). In contrast with Fig. 6, no direct relationship between the two parameters is observed. WVP is actually a combined process in which vapour first enters the sample, and egresses again with evaporation in the direction towards the sink, with some condensation occurring inside the sample depending on pore sizes. From the results, the amount of capillary porosity seems to be less significant than pore volume and connectivity. It is the total connected pore volume which shows a better correlation with WVP. Since vapour diffusion is a mechanism that involves the whole pore volume of the sample the difference seems natural, as concrete mixes show different ratios between capillary and total porosities. Similarly, Saeidpour and Wadsö (22) concluded, from studies made on mortars with slag, that the volume of capillary pores does not affect the diffusion transport significantly. Consequently, a linear relationship is derived only when the whole connected pore system (capillary and gel pores) is considered in the porosity values. Therefore, only total porosity is compared with WVP for the rest of the concrete mixes.

Since the diffusion mechanism is related to pore structure, for this transport process the impact of the SCMs on tortuosity and connectivity is more relevant than the impact on total porosity. Figure 8 shows the relationship between WVP and porosity for the different SCMs and the three MGs studied. Results from concrete mixes O35-2 and O40 are added for reference. It may seem unexpected that the lowest MG leads to the highest WVP, however in the computation of the WVP, MG is in the denominator and is simply considered as the different between internal and external RHs. Then, for the same WVT, a lower MG leads to a higher value of WVP. The simplified assumption of a linear distribution of the MG does not account for the ability of concrete to retain liquid water in pores due to the pressure in the internal pore network, which then reduces WVT. If this is the case, the actual MG inside the sample is actually lower than the difference between the RH in the two surrounding environments. Concrete is a hygroscopic material due to its C-S-H content, and this causes differences regarding water transport in comparison with other building materials (bricks, plaster). For this reason, it is very likely that different WVP values would have been obtained if the samples would have been dried below the RH in the sink chamber, so that only moisture adsorption occurred during the experiments. Still, from a practical point of view, it seems better to refer to WVP as a response to the difference in RH in the two external environments, and for increased description maybe to also include the consideration of whether the transport process is occurring after absorption or desorption in the sample to account for hysteresis effects.

The reduction in w/cm (from 0.40 to 0.35) led to a decrease in the total porosity of P35 and O35-2 in comparison to O40, LP40, SLP40 and S40.

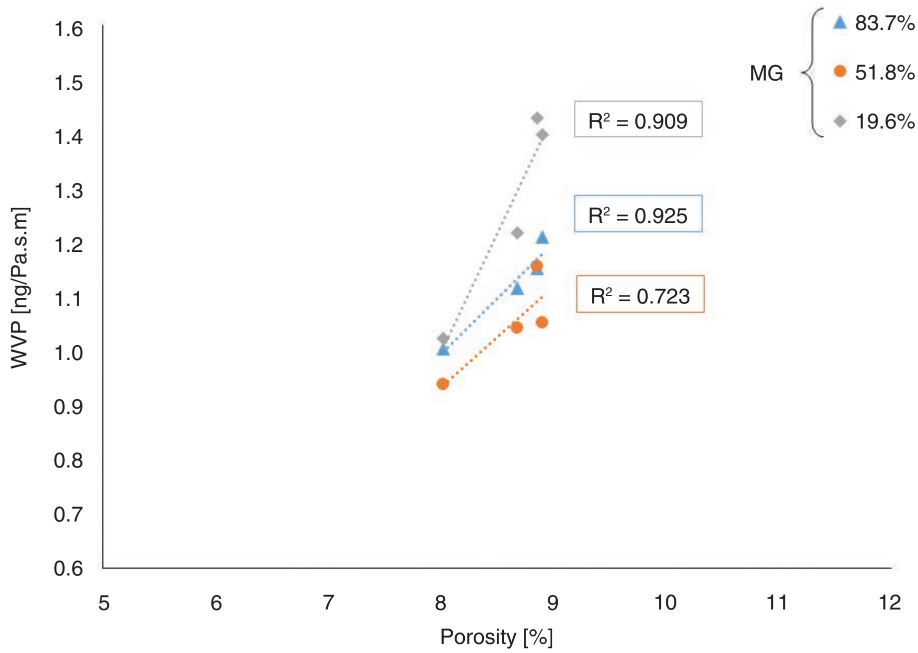

FIG. 6. Relationship between WVP and porosity of pure OPC concrete mixes for the different w/cm and MGs studied

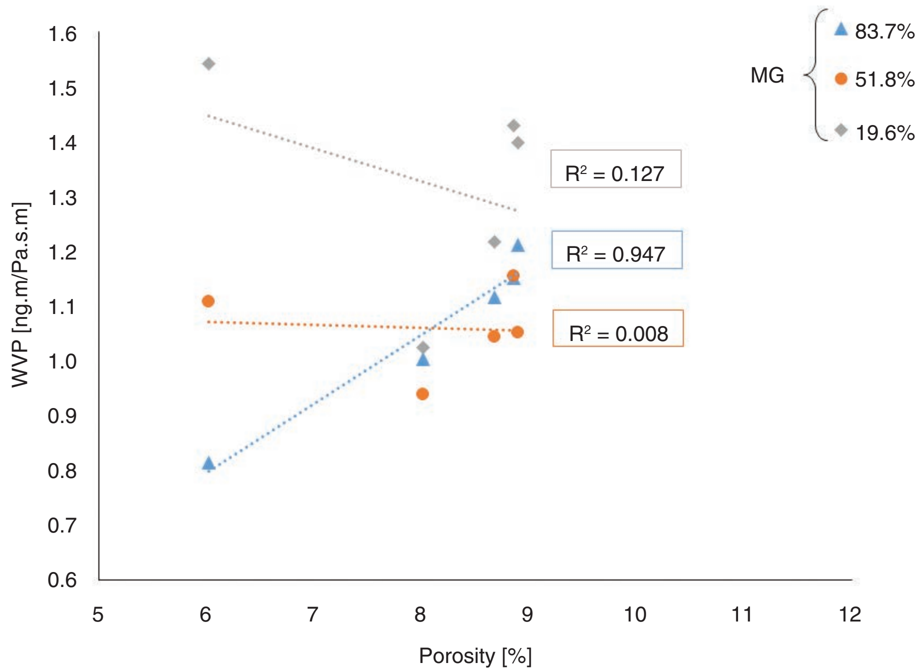

FIG. 7. Relationship between WVP and capillary porosity of pure OPC concrete mixes for the different w/cm and MGs studied

However, this reduction is not reflected in the WVP. The difference obtained between P35 and O35-2 shows that the reaction of the natural pozzolan only compensated for the reduction in clinker content to a limited extent. Conversely, higher total porosity but lower WVP than LP40 is obtained for the concrete with GGBFS, showing significant pore refinement that also leads to the lowest CSRs among the studied mixes. It should also be mentioned that the replacement ratio of clinker by natural pozzolan in PPC is higher than the replacement ratio by GGBFS and limestone in S40, LP40 and SLP40, meaning that differences between the natural pozzolan and GGBFS are lower in terms of

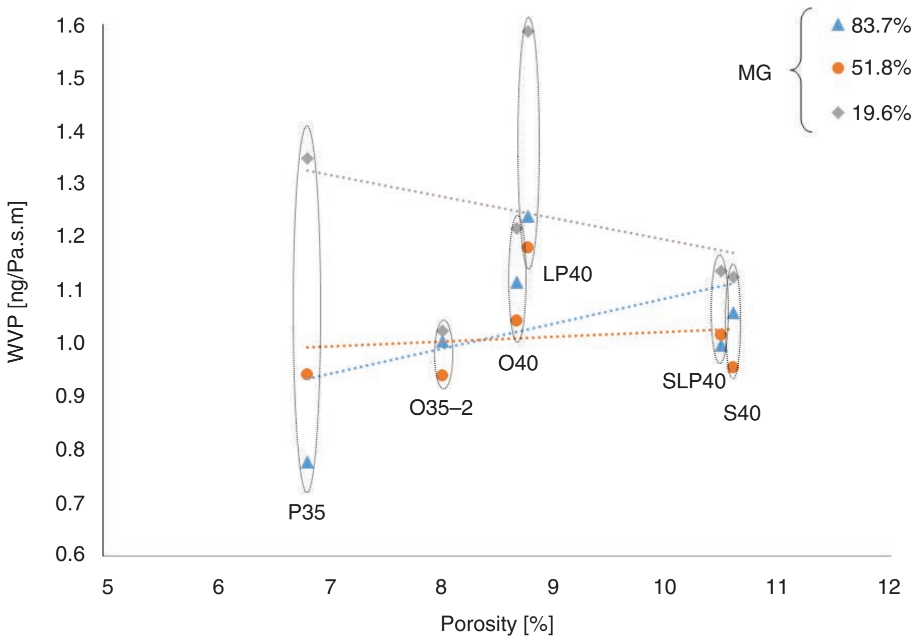

FIG. 8. Relationship between WVP and porosity for the different SCMs and MGs studied

specific efficiency. For the case of LP40, the results show the non-pozzolanic nature of this SCM, as the highest values of WVP are obtained with no pore refinement capacity as in the case of concrete mixes with GGBFS or natural pozzolan. Even the concrete mix with a ternary binder, SLP40, has lower values of WVP than LP40. This indicats that GGBFS is able to enhance the pore structure even with a lower clinker content when comparing SLP40 to LP40. Therefore, the eventual formation of carboaluminates due to the inclusion of LP in concrete seems to have little significance in this respect in comparison with the effect of dilution of hydraulic phases.

Figure 9 shows the relationship between WVP and CSR of pure OPC concrete mixes for the different w/cm and MGs studied. The increase in the w/cm ratio normally leads to an increase in both WVP and CSR. From these results, a nearly linear trend between WVP and CSR is observed for OPC.

Figure 10 shows the relationship between WVP and CSR for mixes with the different SCMs and MGs studied. These results do not correspond well with the linear relationship between CSR and WVP observed for OPC concrete. S40 has the lowest CSR and WVP values for all MGs showing that both tests reflect the influence of GGBFS on pore connectivity. In contrast, LP40 has less porosity than S40, and still has a higher value of CSR and WVP than S40 for all MGs. This reflects the pore filling action of the studied GGBFS at advanced ages. Furthermore, the action of the GGBFS causes an increase in the smallest pore fraction which, based on the comparison made with capillary pores, is a more influential fraction in the diffusion phenomena. This is connected to the fact that tortuosity, connectivity and pore size distribution of the pore system are more relevant for transport properties than total pore volume. In the case of SLP40, there is a combined action of dilution given by the LP addition and pore refinement given by the presence of GGBFS. Still, with lower clinker content than LP40, SLP 40 shows better results, indicating the prevalence of the pore refinement action over the dilution effect.

5. CONCLUSIONS

The cup method test was used to measure the isothermal WVP in plain and blended concrete mixes including GGBFS, LP and NP. The WVP was effective in showing the influence of SCMs since the diffusion mechanism is related to pore structure, and it was found that the impact of the SCMs on tortuosity and connectivity is more relevant than the impact on total porosity for this transport process.

Significant benefits were obtained in a binary concrete mix with the incorporation of GGBFS. The reduction in the WVP in comparison with the control concrete was registered, with a higher effect of GGBFS for larger MG's. For the case of the ternary concrete mix with GGBFS and LP, only a slight improvement in comparison with the control concrete was found for the different MGs studied. However, as the clinker content was reduced by the presence of both GGBFS and LP, the results showed the large extent of the pore refinement action by the

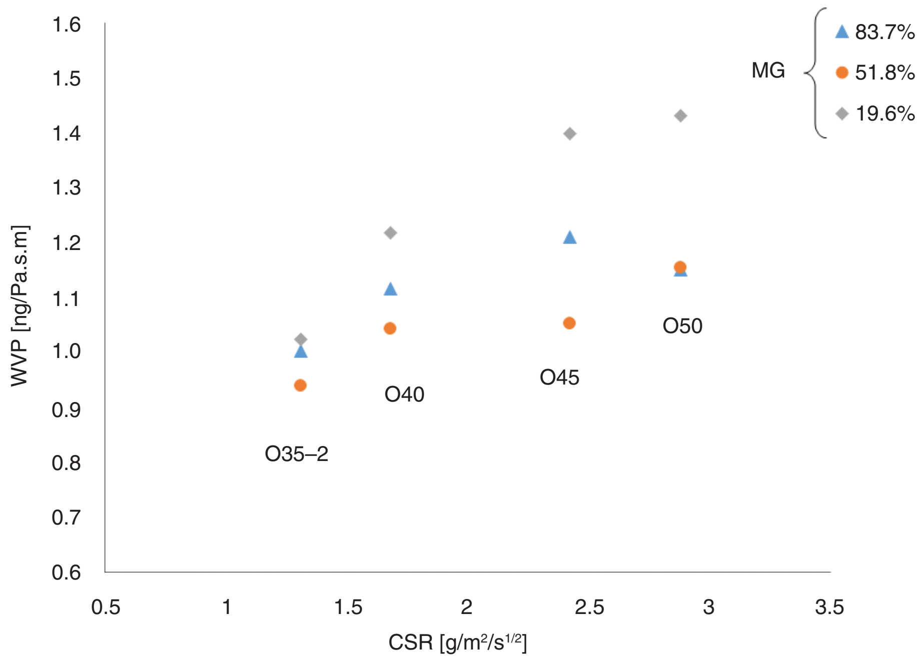

FIG. 9. Relationship between WVP and CSR of pure OPC concrete mixes for the different w/cm and MGs studied

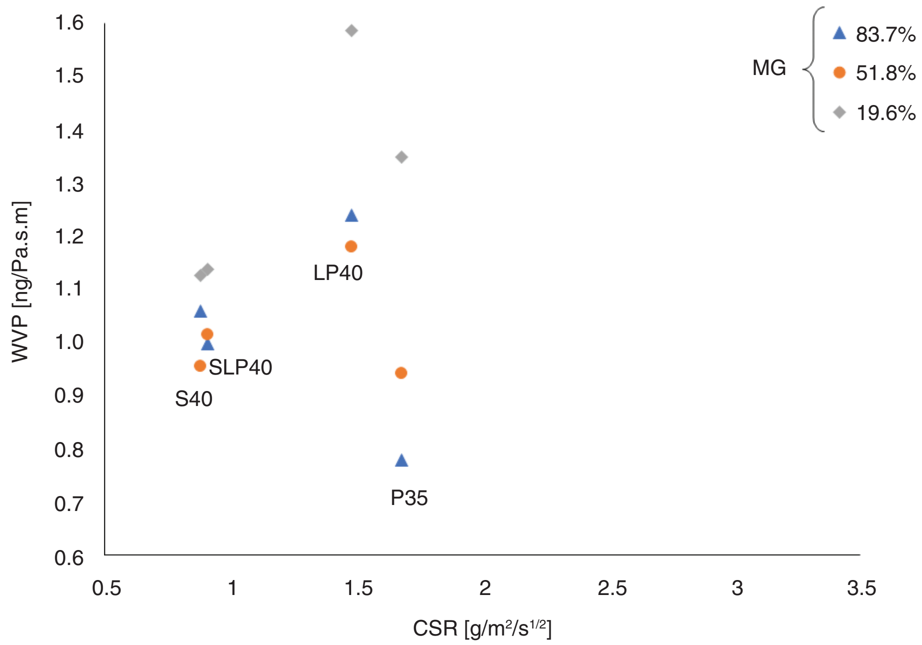

FIG. 10. Relationship between WVP and CSR for the different SCMs and MGs studied

studied GGBFS, which compensates for the dilution effect of LP. Accordingly, for the concrete incorporating LP only, the WVP increased for all cases, as a consequence of the dilution effect that increases the effective water to cement ratio.

The difference between the effects of OPC and pozzolanic Portland cement on WVP was not significant for the low w/cm ratio investigated. It should be noted that in this case, the replacement ratio was higher than for the case of substitution by GGBFS, LP, or both, resulting in less clinker content in the concrete mix.

A linear trend between CSR and WVP for OPC concrete was found, with w/cm having proportional effects on both properties. However, this tendency was not replicated when SCMs were used. Since

SCMs act by shifting the pore size distribution to smaller ranges, it can be concluded that WVP is strongly influenced by a different range of pore sizes than those affecting CSR.

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