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The permeability of Portland limestone cement concrete

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Abstract

The effect of limestone addition on the air permeability, water permeability, sorptivity, and porosity of limestone cement concrete has been investigated. Six Portland limestone cements (PLCs) with different limestone content (10–35% w/w) were produced by intergrinding clinker, gypsum, and limestone. A water-to-cement ratio (w/c) of 0.70–0.62—depending on the cement strength class—was used to prepare concrete of the compressive strength class C20/25 of EN 206-1. A modified commercial triaxial cell for 100-mm-diameter samples was used for the determination of the gas (N_2) and the water permeability of concretes. In addition, the sorptivity and porosity of the samples were measured, while thin sections of the concrete specimens were examined by means of optical microscopy. It is concluded that the PLC concrete indicates competitive properties with the ordinary Portland cement (OPC) concrete. Furthermore, the limestone addition has a positive effect on the water permeability and the sorptivity of concrete.

Keywords: Concrete; Blended cement; Limestone; Permeability; Adsorption

1. Introduction

The use of Portland limestone cements (PLCs) seems to have many benefits, both technical and economical [1–3]. In addition, the European Standard EN 197-1 identifies four types of PLC containing 6–20% limestone (types II/A-L and II/A-LL) and 21–35% limestone (types II/B-L and II/B-LL), respectively [4]. It is expected that the future world production of PLC will be continuously increased, but the wide use of limestone cement requires a thorough knowledge of the cement and concrete properties.

As far as the PLC is concerned, the research work is focused on three areas. The first one is the effect of limestone on the cement performance [5–8]. The second one deals with the participation of limestone in the hydration reactions of clinker [9–19], while the third one concerns the production process and specifically the intergrinding of clinker and limestone [20–22]. Although there is a disagreement in many partial topics, the knowledge level is satisfactory and continuously extended.

Concerning the PLC concrete, the few available references are focused on the effect of limestone on the concrete properties and behavior [23–30].

The recent years, there is a much interest in the "thaumasite form of sulfate attack" (TSA) correlated with the use of PLC and calcareous aggregates. Recent research work shows that PLC is susceptible to the thaumasite formation, due to sulfate attack at 5 °C, after only a few months exposure to sulfate solutions [31–37].

The present work deals with the concrete permeability, which is one of the most important parameters influencing the durability of concrete and finally its performance. In this paper, the effect of limestone content on the gas permeability, water permeability, sorptivity, and porosity of limestone cement concrete is investigated. This work is a part of a project, developed in our laboratories, concerning the properties of PLC and concrete.

2. Experimental

2.1. Materials and cement production

The chemical and mineralogical composition (Bogue) of the used clinker is shown in Table 1. The chemical analysis

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Table 1
Chemical and mineralogical composition and moduli of clinker

Chemical composition (% w/w)	
SiO_2	21.96
Al_2O_3	5.15
Fe_2O_3	3.78
CaO	65.95
MgO	1.76
K_2O	0.56
Na_2O	0.12
SO_3	0.52
Mineralogical composition (% w/w)	
C_3S	61.59
C_2S	16.48
C_3A	7.27
C_4AF	11.50
Moduli	
Lime saturation factor (LSF)	94.20
Silica ratio (SR)	2.46
Alumina ratio (AR)	1.36
Hydraulic modulus (HM)	2.14

of the limestone is given in Table 2. The limestone meets the requirements of EN 197-1 and belongs to the type LL. The main constituent of the limestone is calcite, while dolomite and quartz are also identified (X-ray diffraction) as minor constituents.

The PLC have been produced by intergrinding clinker, limestone, and gypsum (5% per clinker weight) in a pro-pilot plant ball mill of 5 kg capacity. The codes of the samples as well as their properties are given in Table 3. The cements LC1–LC4 contain 0%, 10%, 15%, and 20% limestone, respectively, and have similar 28-day compressive strength (48–51 N/mm^2, strength class 42.5R of EN 197-1). The cements LC5–LC7 contain 20%, 25%, and 35% limestone, respectively (33–40 N/mm^2, strength class 32.5R of EN 197-1). The compressive strength of the cements was measured according the EN 196-1.

Table 2
Chemical composition and characteristics of limestone

Chemical composition (% w/w)	
SiO_2	0.55
Al_2O_3	0.40
Fe_2O_3	0.17
CaO	53.47
MgO	1.02
K_2O	0.03
Na_2O	0.01
LOI	43.13
*Chemical characteristics**	
$CaCO_3$ (%)	95.5
MBA (g/100g)	0.1
TOC (%)	–

*According the EN 197-1.

Table 3
Characteristics of the tested cements

Sample	Composition (%)		Specific surface (m^2/g)	Compressive strength (N/mm^2)			
	Clinker	Limestone		1 day	2 days	7 days	28 days
LC1	100	0	260	11.9	21.3	35.3	51.1
LC2	90	10	340	11.2	20.9	36.3	47.9
LC3	85	15	366	12.9	22.7	37.7	48.5
LC4	80	20	470	14.9	24.3	38.0	48.1
LC5	80	20	325	7.6	17.2	28.1	39.8
LC6	75	25	380	9.7	17.8	31.4	40.0
LC7	65	35	530	9.8	17.0	26.2	32.9

2.2. Preparation of specimens

The mix proportions are given in Table 4, while the aggregate grading is presented in Table 5 (maximum aggregate size: 16 mm). The water-to-cement ratio (w/c: 0.70, 0.62), as well as the cement content (270 and 330 kg/m^3) of concrete was selected to be similar with the used in field constructions to have concrete of the compressive strength class of C20/25 (EN 206-1), which is widely used in Greece.

The properties of fresh concrete are given in Table 6. The slump of the mixes was in the range of 110–130 mm (class S3 of EN 206-1), using plasticizer where needed (LC7, Pozzolith 390N). The density of the fresh concrete varies from 2390 to 2417 kg/m^3.

The concrete compressive strength at 28 days is given in Table 6. The concrete LC1 belongs to the compressive strength class C25/30 of EN 206-1, while concretes LC2–LC7 belong to the compressive strength class C20/25 of EN 206-1.

The concrete specimens were cast in cylindrical steel molds of 100 mm diameter and 200 mm height. The specimens remained in the molds for 24 h and then they were demolded and placed in water in the curing room ($T = 20 \pm 2$ °C) for 27 days.

The air permeability tests were applied to a concrete cylinder of 100 mm diameter and height varied between 45 and 50 mm. According to literature, the samples were oven-dried at 105 °C, until a weight change of less than 0.1% over

Table 4
Concrete mix proportions

Sample	w/c	Concrete composition (kg/m^3)				
		Cement	Water	Fine Aggregate	Medium Aggregate	Plasticizer
LC1	0.70	270	189	912	1028	–
LC2	0.70	270	189	912	1028	–
LC3	0.70	270	189	912	1028	–
LC4	0.70	270	189	912	1028	–
LC5	0.62	330	205	888	1002	–
LC6	0.62	330	205	888	1002	–
LC7	0.62	330	205	888	1002	2.4

Table 5
Aggregate grading

Sieve size (mm)	Cumulative passing (% w/w)	
	LC1–LC4	LC5–LC7
0.25	11	9
0.50	19	18
1.0	32	32
2.0	40	40
4.0	52	52
8.0	70	70
16.0	100	100

24 h was observed [38]. The drying period of the specimens was 3–5 days.

The water permeability tests were applied to a concrete cylinder of 100 mm diameter and height varied between 45 and 50 mm. To saturate the samples, they were boiled in deionized water for a period of about 5 h prior to testing, subsequently left to attain room temperature, and immersed in the same deionized water in which they were boiled. The saturation of the samples with water is considered necessary to eliminate the presence of gas bubbles in the samples' pores and decrease the duration of the water permeability test [38,39].

The sorptivity was measured in a concrete cylinder of 100 mm diameter and 125 mm height, oven-dried at 105 °C for 24 h.

2.3. Measuring techniques

A modified commercial triaxial cell for 100-mm-diameter samples, operating to maximum cell pressure of 1.7 N/mm^2, was used for the determination of the gas (N_2) permeability of the specimens. The outline of this device is presented in Fig. 1, while the detailed procedure is described in a previous work [28]. The gas permeability measurements were carried out for inlet pressure (P_1) of 0.3, 0.5, 0.7, 0.9, and 1.1 N/mm^2, outlet pressure (P_2) of 0.1 N/mm^2, and cell pressure of 1.3 N/mm^2. The value for the intrinsic permeability (K) was calculated at each different inlet pressure according to the following Eq. (1) [38–40]:

$$K = \frac{2QLP_2\eta}{A(P_1^2 - P_2^2)} \quad (1)$$

where K is the intrinsic permeability (m^2), Q is the rate of flow of gas (m^3/s), L is the specimen thickness (m), A is the cross-sectional area of the specimen (m^2), η is the dynamic viscosity of gas (N s/m^2), and P_1 and P_2 are the inlet and outlet pressure, respectively (N/m^2).

Due to gas slippage, it is not accurate to determine the intrinsic permeability as the average of the calculated values of K at the different inlet pressure values. Therefore, a method mentioned in the literature [28,38,39] was applied to correct the taken values of gas permeability. Using regression analysis, the relation between the gas permeability and the inverse of mean pressure ($1/P_m$, where $P_m = (P_1 + P_2)/2$) was determined. The coefficient b of the resulted equation $K = a(1/P_m) + b$ gives the correct value of the gas intrinsic permeability (K_g).

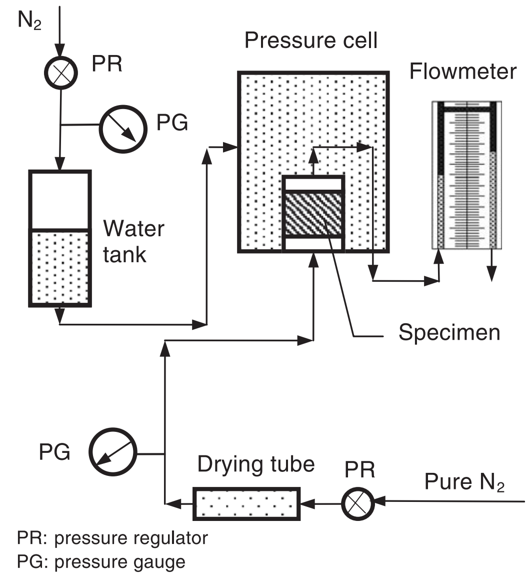

Fig. 1. Outline of the device used for the measurement of gas permeability.

The same triaxial cell was used for the determination of the water permeability of the specimens. The outline of this device is presented in Fig. 2. The water permeability measurements were carried out for inlet pressure of 0.5 N/mm^2, outlet pressure of 0.1 N/mm^2, and cell pressure of 0.7 N/mm^2. Water permeability (coefficient of permeability) K_w was determined according to Eqs. (2) and (3) [38–40], when the steady-state flow was attained (water flow rate at inlet equals water flow rate at outlet). Depending on the samples, the test duration was 5–7 days.

$$K = \frac{QL\eta}{A\Delta h\rho g} \quad (2)$$

where K is the water permeability (intrinsic or absolute permeability) (m^2), Q is the rate of flow of water (m^3/s), L is

Table 6
Concrete properties

Sample	Fresh concrete			Hardened concrete
	Slump (mm)	Flow (mm)	Unit weight (kg/m^3)	Compressive strength at 28 days (N/mm^2)
LC1	130	460	2400	31.9
LC2	120	440	2395	27.4
LC3	120	420	2400	27.3
LC4	110	420	2394	28.0
LC5	120	440	2417	28.2
LC6	110	420	2410	26.5
LC7	110	400	2390	26.6

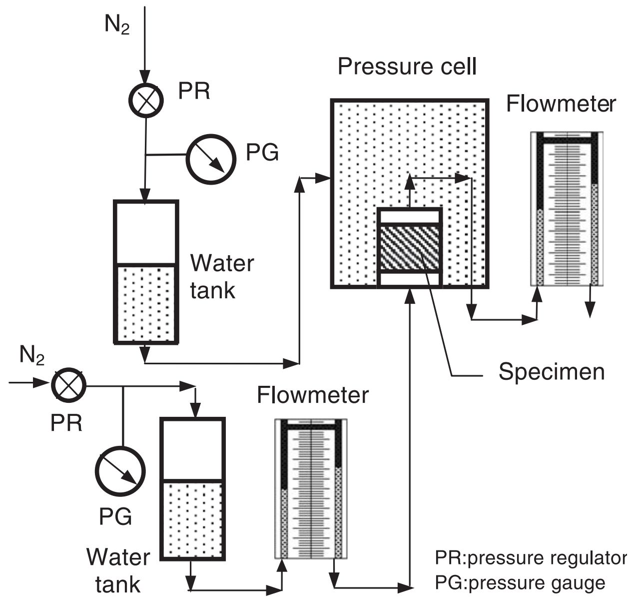

Fig. 2. Outline of the device used for the measurement of water permeability.

the specimen thickness (m), η is the dynamic viscosity of water (N s/m^2), A is the cross-sectional area of the specimen (m^2), Δh is the drop in hydraulic head through the specimen (m), ρ is the density of the water (kg/m^3), and g is the acceleration due to gravity (m/s^2).

$$K_w = \frac{K\rho g}{\eta} \quad (3)$$

where K_w is the water permeability (coefficient of permeability) (m/s).

A schematic diagram of water absorption test is given in Fig. 3. The lower part of the sides of the specimen adjoining the inflow face was sealed with an adhesive tape. The specimen was rested on rods to allow free access of water to the inflow surface and the tap water level was kept not more than 5 mm above the base of the specimen. The quantity of the absorbed water was measured at 10, 20, 30, 45, and 60 min after starting the test. Since the cumulative water absorption per unit area of the inflow surface (i)

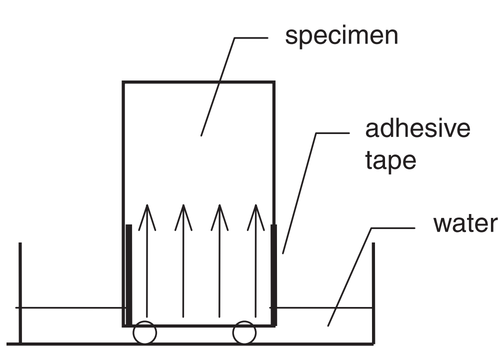

Fig. 3. Schematic diagram of water absorption test.

increases in relation to the square root of elapsed time (t), the sorptivity S is defined as the slope of the i against $t^{0.5}$ plot and is determined using regression analysis [29,40].

The total porosity of the specimen was measured with a Carlo Erba Hg porosimeter.

Thin sections of the concrete samples were examined by means of optical microscopy using transmitted ordinary and crossed polarized light. This technique has been used to examine the homogeneity of the cement paste, the porosity of the samples, and the aggregate–paste interface zone.

3. Results and discussion

Table 7 presents the gas permeability (K_g), the water permeability (K_w), the sorptivity (S), and the total porosity (P) of the tested specimens. The results given in Table 7 are the average values of three different specimens. Fig. 4 shows the effect of the PLC on the studied concrete properties. In the same figure, the concrete compressive strength after 28 days, the cement type, and the strength class are also presented.

In general, PLC concretes exhibit higher gas permeability values (with the exception of LC7) compared with the OPC concrete. It must be noted that these specimens also showed lower compressive strength (Fig. 4). The gas permeability of the PLC concrete varies from 2.65×10^{-17} to 3.03×10^{-17} m^2, while the OPC concrete presents a gas permeability of 2.26×10^{-17} m^2. The concrete with PLC containing 35% limestone shows the lower gas permeability.

On the contrary, concrete with PLC exhibits lower water permeability values, compared with the OPC concrete. The water permeability of the PLC concrete varies from 1.81 to 2.30×10^{-11} m/s, while the OPC concrete presents a water permeability of 2.39×10^{-11} m/s. The concrete with PLC containing 20% limestone (LC5) shows the lower water permeability. The limestone addition seems to affect in a positive way the water permeability of concrete.

PLC concrete exhibits slightly lower sorptivity values, compared with the OPC concrete. The sorptivity varies from 0.220 to 0.238 mm/min$^{0.5}$, while the OPC concrete presents a sorptivity of 0.237 mm/min$^{0.5}$. The limestone addition seems to affect in a positive way the water absorption of concrete. It must be noted that the estimated sorptivity

Table 7
Gas permeability, water permeability, sorptivity, and porosity of the tested samples

Code	K_g ($\times 10^{-17}$ m^2)	K_w ($\times 10^{-11}$ m/s)	S (mm/min$^{0.5}$)	P (%)
LC1	2.26	2.39	0.237	12.48
LC2	2.65	2.30	0.238	12.30
LC3	2.80	2.22	0.226	12.31
LC4	2.95	2.00	0.220	13.14
LC5	3.03	1.81	0.228	12.94
LC6	2.82	2.07	0.229	13.62
LC7	2.10	2.23	0.224	14.64

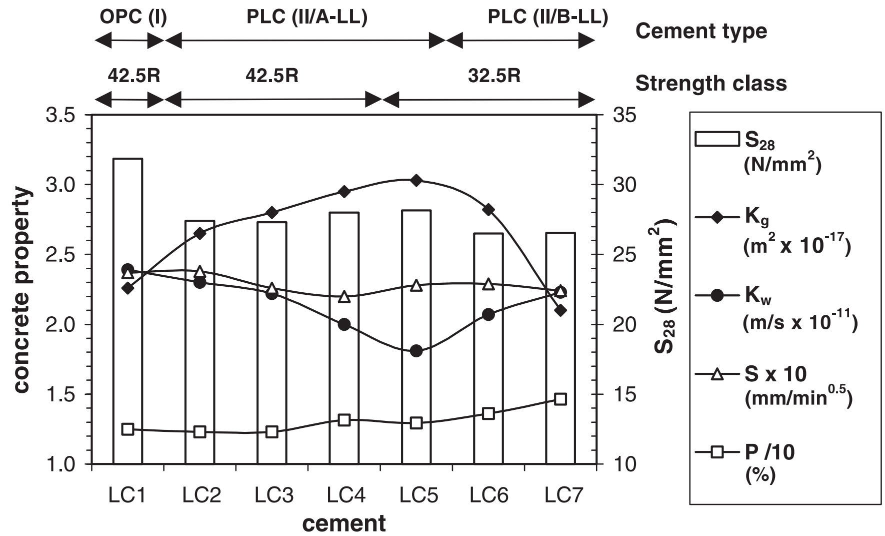

Fig. 4. Effect of PLC on concrete properties (S_{28} = compressive strength at 28 days; K_g = gas permeability; K_w = water permeability; S = sorptivity; P = total porosity).

coefficient only evaluates the large capillary pores, due to the high w/c of the studied concretes.

Concrete, based on PLC that contain up to 15% w/w limestone, has the same porosity as the OPC concrete. Further increase of the limestone content causes a relative increase of the concrete porosity.

To indicate more clearly the contribution of the limestone to the concrete properties, the ratio (p/p_0) has been used, where p is the value of a specific property of PLC concrete and p_0 is the value of the same property in OPC concrete. Therefore, (p/p_0) values less than 1 indicate that limestone favors the concrete property and improves the concrete performance. Fig. 5 illustrates the mean value and the deviation of (p/p_0) for the measured concrete properties. The (p/p_0) values presented in Fig. 5 are the average values of the six PLC concretes studied (LC2–LC7). Although the PLC concretes properties are close to the OPC concrete properties, it is seen, for the w/c used, that the limestone

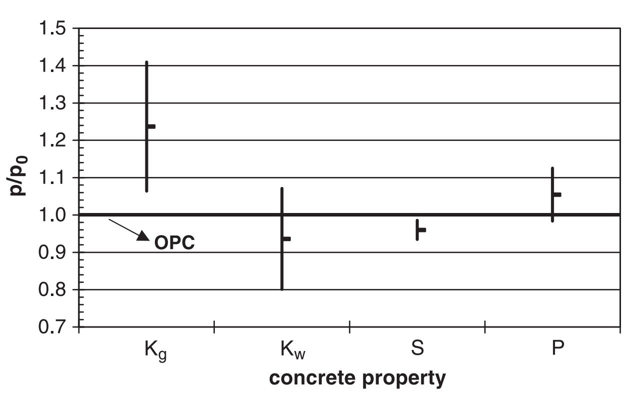

Fig. 5. Mean value and deviation of PLC concrete properties compared to OPC concrete properties (p = value of a specific property of PLC concrete; p_0 = value of the same property in OPC concrete; K_g = gas permeability; K_w = water permeability; S = sorptivity; P = porosity).

addition has a positive effect on the water permeability and sorptivity, while the gas permeability and the porosity of concrete are increased.

The above results show that the limestone affects the water permeability and sorptivity in a different way than the gas permeability and porosity. The permeability of concrete is not a simple function of its porosity, but depends also on the size, distribution, shape, tortuosity, and continuity of the pores. In addition, the cement particle size distribution affects the concrete permeability [40]. In the case of PLC, which has a higher fineness and a lower clinker content than OPC, the factors that affect the pore system of concrete are (a) the filler effect and (b) the (clinker) dilution effect. The above two factors influence the total volume and size distribution of pores and finally affect the concrete permeability. Our results show that gas permeability is closely related with the porosity. On the other hand, water permeability and sorptivity seem to be affected by the size and kind of pores. In any case, further investigation is needed.

The samples were examined under an optical microscopy to have an overview of the concrete structure. Both ordinary and crossed polarized light was used. The images were selected to be representative as far as the condition of the cement paste and the interface of paste–aggregate are concerned. There was not any considerable differentiation among the samples. In all samples, the cement paste appears to be uniform in color. It indicates that water is well distributed resulting in a homogeneous mixture. Besides, the entrapped air is uniformly distributed in the form of small rounded bubbles. Finally, neither unusual cracks nor excessive entrapped air voids were observed in any sample. Special attention was paid on the aggregate surface but we did not observe any precipitation of secondary hydration products. Such precipitation has been previously reported in

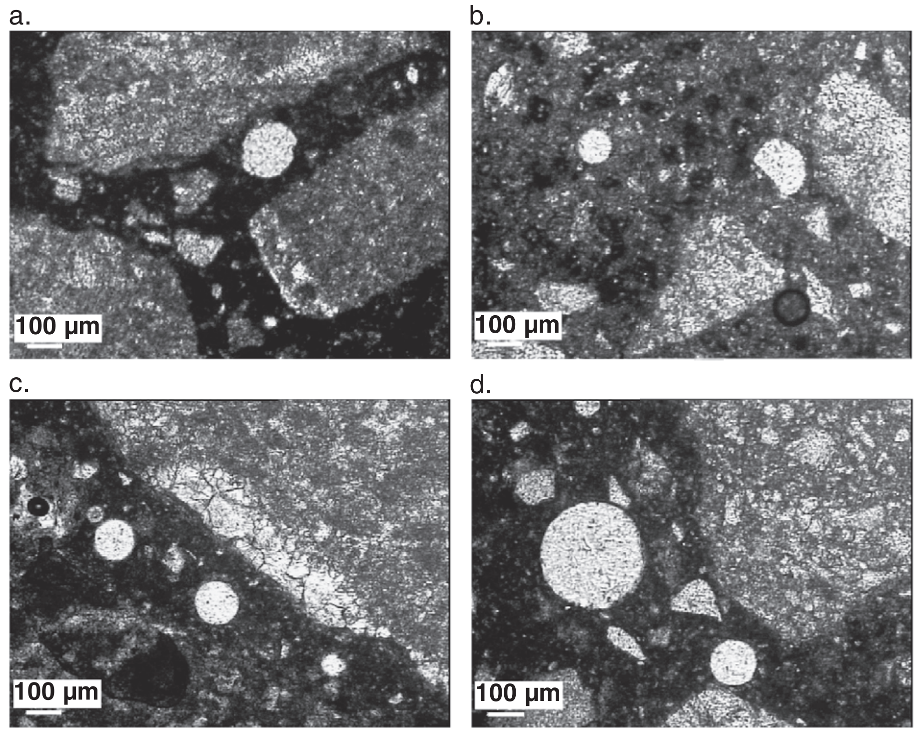

Fig. 6. Thin sections of concrete samples: (a) LC1, (b) LC2, (c) LC4, and (d) LC7.

limestone concrete based on high C_3A clinker [28]. In Fig. 6, pictures from samples LC1, LC2, LC4, and LC7 are presented.

Reviewing all the above measurements and although that some phenomena need more thorough investigation, the final sense is that, in the case of high w/c ratio, the presence of limestone does not affect considerably the permeability characteristics of the concrete. In addition, our previous studies have shown that PLC have many benefits concerning their mechanical and physical properties as well as the workability characteristics and the strength development of concrete [8,17,18,28–30]. It is concluded that concrete based on PLC (type II/A-LL and II/B-LL, strength class 42.5R and 32.5R-EN 197-1) has a satisfactory quality and competitive properties with OPC concrete. At the same time, all the technical and economical benefits, coming from the use of an abundant and not expensive mineral as a cement main constituent, are fully exploited.

4. Conclusions

The following conclusions can be drawn from the present study:

- ◆ PLC concrete indicates competitive properties with the OPC concrete.
- ◆ Concrete, based on PLC, exhibits higher gas permeability values than the OPC concrete.
- ◆ The limestone addition has a positive effect on the water permeability and the sorptivity of concrete.
- ◆ Limestone content up to 15% w/w does not alter the concrete porosity.

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