Kinetics of the water absorption in GGBS-concretes: A capillary-diffusive model

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Abstract. We study the kinetics of absorption of water in Portland cement concretes added with 60, 70 and 80% of granulated blast furnace slag (GGBS) cured in water and at open air and preheated at 50 and 100ºC. A mathematical model is presented that allows describing the process not only in early ages where the capillary sorption is predominant but also for later and long times where the diffusive processes through the finer and gel pores are considered. The fitting of the model by computerized methods enables us to determine the parameters that characterize the process: i.e., the sorptivity coefficient (S) and diffusion coefficient (D). This allows the description of the process for all times and offers the possibility to know the contributions of both, the diffusive and capillary processes. The results show the influence of the curing regime and the preheating temperature on the behavior of GGBS mortars.

1. Introduction

The role of water in the mechanism of deterioration of porous building materials has been recognized for a long time. The conjugate action of mechanical and chemical damages arising for instance from freeze/thaw cycles, shrinkage-induced cracking, salt crystallization and leaching (Addleson 1972) may lead to rapid degradation of the bulk mechanical properties of building materials and significantly reduce the service life of constructions. The durability of building structures is thus critically determined by the rate at which water infiltrate and move through the porous structure. In addition the penetration of water in building materials provides a mechanism and path for the penetration of deleterious materials like chloride and sulfate ions which can lead to the corrosion of steel reinforcement in concretes (Thoft-Christensen 2001, Mejlhede, *et al.* 1999, Thoft-Christensen 2002, Luping 1996, Mironova, *et al.* 2002). Sorptivity and diffusion are the primary mechanisms that control the transport of deleterious materials inside of the concrete (Martys and Ferraris 1997). The diffusion corresponds to the ionic movement under the action of a concentration gradient (it occurs fundamentally when the material is dampened) and sorptivity is the property that characterizes the

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disposition of a porous material to absorb and to transmit water by capillarity.

Because of its economic implications, the problem of water movements in porous building materials has received great attention in the past and also at present. A better understanding of the water absorption and moisture transfer can therefore reduce or prevent damage in building materials. For that reason these phenomena are of interest to physicist and engineers.

This phenomenon has been the object of intensive research (Martys and Ferraris 1997, Hall and Kam 1986, Ho and Lewis 1987, Hall and Yau 1987, Hall 1989, Sabir, *et al.* 1998, Ohdaira and Mazuzawa 2000, Goual, *et al.* 2000, McCarter, *et al.* 2001a, McCarter, *et al.* 2001b) where different methods of measuring the uptake and movement of water in these materials (gravimetry, microwave method, electrical conductivity, relative humidity, ultrasonic method, nuclear magnetic resonance, etc) have been used.

It is known that the use of finely divided siliceous materials (such as fly ash, ground granulated blast furnace (GGBS or slag), rice husk ash, silica fume, etc) in concrete is the most direct, technically feasible and economically attractive solution to the problem of reinforced concrete durability.

In particular, these materials greatly improve the durability of concrete through control of high thermal gradients, pore refinement, depletion of cement alkalis and the capability for continued long-term hydration or pozzolanic reaction (Swamy 1997). In developing countries where there is increasing demand for construction, the technical advantages of pozzolans and slag in concrete are complemented by other economic, ecological and environmental considerations. These materials reduce substantially not only the energy consumption in the production of concrete, but also help to reduce environmental pollution through reduced emission of carbon dioxide.

Slag is a by-product from the production of steel. During production, liquid slag is rapidly quenched from a high temperature by immersion in water (Metha 1993). The slag is a glassy, granular, non-metallic product that consists essentially of silicates and aluminosilicates of calcium and other bases (Klieger and Joseph 1994).

The addition of slag (GGBS) diminishes the large pores and reduces porosity. It is due to the hydration of slag (Metha 1983) which reacts with calcium hydroxide (originated during the hydration of Portland cement) to form calcium silicate hydrate (CSH) gel with very small pore size. Thereby the slag increases the strength of the concrete product in comparison with the concrete based on ordinary Portland cement OPC (Kalid, *et al.* 2002, Mejía, *et al.* 1997, Mejía and Talero 1996, Thomas and Bamforth 1999).

Ho, *et al.* (1986) showed that concrete quality with response to interrupted curing is improved by the incorporation of slag in concrete. The rate of water absorption by capillary action is significantly decreased when slag is used. GGBS concrete is substantially more resistant to chloride diffusion than Portland cement concrete of the same strength grade. The uses of slag will considerably increase the service life of structures exposed to chloride environment and it wills sufficient cover to the steel reinforcement. Swamy (1997) has presented a critical and valuable evaluation about the use of slag in concrete.

From scientific point of view and for numerous applications, simple but accurate models describing the water transport in mortars and concretes are important. Several models (which involve both analytical and numerical models) have been developed for describing this phenomenon in mortars and concretes without and with additions (Martys 1997, Hall and Yau 1987, Küntz and Lavallé 2001, Holm and Künzel 2000, Martys 1999, Claisse, *et al.* 1997, Wirquin, *et al.* 2000), but they do not always agree with the experimental results. Several researchers describe the water absorption in mortars and concretes in early ages only where the process of capillary absorption

prevails but they dont describe the complete process where the moisture diffusion through the finer or gel pores may be considered. These finer pores increase its importance with time (Martys 1997). The modeling of water absorption in mortars and concretes that involves both capillary and diffusive processes and allows for the computing of the kinetic coefficients of the process is scarce. For that reason, a model that considers both the capillary and diffusion phenomenon and allows us to explain the process for all ages will be of great importance.

The goal of the present paper is to develop a mathematical model for describing the water transport in GGBS concretes. This model allows us to describe the process not only in early ages where the capillary sorption prevails but also for large times where the diffusive processes through the finer and gel pores may be considered. This gives the possibility to know the contributions of the diffusion and capillary action in the water transport in concretes, which is very useful to predict the penetration of chloride or other ions that deteriorate the steel reinforcement in concretes. The fitting of the model by computerized methods (by applying non-linear regression techniques fundamentally) lets us determine the kinetic parameters, such as sorptivity coefficient and diffusion coefficient. It allows us to characterize the kinetics of the water transport in GGBS concretes with 60, 70 and 80% of slag. Besides, the influence on the water sorption process of different curing regimes (curing in water and at open air) as well as the effect of changing the preheating temperature is analyzed.

2. Experimental

The experimental data presented in this paper were obtained by Mejías and Talero (1996, 1997).

The samples used correspond to cylindrical mortars (with a diameter and height of 50.8 and 50 mm respectively) manufactured with OPC (Table 1), sand, and GGBS additions of 60, 70 and 80% by weight. The chemical composition of GGBS showed high contents of silica and aluminum oxides above 50%, followed of calcium, magnesium and iron about 42%. Further details are in reference (Mejía, *et al.* 1997).

A curing under water and at open air by 15 days was used and a posterior preconditioning of the samples at 100ºC during 6 h and cooling in desiccator for 24 h was applied. Other samples cured

Table 1 Chemical composition of the OPC

Cement	SC_3	SC_2	AC_3	FAC_4
OPC	46.95	19.98	14.69	5.33

Table 2 General data for the samples

Sample designations	Compositions	Curing regime (15 days)	Preconditioning temperature
S80W100	OPC+80% slag	Water	100ºC
S70W100	OPC+70% slag	Water	100ºC
S60W100	OPC+60% slag	Water	100ºC
S80W50	OPC+80% slag	Water	50ºC
S80A100	OPC+80% slag	Air	100ºC

under water were dried at 50ºC during 48 h and cooled in desiccator. Table 2 shows the general data for the samples.

One of the ends of the samples was placed on a thin sponge (3 mm) to absorb water. The other faces were sealed with an impermeable organic substance. The weight of the specimens was measured intermittently for about two weeks. Figs. 1 and 2 show a scheme of the sorption test and the cumulative water absorption versus square root of time respectively.

3. Formulation of the problem

The process of water transport in concrete has been studied by many researchers. In most of the cases only the process for early ages in dry samples and without additions has been explained satisfactorily. It is frequently found that if a mortar or concrete surface is exposed to wetting by water then the cumulative water absorption (normalized to the exposed surface area) M/A (in kg/m^2) is proportional, during the initial absorption period, to the square root of elapsed wetting time t: (Hall 1989)
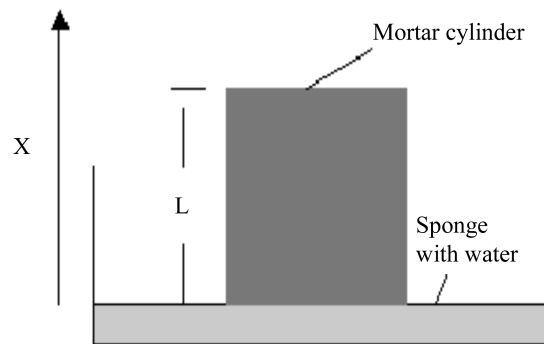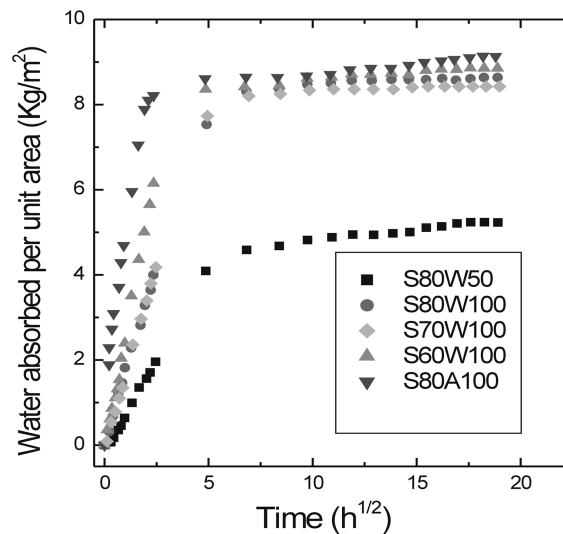

Fig. 1 Scheme of the sorption test

Fig. 2 Water absorbed for unit area for concretes added with 60, 70 and 80% GGBS

$$\frac{M}{A} = St^{1/2} \tag{1}$$

where S is the sorptivity coefficient measured in $kg/(m^2 \cdot h^{1/2})$. It is easily determined from the slope of the linear part of the M/A versus $t^{1/2}$ curve.

After a period of sorption the initial rate of ingress observed decreases as the water has accessed all the larger capillary pores. The decrease in gradient of the straight line portion of the water uptake versus square root of time indicates that sorptivity is now occurring via the finer pores and indicates the increasing importance of small pores with time (Martys and Ferraris 1997). Besides, some materials with extremely coarse pore structure experience little capillary suction and may show significant deviation from linearity after prolonged wetting.

It is widely known that in dry or partially dry mortars or concretes the predominant mechanism in the water absorption is the capillary suction (through the capillary pores) and when the time lapses, and the material begins to be saturated, the predominant mechanism is diffusion (through the finer and gel pores) (Martys and Ferraris 1997, Xi, *et al.* 1994).

However, both processes should coexist simultaneously from early ages (a capillary process through the big capillary pores and a diffusive process through the small pores and gel pores) mainly in GGBS concretes where the quantity of gel pores increases and the big capillary pores decrease considerably.

To model the capillary and diffusive processes we consider that the cumulative water absorption with time can be expressed in general as:

$$\frac{M}{A} = St^{1/2} + \text{diffusive term} \tag{2}$$

the first term on the right can be written as (Martys and Ferraris 1997):

$$N\rho \left\{ 1 - \exp\left(-\frac{St^{1/2}}{N\rho}\right) \right\} \tag{3}$$

where N is a constant related to the distance from the concrete surface over which capillary pores control the initial sorption and ρ is density of the water. This term was constructed so that at early ages, in the limit $t^{1/2} < N\rho / S$, expansion of the exponential gives $N\rho \left\{ 1 - \exp\left(-\frac{St^{1/2}}{N\rho}\right) \right\} \approx St^{1/2}$ which is the same as Eq. (1).

The diffusive term in Eq. (2) is obtained considering the physical diffusion of water and solving the continuity equation or second Fick's law, in non-stationary states with the initial and boundary conditions imposed by the given situation. The samples are permeable cylindrical mortars with zero water (considering the water used in the test) as initial condition. In our case (as above mentioned) all the faces were sealed except the surface in contact with the water. This geometry (Fig. 1) simplifies the mathematical problem to determine the diffusion coefficient, which not depends on the geometry and boundary conditions. The diffusion coefficient (D) is more likely governed by the nature of the substance and the characteristics of the concrete. Accordingly, the diffusion problem is reduced to a unidirectional treatment where the diffusion coefficient D is assumed constant for each test. The mathematical problem is given by the equation:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \tag{4}$$

where C is the water concentration and t the time.

The initial and boundary conditions are given by:

$$C(x, 0) = 0 \quad \forall x \in (0, L) \tag{5}$$

$$C(0, t) = Co \quad \forall t \tag{6}$$

$$\frac{\partial C}{\partial x} = 0 \quad \text{in } x = L, \ \forall t \tag{7}$$

The first boundary condition (Eq. 6) shows the invariance of water concentration (Co) in the surface for each sample.

The second boundary condition (Eq. 7) expresses the impermeability of the top of the sample in $x=L$.

In order to solve the boundary problem the Fourier method (Jost 1960, Carslaw and Jaeger 1970) or the method of Laplace Transform (Cranck 1975) can be used. The result is:

$$C = Co\left\{1 - \frac{4}{\pi}\sum_{n=0}\frac{1}{(2n+1)}\exp\left[-\frac{(2n+1)^2}{4L^2}\pi^2 Dt\right]\sin\frac{(2n+1)}{2L}\pi x\right\} \tag{8}$$

This solution expresses the water concentration profiles along the (x) coordinate and the (t) time in all the section of the sample.

The amount of water absorbed through the area A of the permeable surface is:

$$M = \iiint_V C(x, t)dxdydz = A\int_0^L C(x, t)dx \tag{9}$$

$$M = ACoL\left\{1 - \frac{8}{\pi^2}\sum_{n=0}\frac{1}{(2n+1)^2}\exp\left[-\frac{(2n+1)^2\pi^2}{4L^2}Dt\right]\right\} \tag{10}$$

Therefore the amount per unit of area expressed in kg/m^2 is given by:

$$\frac{M}{A} = CoL\left\{1 - \frac{8}{\pi^2}\sum_{n=0}\frac{1}{(2n+1)^2}\exp\left[-\frac{(2n+1)^2\pi^2}{4L^2}Dt\right]\right\} \tag{11}$$

substituting the Eq. (11) as second term in Eq. (2), we obtain:

$$\frac{M}{A} = C\rho\left\{1 - \exp\left(\frac{-St^{1/2}}{C\rho}\right)\right\} +$$

$$CoL\left\{1 - \frac{8}{\pi^2}\sum_{n=0}\frac{1}{(2n+1)^2}\exp\left[-\frac{(2n+1)^2\pi^2}{4L^2}Dt\right]\right\} \tag{12}$$

Eq. (12) represents a capillary-diffusive model. It expresses the mass of water per unit of area incorporated to the sample considering both, the capillary sorption through the large pores (first term) and water diffusion through the small pores and very finer gel pores (second term).

4. Results and discussion

Fig. 2 shows the experimental results of the sorption test for the samples S80W100, S70W100, S60W100, S80W50 and S80A100. A quick increase of the amount of water absorbed at early ages for all samples is obtained, showing the samples S80A100 and S80W50 the biggest and smaller rate of absorption at early ages respectively. Later on the initial rate of ingress observed decreases (the water has accessed all the large capillary pores) until a stabilization of the curve or equilibrium (saturation) is reached for large times in dependence of the analyzed sample.

The samples S80W100, S70W100 and S60W100 with 60, 70 and 80% of GGBS show a similar behavior. Big differences from the qualitative point of view are not obtained.

Qualitatively, a bigger rate of initial sorption is obtained for the sample S80A100, followed by S60W100, S70W100 and S80W100. The amount of incorporated water for large times (equilibrium) is very similar for these samples. On the other hand, the sample S80W50 shows the smaller rate of initial sorption and the smaller amount of incorporated water for large times.

For samples curing at open air (S80A100) a greater rate of water absorbed is qualitatively obtained in comparison with the samples (S80W100) curing under water. Also, the sample S80W50 preheating to 50°C shows a smaller sorptivity and a smaller amount of incorporated water at equilibrium in comparison with the sample S80W100 preheating to 100°C.

Figs. 3a, 3b, 3c, 4 and 5 present the water absorbed for unit area (M/A) versus time (*t*) for S80W100, S70W100, S60W100, S80W50 and S80A100 samples. Solid lines represent the curves of the fitted model. The fitting of the model (Eq. (12)) permitted us to determine the parameters *C, S, D* and *Co* in each case. The values of these parameters for all samples are given in Table 3.

In the figures the correlation (*r*) and multiple determination coefficients (R^2) are shown. These statistical parameters related with the good correspondence between the mathematical model and the experimental data, are calculated in the non-linear regression process (fitting process of the model).

According to the values of the coefficients *S* and *D* the samples S80W100, S70W100, S60W100
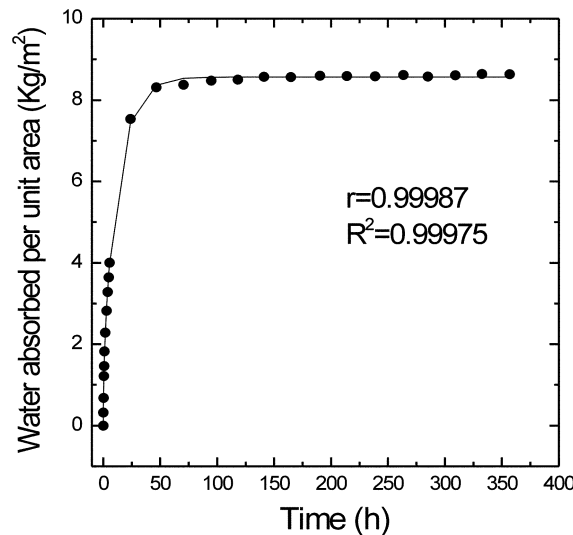

Fig. 3a Water absorbed for unit area for the sample S80W100 with 80% GGBS, curing in water and preconditioned at 100°C
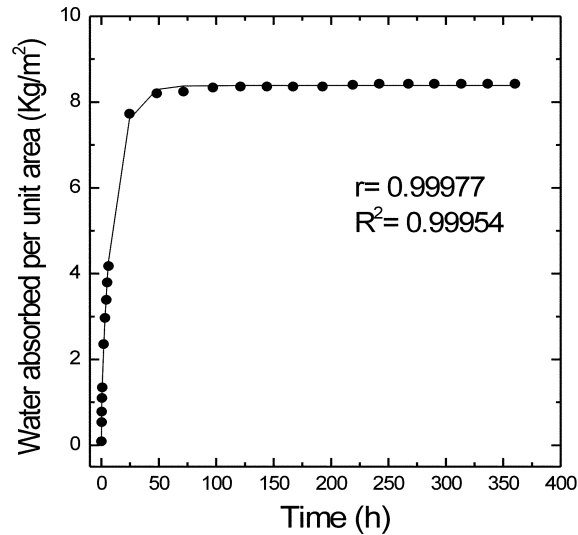
Fig. 3b Water absorbed for unit area for the sample S70W100 with 70% GGBS, curing in water and preconditioned at 100°C
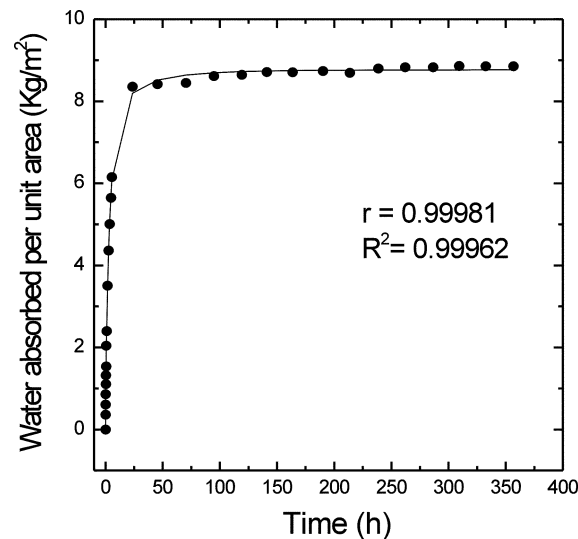

Fig. 3c Water absorbed for unit area for the sample S60W100 with 60% GGBS, curing in water and preconditioned at 100°C

with 60, 70 and 80% GGBS cured under water and preheating to 100°C show a similar behavior which agree with the qualitative analysis carried out previously. The sample S80W100 curing in water shows a less sorptivity and diffusion (less S and D) in comparison with S80A100 curing at open air and therefore a bigger opposition to the uptake and water transport in the concretes. It is due to the influence of the curing regime on the pore structure. The curing in water facilitates the refinement of the pore structure in GGBS mortars and therefore leads to high degree of
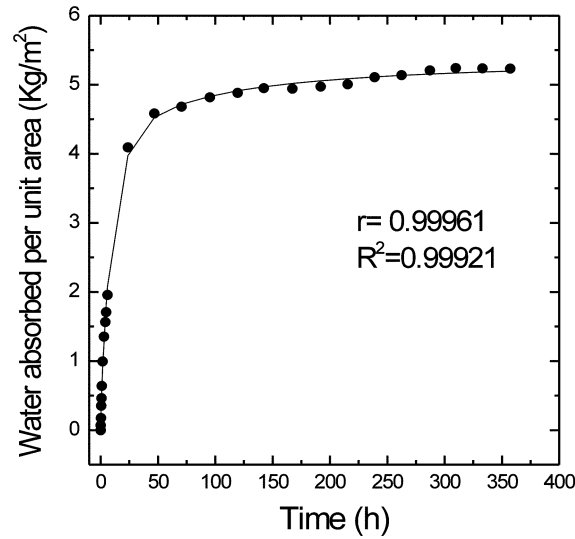
Fig. 4 Water absorbed for unit area for the sample S80W50 with 80% GGBS, curing in water and preconditioned at 50°C
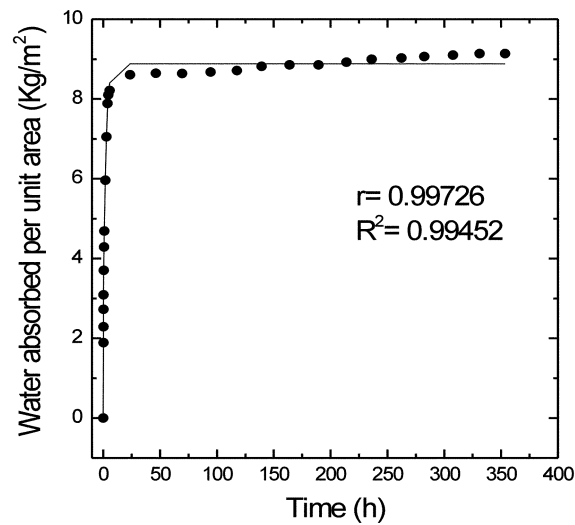

Fig. 5 Water absorbed for unit area for the sample S80A100 with 80% GGBS, curing at open air and preconditioned at 100°C

impermeability (Kalid, *et al.* 2002). In other words, the lack of curing in water in slag cement concretes affects adversely the development of a very finer pore structure. On the other hand curing at open air increases the absorption and porosity of slag cements (Kelham 1988, Dhir and Byars 1991).

The changes in the diffusion coefficients (*D*) show the role of the diffusive processes and the importance of its consideration as well as their dependence on the structure of pores affected by the different curing regimes.

Table 3 Values of the parameters for all samples

Mixture	S (Kg/m² s^{1/2})	D (m²/s)	Co (Kg/m³)	N (m)
S80W100	$3,16. 10^{-2}\pm0,25. 10^{-2}$	$1,38. 10^{-8}\pm0,08. 10^{-8}$	$140,52\pm5,42$	$1,51. 10^{-3}\pm0.14. 10^{-3}$
S70W100	$2,75. 10^{-2}\pm0,24. 10^{-2}$	$2,50. 10^{-8}\pm0,23. 10^{-8}$	$137,34\pm8,32$	$1,52. 10^{-3}\pm0.15. 10^{-3}$
S60W100	$2,81. 10^{-2}\pm0,15. 10^{-2}$	$6,10. 10^{-8}\pm0,27. 10^{-8}$	$94,62\pm3,86$	$4,02. 10^{-3}\pm0.20. 10^{-3}$
S80W50	$4,80. 10^{-3}\pm0,45. 10^{-3}$	$2,83. 10^{-8}\pm0,14. 10^{-8}$	$63.75\pm3,70$	$2,17. 10^{-3}\pm0.17. 10^{-3}$
S80A100	$21,31. 10^{-2}\pm2,1. 10^{-2}$	$1,28. 10^{-7}\pm0,08. 10^{-7}$	$121,52\pm6,29$	$2,75. 10^{-3}\pm0.26. 10^{-3}$

The results obtained for the samples S80W100 and S80W50 allows us to analyze the effect of the preconditioning temperature. A decrease of the sorptivity S (see Table 3) is obtained when diminish the preconditioning temperature. It is due fundamentally to strong role that plays the degree of saturation on the control of water uptake (Martys and Ferraris 1997).

Water flows through the big capillary pores by action of the capillary force, being the flow rate determined by the pore structure and the local content of water. Therefore the capillary forces are bigger in dry materials as S80W100 (preheating at 100⁰C) that in partially dry materials as S80W50 (preheating at 50ºC) and become minimum in completely saturated materials.

It should also be taken into account that for preconditioning temperatures around 100⁰C microcracking and other undesirable effects can be generated (Holm and Künzel 2000, Kelham 1988), which contributes to a bigger capillary suction.

The variation of the diffusion coefficients D with the preconditioning temperature is not significant; a little increase in the partially saturated sample S80W50 in comparison with the completely dry sample S80W100 is noticed.

5. Conclusions

1. The capillary-diffusive model proposed here describes the kinetics of the water absorption in GGBS concretes with enough accuracy. The kinetic coefficients such as sorptivity and diffusion coefficient should be previously determined in the fitting model process. The model describes the water absorption process for all times and allows us to know the contributions of the diffusion and capillary action in the water transport in GGBS cement concretes.

2. The variation of the % GGBS in the range 60-80% in materials with the same curing regime and preconditioning temperature doesn't influence considerably on the behavior of the materials. This is manifested in the little variation of the coefficients S and D.

3. The curing regime has important effects on the permeability of GGBS concretes. The samples cured in water show best characteristics of impermeability (less S and D) that the samples cured at open air.

4. The preconditioning temperature (for the same curing regime) is a factor that shows the influence of the saturation degree on the control of the water uptake. According to the S values, the samples preconditioned at 50⁰C show less sorptivity in comparison with the samples preconditioned at 100ºC.

5. The model proposed considers that the capillary sorption and diffusion are the principal mechanisms for the water absorption in GGBS cement concretes but it doesnt consider (for

simplicity and for having a less contribution) other mechanisms (such as permeation, osmosis, thermal migration, etc) that may be present in the uptake and movement of water in these materials. Future studies include the analysis of the possibility of existence (or not) of correlation effects between both capillary and diffusive process.

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