Limitations of sorptivity and water permeability for the estimation of the chloride penetration rate in concrete regarding the accomplishment of prescriptive design for durability in the marine environment

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ABSTRACT

This paper presents an analysis of experimental data from conventional concrete regarding sorptivity and penetrability under pressure comparing these parameters to chloride diffusion rate determined in the laboratory and in actual marine environment. Prescriptions for durability assurance of reinforced concrete structures is based on the qualitative characterization of transport properties. For the specific case of the marine environment, it is required to assess the resistance of concrete to chloride ingress. The results show the limitations of both parameters as prescriptive indexes, with capillary absorption rate showing some advantages over water penetration under pressure.

Keywords: capillary absorption; water penetration; chloride; durability; prescriptive design.

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Limitaciones de la velocidad de absorción capilar y la permeabilidad al agua para la estimación de la velocidad de penetración de cloruro en hormigón respecto a la consecución del diseño prescriptivo para durabilidad en ambiente marino

RESUMEN

Este artículo presenta un análisis de datos experimentales de hormigón convencional respecto a la velocidad de absorción capilar y la penetración de agua a presión comparando estos parámetros con la velocidad de difusión de cloruro determinada en el laboratorio y en ambiente marino real. Las prescripciones para el aseguramiento de la durabilidad de estructuras de hormigón armado están basadas en la caracterización cualitativa de las propiedades de transporte. Para el caso específico del ambiente marino, se requiere evaluar la resistencia del hormigón al ingreso de cloruro. Los resultados muestran las limitaciones de ambos parámetros como índices prescriptivos, con la velocidad de absorción capilar mostrando algunas ventajas sobre la penetración del agua bajo presión.

Palabras clave: absorción capilar; penetración de agua; cloruro; durabilidad; diseño prescriptivo.

Limitações da taxa de absorção capilar e da permeabilidade água para a estimativa da taxa de penetração de cloreto em concreto em relação à realização do design prescritivo para durabilidade em ambiente marinho

RESUMO

Este artigo apresenta uma análise de dados experimentais de concreto convencional em relação à velocidade de absorção capilar e à penetração de água sob pressão para comparar estes parâmetros com a velocidade de difusão de cloretos determinada no laboratório e no ambiente marinho real. Os requisitos para a garantia da durabilidade das estruturas de concreto armado baseiam-se na caracterização qualitativa das propriedades de transporte. Para o caso específico do meio marinho, é necessário avaliar a resistência do concreto à ingestão de cloreto. Os resultados mostram as limitações de ambos parâmetros como índices prescritivos, com a velocidade de absorção capilar apresentando algumas vantagens sobre a penetração de água sob pressão.

Palavras chave: absorção capilar; penetração de água; cloreto; durabilidade; desenho prescritivo

1. INTRODUCTION

Most of the concrete deterioration mechanisms are related to the performance of cover concrete. This concrete layer is responsible for the durability of the structure to the degree that it provides physical and chemical protection to reinforcement against external aggressive substances. Design methods for durability look into the characteristics of this cover concrete for assessing a certain lifespan.

The service life of a structure regarding the degradation of constituents can be explained by applying the model proposed by Tuutti (Tuutti, 1982). The stipulated service life is the period during which the service requirements must be met, with a level over the acceptable minimum regarding safety, comfort and aesthetics. For this, the exposure conditions to which the structure will be exposed must be considered.

In general, the regulation considers periods of stipulated service life of 50 or 100 years, provided that cracking is controlled, and concrete is properly placed, compacted and cured. Minimum requisites for the properties and depth of cover concrete are defined. This prescriptive approach is widely accepted, but it is limited regarding the accuracy of the projected service life (Rostam, 2000). The general classification of environments and target parameters impedes to consider all the intervening factors. These parameters are often qualitatively restricted, or an indeterminate quantification is established for them (Anoop et al., 2002), and the actual service lifespan cannot be accurately estimated. Reinforcement corrosion is one of the most investigated deterioration processes design for durability of reinforced concrete structures.

Tuutti's model divides the corrosion process into two development periods covering the service lifespan (Tuutti, 1982): initiation and propagation. During the initiation period, the incubation of the conditions necessary for the beginning of the degradation develops. During the propagation period, the situation progressively worsens with lower and lower performance level to the moment in which the deterioration degree of the structure is such that it does not comply with the minimum service conditions required. In general, the time required for reinforcement depassivation is conceived as the initiation period, while cracking and spalling are conceived within the propagation period (Tuutti, 1982; DURAR, 1997; Rostam, 2000).

The influencing factors for the initiation period in the marine environment are classified in internal and external. Internal factors are mainly related to the characteristics of cover concrete. Among these factors the most important are those that determine the material resistance to chloride ingress: porosity of the matrix (Collepardi et al. 1970; Monosi et al. 1989) (determined by the w/b ratio, compaction degree, curing), content and type of cement (Collepardi et al., 1970; Glass and Buenfeld, 2000), porosity of the interfacial transition zone (Delagrave et al., 1997a), and porosity of aggregate (Fernández Luco, 2001). The aggressiveness of the environment defines external factors (Sandberg et al., 1998; Traversa, 2001; Andrade et al., 2002; Traversa and Di Maio, 2002; Di Maio et al. 2004), characterized by the average temperature and relative humidity, incidence of winds, rain, distance and height with respect to sea level. Finally, the presence of protective surface layers on the structure (paint or finishing) must be considered as these reduce the exposure level (Di Maio et al., 2000). All these factors determine the time required to chlorides to reach reinforcement.

Chloride ingress into reinforced concrete leads to reinforcement pitting. When the chloride threshold content on reinforcement surface is achieved, steel depassivates and starts corroding if oxygen and moisture are available. Therefore, cover concrete must prevent this situation as long as possible. Its transport properties define the time required for corrosion to start. Lower transport rate of chlorides through cover concrete will allow a longer service life of the structure.

The resistance of cover concrete to chloride ingress is usually defined by the apparent diffusion coefficient (Collepardi et al., 1970), D_{ap}, which establishes the greater or lesser rate at which ions enter into concrete. The performance-based design applies this coefficient to compute a certain service life of the structure for a certain cover depth. However, D_{ap} is generally not explicitly included in construction regulation as a design parameter for concrete, as its measurement is very time-consuming. Some uncertainties remain regarding the application of diffusion coefficients for service life prediction, as real conditions of exposure are very difficult to simulate in short-term tests, particularly to consider the dependence of the apparent diffusivity on the chloride surface concentration (Andrade et al., 2000). In consequence, design engineers show reluctance to use complex models for the prediction of chloride ingress into concrete, and they are little prone to introduce them into Codes or Standards (Andrade et al., 2013). Instead, the correlation between chloride diffusivity with other properties of concrete is frequently considered for the design. This approach is the basis for the prescriptive design.

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Prescriptive criteria are basically maximum or minimum values for different concrete properties to satisfy. These properties may include concrete strength, water/binder ratio, water absorption, capillary absorption rate, water penetration under pressure, cement content and type. All these factors are reported as the main controlling parameters for durability, and on this basis, limits are established from reference values that have proved to provide long-term durability of reinforced concrete exposed to chlorides in the laboratory or in service.

Because of practical reasons, prescriptive criteria is widely included in regulations for durable reinforced concrete structures. Guidelines regarding the durability of reinforced concrete in the marine environment are included in regulations (BS EN 206, 2013; ACI 201.2R, 2016; CIRSOC 201, 2005), where prescriptive criteria are generally established to ensure a service life of 50 years. First, the type of chloride exposure must be defined and rated according to the aggressiveness level. Then, corresponding characteristics for cover concrete are required. These types of limits are easy and reliable when examined. However, prescriptive design for durability does not allow inferring the actual service lifespan. Modern regulations are progressively including performance-based design criteria, but great effort is required in this sense due to extensive experimental work required for the validation of models.

This paper reports comparisons between the chloride diffusion coefficient in saturated and unsaturated conditions and technological properties such as w/b ratio, compressive strength, sorptivity and water penetration under pressure. The main connecting and disconnecting aspects regarding these comparisons are analyzed on the basis of the experimental results.

2. PRESCRIPTIVE DESIGN FOR DURABILITY IN THE MARINE ENVIRONMENT

2.1. Water/binder ratio
The capillary porosity of the cementitious matrix is a function of the w/b, given certain hydration and compaction degrees. During the initiation period, the availability of connected porosity that allows moisture and ion transport is essential for the development of the deterioration process. The limitation to a maximum w/b ratio leads to a decrease in the volume of capillary porosity in concrete. This reduced w/b ratio must be complemented with proper curing treatment that allows sufficient progress of cement hydration, as well as efficient consolidation that eliminates macropores.

The influence of w/b on chloride diffusivity of concrete has been widely investigated (ACI 222, 2003). However, its utility as a prescription parameter cannot be only sustained by the confirmation of incidence. JSCE proposes a potential relationship between w/b and diffusivity through concrete (Song et al., 2008), but, as said, other interrelated affecting parameters cause significant deviation from this relationship. Therefore, concrete properties significantly related to the w/b ratio are generally prescribed. This is also due to the difficulties in controlling the w/b ratio in the field, as no reliable experimental method is available.

Regarding active corrosion of reinforcement, the flux of oxygen through concrete is also a function of the reduction of w/b (ACI 222, 2003). Both chloride and oxygen diffusion are connected with the relationship between permeability and w/b.

2.2. Cement content
Cement content is determining of the durability in the marine environment in several aspects. First, for a certain w/b, more cement means larger volume of paste in concrete. The use of water reducing admixtures may contribute to reducing the cement content in concrete. The reduction of paste content in concrete is potentially a beneficial effect, as aggregates are usually less porous than the

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matrix and increasing their contents helps in reducing transport properties of concrete provided that proper compaction is applied.

Conversely, C_3A contained in cement is the largest contributor to the chloride binding capacity in concrete, and this feature is dependent on the type and content of cement (Andrade, 1993; Delagrave et al., 1997b). Chloride binding is a delaying process of chloride ingress, and service life increases when chloride binding is enhaced. Then, increasing cement content in concrete means increasing chloride binding capacity.

Supplementary cementitious materials contained in cement affect durability in the marine environment by two opposite effects, dilution and pozzolanic action. These effects are not so marked in the results of accelerated test methods, but they are verified more extensively with time. For this reason, some procedures make attempts to consider the late reactivity of supplementary cementitious materials. In any case, significant changes with time are caused depending on the cement type.

2.3. Compressive strength

As said, the w/b ratio is the most important parameter regarding transport properties of concrete. This design tool is difficult to be controlled in the field. Then, its relationship with compressive strength, which has been extensively proved and explained on the basis of the conformation of the pore structure, is used for design. This direct relationship is the most developed in the field of concrete technology. Therefore, a strong basis for the use of the compressive strength as an evaluation parameter for virtually any other property of concrete, including chloride diffusivity, is available. As expected, diffusivity in saturated state decreases consistently with increasing compressive strength, and simple empirical relationships between compressive strength at 28 days and chloride diffusivity of concrete have been determined (Frederiksen et al., 1997). These relationships are probably affected by entrained air. Differences caused by the cement type and practices for accelerating strength gain (curing treatment, additives) are also to be expected.

However, the determining factor for the use of compressive strength as a control parameter is its practicality, cost and universality. Then, it is very easily implemented and interpreted. The application of this control parameter has shown a very variable degree of success, and this is the reason for the application of more comprehensive control parameters for durability in the marine environment.

2.4. Sorptivity

The water capillary absorption rate is one of the transport mechanisms through which chloride can penetrate into concrete in the marine environment. This property is an effective descriptor of the pore structure of concrete. Capillary absorption develops in unsaturated concrete, and it can transport chloride into concrete as the wet front progresses. However, pure diffusion takes place only when no liquid flux is produced, and in this sense, it is different from capillary absorption. The pore size ranges participating in both processes and the processes themselves are different. In spite of this, fair correlations between sorptivity and chloride diffusion are presented in the literature (Basheer, 2001; Kropp and Basheer, 2000). These relationships depend on the applied test methods for the determination of each property, which are very sensitive to preconditioning. In this sense, a high sensitivity of the value obtained for sorptivity to test conditions and proportions of constituents of concrete has been documented (Taus, 2010; Bjevović et al., 2015). Moreover, particular consideration must be made for its contrast with chloride diffusion in unsaturated conditions, as in this case only part of the pore structure intervenes in the transport process. The direct correlation between chloride diffusivity and sorptivity may be therefore affected by the saturation degree of concrete.

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2.4. Permeability

In the case of structures subjected to a hydrostatic pressure difference, permeability is the parameter that best represents penetration of aggressive substances from the external environment, e.g. transport of chloride in seawater structures. Furthermore, high resistance to chloride penetration for low permeability concrete has been obtained in connection with the limited connectivity of the pore structure (CCAA, 2009). Again, the processes of water penetration under pressure and chloride diffusion are different, especially considering their correspondence with the saturation degree of concrete.

3.   MATERIALS AND METHODOLOGY

The studied concrete mixes were 28 in total with multiple batches for most of them, making a total of 73 batches. Proportions of studied concretes correspond to w/b between 0.39 and 0.61, with cement contents between 425 and 250 kg/m^3. Three types of portland cement used were: Ordinary (OPC), Limestone (LPC, incorporating 17% limestone), and Composite (CPC, 17% and 12% incorporation of limestone and slag, respectively). Crushed granite aggregates with maximum sizes of 19 and 25 mm were used as coarse aggregate. Fine aggregate was siliceous river sand. Materials with negligible chloride contents were used. Chloride content of concrete coming from constituents (IRAM 1857, 2000) was in all cases lower than 0.03%. For more details on the proportions of these concretes please refer to (Violini et al., 2006; Taus et al., 2008; Villagrán Zaccardi, 2012).

Tests were conducted to determine compliance of concrete mixes to prescriptive parameters. These include compressive strength, measured on cylindrical specimens of 15x30cm in diameter and height, compacted and tested according to IRAM 1524, 1546 and 1553, capillary absorption rate, determined according to IRAM 1871 (IRAM 1871, 2004), and water penetration under pressure according to IRAM 1554 (IRAM 1554, 1983). All specimens were compacted manually, demolded after 24 hours of casting, and cured in a humid chamber (Temp: 23 ± 2 °C; RH> 95%) until the age of 28 days.

Chloride transport rate was evaluated in prismatic specimens 7.5x15x25 cm^3. After the curing treatment, these specimens were waterproofed with chlorinated rubber paint on all sides excepting the molding surface, from which unidirectional chloride ingress was allowed. Specimens were exposed in a natural marine environment and immersed in sodium chloride solution, with the ingressing face set as the horizontal upper side. Thus, they were exposed with the same position they were molded.

The natural marine environment exposure was in the city of Mar del Plata, Argentina, approximately 50 meters from the shoreline and 5 m above mean sea level. Direct contact between samples and seawater did not occur at any time during exposure, and the only source of chlorides was the sea spray.

Specimens exposed in immersion were first saturated for 24h in saturated lime water, and then submerged in 30 g/l NaCl solution maintained at 23 ± 5 °C until profiling.

Chloride ingress profiles were analyzed after 12 months of exposure for specimens in the marine environment and after 6 months for specimens in immersion. First, painted sides were discarded in approximate thickness of 1 cm. Then, cuts parallel to the ingress face were made, obtaining progressive slices of about 5 mm. Average depths from the ingress surface were measured for each slice. All cuts were made in dry condition with a diamond disc. Afterwards, the slices were pulverized, and acid-soluble chloride contents were determined in accordance with IRAM 1857, method C (IRAM 1857, 2000).

Data regressions to the most common solution of the Fick's second law, Equation (1) were performed, and values for D_{ap} were determined. In the cases in which non-Fickian behaviour was detected, the procedure indicated in (Andrade et al., 2015) was followed.

$$C_{(x,t)} = C_s \left(1 - erf\left(\frac{x}{2\sqrt{D_{ap} \cdot t}}\right)\right) \tag{1}$$

where $C_{(x,t)}$ is the chloride content at depth x, in time t, erf is the error function, C_s is the apparent surface chloride content at time t, and D_{ap} is the apparent non-stationary diffusivity.

4. RESULTS AND DISCUSSION

Chloride penetration is driven by the concentration gradient between the surface and the interior of concrete. In consequence, chloride contents that penetrated into concrete in immersion were up to three times higher than those determined in concrete in atmospheric marine exposure. Moreover, unlike during penetration under atmospheric exposure, in saturated concrete, the entire porosity is occupied by pore solution, which constitutes the medium through which chloride enters. Then, faster penetration in saturated than in unsaturated concrete is obvious. The content of pore solution will mainly define the correlation between penetration rates in submerged specimens and exposed to marine environment. This content does not remain constant in specimens under atmospheric exposure, as it is influenced by weather conditions, especially in the more external zone. Therefore, the comparison should take note that both ingress mechanisms are not fully equivalent, and the contrast is merely empirical. Moreover, in a marine environment with different climatic conditions from the one considered in this study, different hygroscopic equilibrium with the environment of concrete will result in a different content of pore solution, and different cycles of wetting and drying.

From the penetration profiles, the direct comparison between the computed values for diffusivity in specimens exposed in the natural marine environment (D_{atm}) and immersed (D_{inm}), is shown in Figure 1. A remarkable trend with little impact of the cement type is noticed. For low penetration rates, greater increments for D_{atm} than for D_{inm} are revealed. This ratio seems to reverse over a certain value, and for high penetration rates, smaller increments for D_{atm} than for D_{inm} are noticed. This may be explained by the concept previously mentioned. Pore size distribution and total porosity are connected. Low diffusivities correspond to concrete mixes with low porosity. In this case, pore liquid remains in a relatively greater volume of small pores of concrete under atmospheric exposure. As porosity increases, larger pores contribute to the penetration of chloride when concrete is in immersion, but they do not remain saturated in concrete under atmospheric conditions.

This is, changes in diffusivity relate more directly to the change in the total pore volume when concrete is saturated, as all porosity contributes to the transport mechanism. On the contrary, only finer pores contribute to transport in unsaturated concrete, and D_{atm} does not continue increasing at the same rate with increasing volume of macropores (Saetta et al., 1993; Climent et al., 2002; Nielsen and Geiker, 2003; Zhang and Zhang, 2014).

Figure 2 shows D_{atm} and D_{inm} compared with w/b. Despite the potentially different chloride binding capacity, no statistically significant differences can be established among the different cement types. The influence of w/b very much hides any difference.

As expected, increasing diffusivity values are obtained with increasing w/b. However, significant variations within each w/b were obtained. The influence of the other variables (hydration degree, maximum aggregate size and cement content) is the primary reason for this. It is particularly important to note that the sum of these influences may result in a similar degree of impact to the variation of the ratio w/b, especially for diffusivities determined in the unsaturated condition.
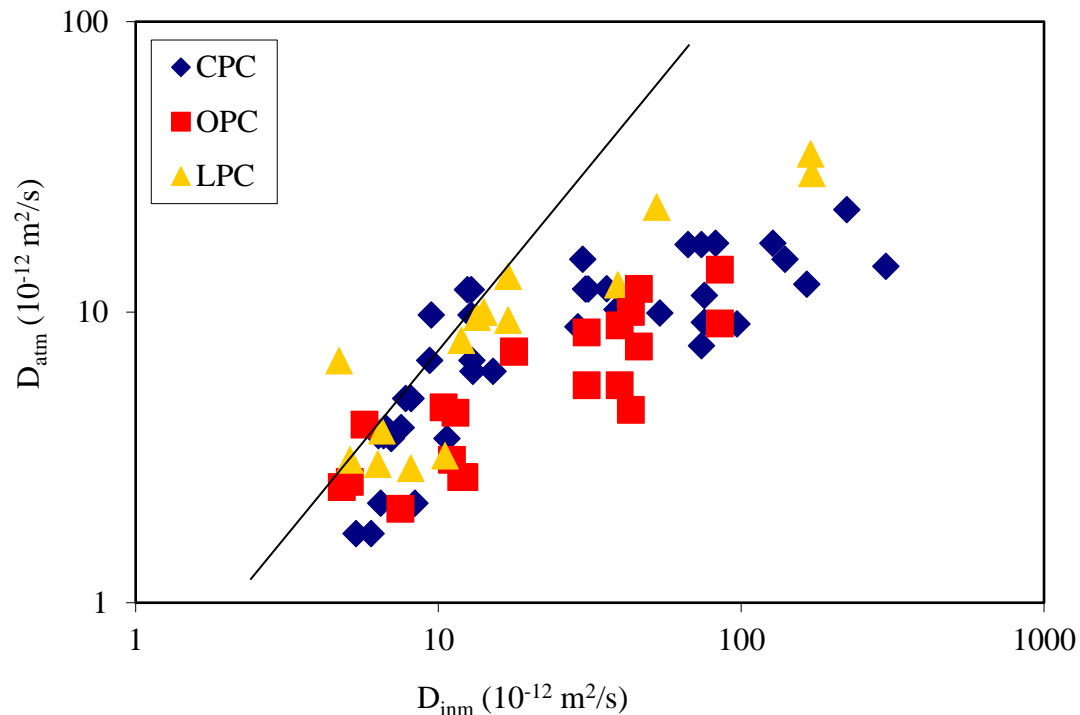

Figure 1. D_{inm} versus D_{atm}.

The differentiation between diffusivity values when in immersion or atmospheric is more pronounced with higher w/b (D_{inm} increases much more than D_{atm} with increasing w/b). Concrete porosity increases with w/b, but this increased porosity results in more content of pore liquid only in the saturated state. Macropores do not affect in the same way to samples exposed to the atmosphere. At unsaturated conditions, the volume of pore liquid is defined by the fine capillary pores, where more or less condensation will occur depending on the relative humidity. In this range of pore sizes, w/b has less significance regarding the volume fraction of pores participating in the transport process.

In this sense, engineering transport parameters, such as sorptivity and penetrability under pressure, are related to pore structure volume and connectivity. They are therefore indirectly connected to chloride diffusivity in saturated concrete (as shown later in Figures 3, 5 and 6).
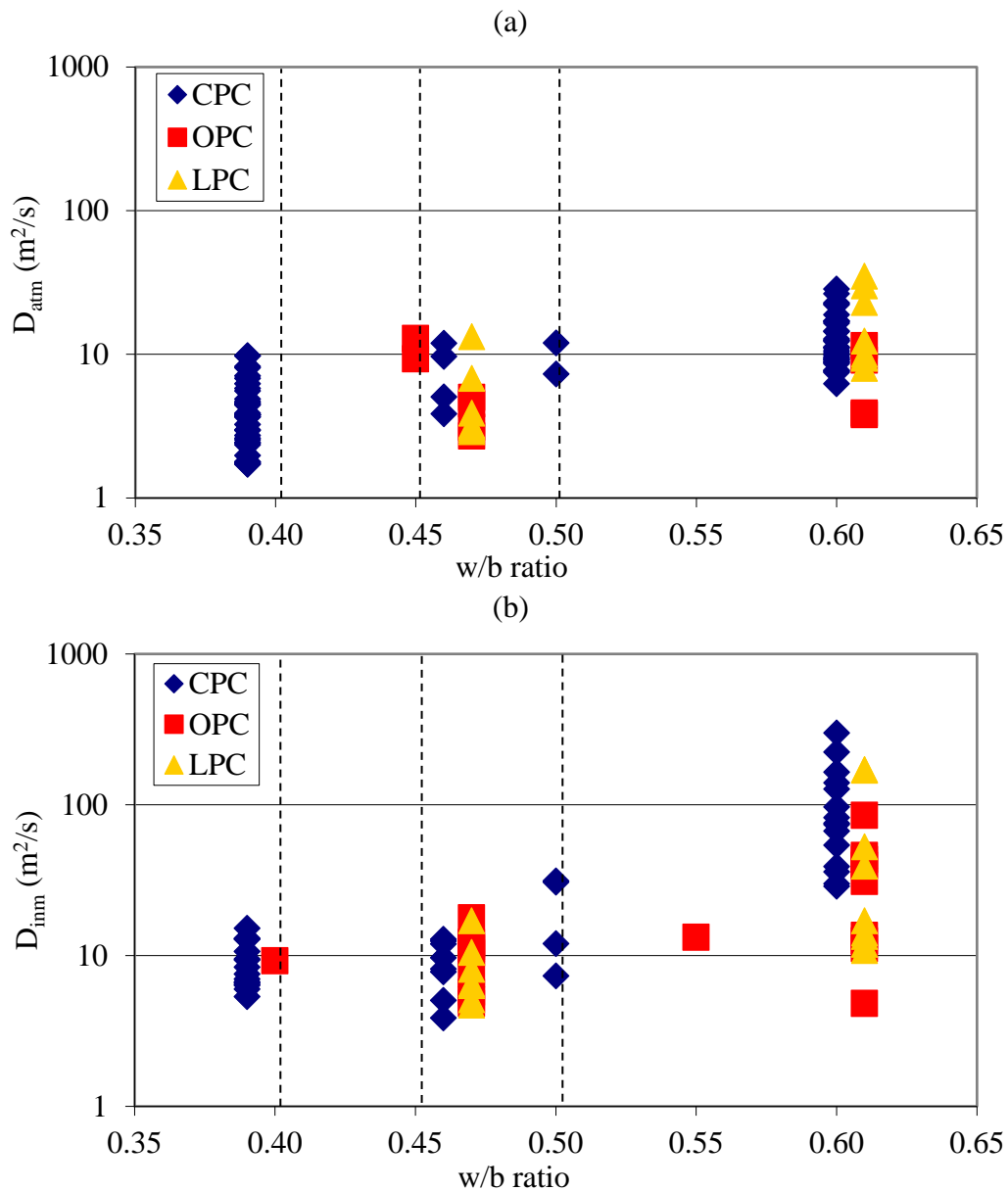
(a)

(b)

Figure 2. (a) D_{atm} and (b) D_{inm}, versus w/b.

Figure 3 shows the comparisons between sorptivity and diffusivity. Again, the variation in sorptivity is higher than the variation in D_{atm}. The variation in capillary absorption rate is more directly connected with D_{inm}. The variability of sorptivity is greater than the corresponding to the w/b. This indicates how unsuitable is the w/b for defining the transport rate in concrete. As in the case of diffusivity, other factors different from the w/b are also affecting the transport rate, and these are taken into account only when the transport property itself is measured. Fig. 2 shows a significant number of conformity values by w/b (values of 0.40, 0.45 and 0.50 are generally accepted depending on the environment and the consideration of reinforced or prestressed concrete). This number is certainly reduced when limits are based on a tolerable limit for sorptivity. It should be mentioned that the correspondence between w/b ratio and capillary absorption rate will certainly be different in concrete made with water-reducing admixtures, which allow lower mixing water (and in consequence also paste volume) for the same consistency level. In that case, it is estimated that the capillary absorption rate may decrease for the same w/b.
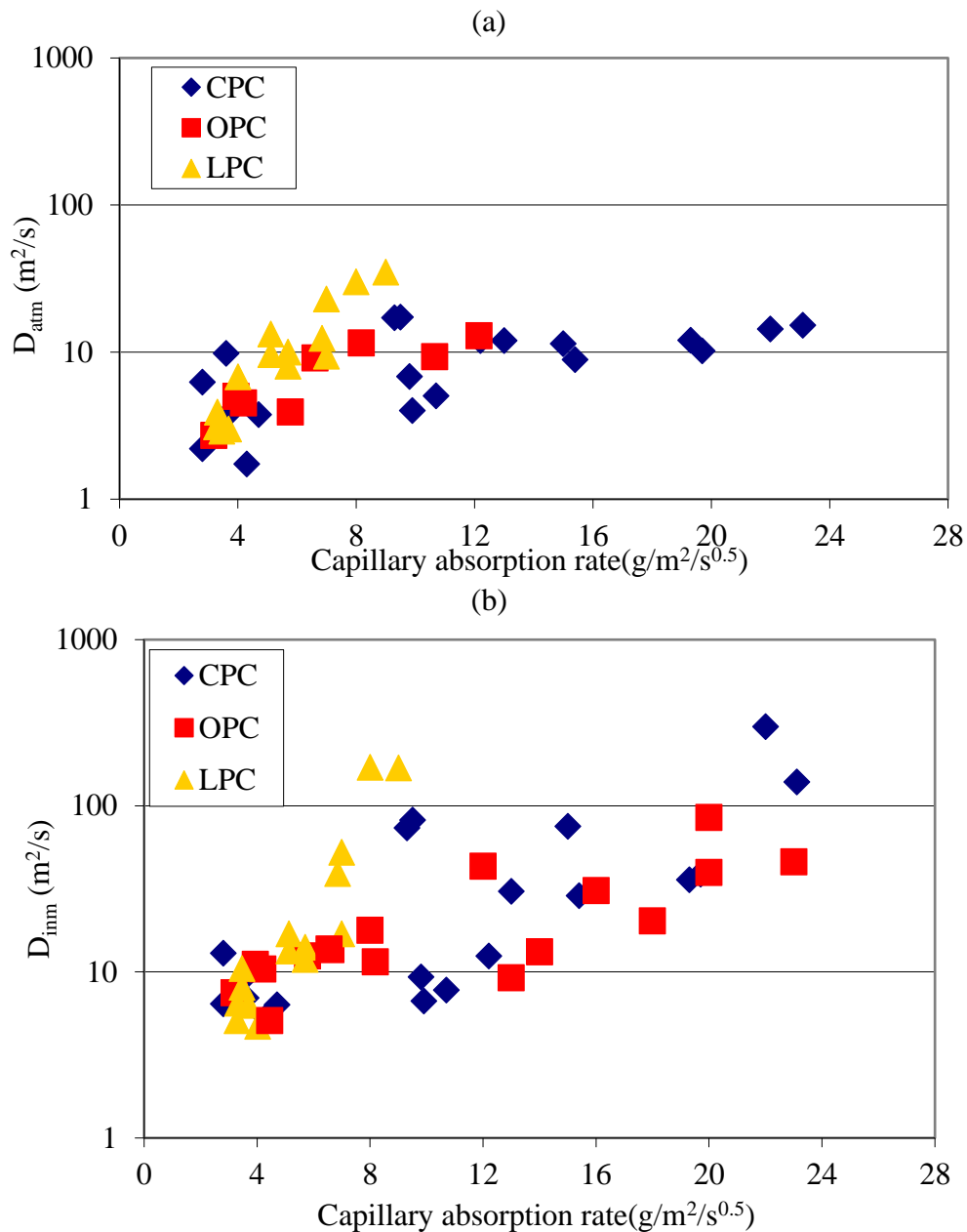
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(a)

(b)

Figure 3. (a) D_{atm} and (b) D_{inm} versus sorptivity.

In Figure 4, the comparisons between diffusivities and compressive strength are presented. An inverse evolution is obtained as a consequence of the opposite relationships of both properties with porosity. A greater dispersion is noted for D_{inm} than for D_{atm}. The number of conformity values according to strength is similar to that according to w/b. Thus, the direct relation between strength and w/b allows a reliable control procedure by the first, in order to assure compliance of the second. This is an aspect of practical necessity due to the lack of reliable methods for experimentally controlling the w/b in fresh concrete. On the other hand, it should be mentioned that strength of cover concrete in the structure will be greatly affected by the consolidation degree, which is in turn highly dependent on the field practices.
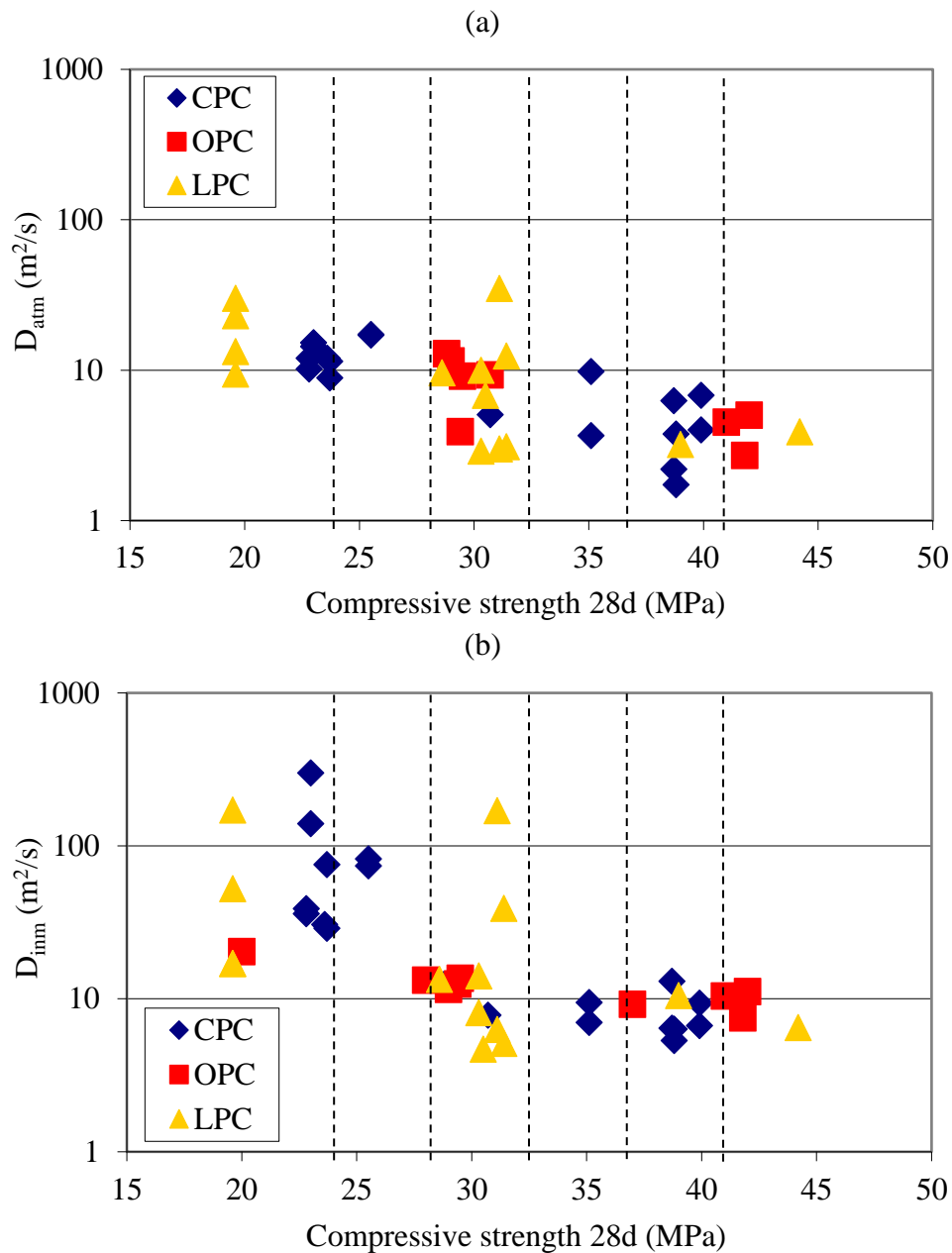
(a)

(b)

Figure 4. (a) D_{atm} and (b) D_{inm} versus compressive strength.

The experimental determination of the compaction index of fresh concrete is then advisable to define the sensitivity of concrete to compaction and establishing an indicative relative risk for the required level of control during placement of fresh concrete. In practice, the lack of this consideration commonly leads to bad results regarding the durability of the structure.

In Figures 5 and 6, the comparisons between diffusivities and water penetration under pressure, mean and maximum, respectively, are presented. The proportion of conformity values shows water penetration under pressure as a less challenging property than sorptivity.

The compliance of both mean and maximum water penetration depths is equivalent. Compaction defects in the specimen are required for obtaining a significant difference between these two parameters. The good correspondence of values shows that this was not the case for any of the tested specimens. With this in mind, it should be remarked the very limited use of the maximum penetration of water under pressure regarding chloride penetration and other transport properties of concrete.
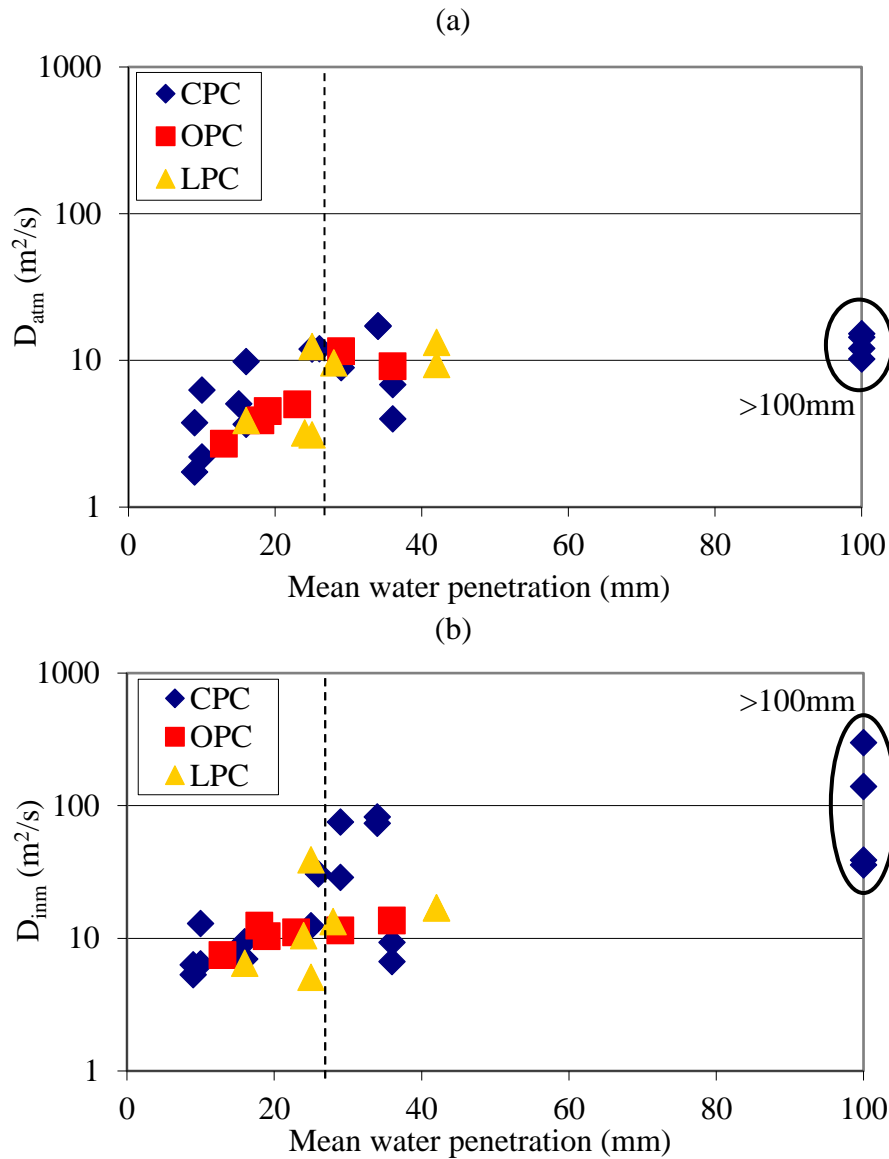
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(a)

(b)

Figure 5. (a) D_{atm} and (b) D_{inm} vs. mean penetration of water under pressure.

Among the analyzed parameters, sorptivity was the one showing the best correlation with chloride diffusivity. Compressive strength, w/b, and water penetration under pressure followed in that order. This is remarkable in the sense that water penetration under pressure may be wrongly recommended over compressive strength for the estimation of chloride penetration in concrete when only considering that transport properties should be better connected to each other.

However, all examined properties showed a better correlation with chloride diffusivity in the saturated condition. For unsaturated concrete, macropores are not participating in the process of chloride ingress, but they do in other processes such as water absorption and penetration. Therefore, unsaturated concrete with high w/b tends to be classified as less durable in the marine environment when tested for sorptivity or water penetration under pressure than for chloride ingress rate itself. The saturation degree of concrete is an important aspect that should always be considered in this regard.
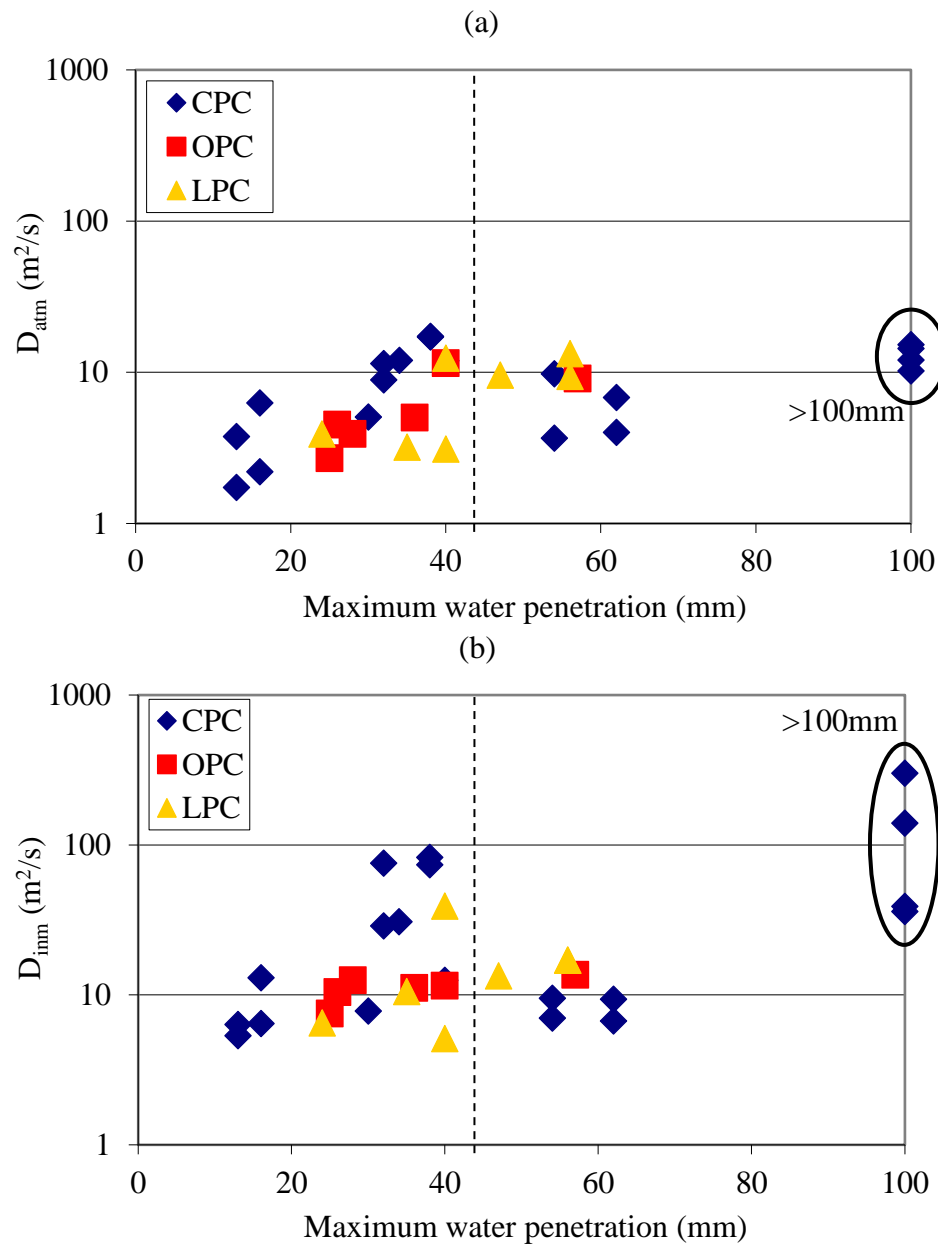
(a)

(b)

Figure 6. (a) D_{atm} and (b) D_{inm} vs maximum penetration of water under pressure.

5.   CONCLUSIONS

The w/b ratio is the technological parameter determining the chloride ingress rate into concrete. However, other parameters also affect this transport property, such as hydration degree, maximum aggregate size, compaction degree, and cement content, and in combination, their effect may exceed the one from the w/b. Therefore, the w/b ratio as a design parameter for durability in the marine environment requires the assistance of complementary prescriptions for transport properties.

The capillary absorption rate demonstrated a consistent correlation with the chloride diffusion rate in the saturated condition. It is convenient to expand the database for this correlation in order to make reliable predictions on this basis with applications in prescriptive design. However, the

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connection between sorptivity and chloride diffusion in unsaturated concrete is less consistent, as the pore size range participating in each transport mechanism is different.

Water penetration under pressure showed little application for the design for durability regarding chloride penetration into concrete. In this sense, compressive strength is considered more practical and reliable for estimating the performance of concrete in the marine environment. Therefore, no added value of water penetration under pressure over compression strength is anticipated. An exception could be made for chloride penetration in saturated concrete, where the chloride penetration rate can be better anticipated by the maximum water penetration under pressure than for the case of unsaturated concrete.

As a result, combined data of capillary absorption rate and compressive strength seem to work well as prescriptive parameters for durability in the marine environment. In many cases, the determination of the capillary absorption rate faces some practical inconveniences for its application in the field, mostly due to its sensitivity to testing variables and the required testing time. The values for prescriptive limits for sorptivity are still a matter of study.

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