Hydrophobe VII *7th International Conference on*
Water Repellent Treatment and Protective Surface Technology for Building Materials
A.Elena Charola and J. Delgado Rodrigues, Editors
LNEC (Laboratório Nacional de Engenharia Civil), Lisbon: 2014 (215-224)

Performance assessment of hydrophobic treatments on different substrates

Carlos Esteves
Instituto Superior Técnico, Lisboa, Portugal, carlos.esteves@ist.utl.pt
Inês Flores-Colen
Instituto Superior Técnico, Lisboa, Portugal, ines.flores.colen@ist.utl.pt
Rosário Veiga
Laboratório Nacional de Engenharia Civil, Lisboa, Portugal, rveiga@lnec.pt

SUMMARY: The present paper presents an experimental study with the aim of studying the hydrophobization of three supports (stone, rendering mortar and an external thermal insulation composite system - ETICS) through the application of three hydrophobic products: one a silica (SiO_2) and titania (TiO_2) based nanostructured dispersion (H_{NST}); a silane/oligomeric siloxane ($H_{Sila/Silox}$); and a siloxane (H_{Silox}). The samples of the untreated and treated supports were characterized by laboratory tests (water absorption by capillarity, drying, water permeability under low pressure with Karsten pipes, water vapor permeability and contact angle using the water droplet methodology) to determine the initial effectiveness of the hydrophobic treatments. The results shown that all products introduced significant changes in the hydric properties of the stone, mortar and ETICS supports, namely in terms of initial water absorption by capillarity, initial drying rate, drying index, resistance to water vapor diffusion and water-repellency assessed through the contact angle.

KEY-WORDS: Rendering mortars; stone; ETICS; hydrophobic products; superficial protection; effectiveness

INTRODUCTION

The protection of material surfaces exposed to environmental actions may be a good solution as it increases the buildings lifetime by preventing or decreasing deterioration. Several surface protection products are available nowadays, but impregnation with hydrophobization agents are the most frequently used. The reduced impact on the aesthetics, the surface water-repellency, and ease of application are some of the key characteristics required for these products [1]. The factors that influence their performance and durability have not as yet been fully investigated, particularly for the most recently developed products. While many studies have addressed the application of hydrophobic treatments to stone, concrete and ceramics, there are fewer ones on the application to renders.

Water has an important role on the properties of porous building materials, reducing the thermal and acoustic performance, facilitating salt efflorescence and accelerating the growth of microorganisms, causing hydrothermal aging and hygiene problems inside buildings [2]. The application of hydrophobic products intends to reduce the rate of water absorption, while not affecting water vapor permeability, diminishing soiling and the growth of biological colonization, among other effects [3, 4, 5].

This paper aims to discuss the performance of three hydrophobic products on limestone, cement-based render and external thermal insulation composite system (ETICS). The three products are: i) silica (SiO_2) and titania (TiO_2) based nanostructured dispersion *(H_{NST})*; ii) a silane/oligomeric siloxane based mixture ($H_{Sila/Silox}$); and, iii) a siloxane based product (H_{Silox}). The samples of the untreated and treated supports were characterized by laboratory tests (water absorption by capillarity, drying, water permeability under low pressure with a Karsten pipe, water vapor permeability and contact angle of water droplet) to determine the effectiveness of the hydrophobic treatments.

MATERIALS

Supports

Three supports were used: i) limestone ("moleanos"), ii) cementitious rendering mortar with 2cm of thickness applied on top of ceramic brick; and, iii) ETICS. Table 1 shows some characteristics of these supports.

Table 1. Average results of water capillary, open porosity and bulk density of the supports.

Support	Capillary coefficient ($kg/m^2.min^{0.5}$)	Porosity (%)	M_{AP} (kg/m^3)
Limestone	0.234	10.96	2385.17
Rendering mortar	0.182	30.23	1441.24
Ceramic brick	0.074	25.49	1979.06
ETICS	0.044	-	-

Hydrophobic products

From the wide range of hydrophobic products available on the market, three were chosen to be tested on the selected supports. Table 2 provides the active ingredient of each hydrophobic product and the application information.

Table 2. Hydrophobic products brief characterization and application information.

Designation	$H_{Sila/Silox}$	H_{Silox}	H_{NST}
Chemical Composition	silane/oligomeric siloxane	siloxanes	silica and titania nanostructured dispersion
Total consumption	1 l/m^2	1 l/2.5 m^2	1 l/9 m^2
Number of coats	2	1	2

The application of the hydrophobic products was made by brush following the recommendations of the manufacturer and with the suggested timing. The hydrophobic products were first applied in one direction and later in the orthogonal direction in order to avoid blank spots. A second coat was applied after 10 min for the $H_{Sila/Silox,}$and after 3 h for the $H_{NST.}$ After the product applications, the samples were kept in a controlled temperature chamber at 23 °C and 50% of relative humidity for at least 7 days in order to complete the polymerization of the products. The samples without hydrophobic products were exposed to the same conditions, so all the samples were subjected to the same conditions before testing. The consumption of each product was noted.

EXPERIMENTAL METHODS

The performance of the hydrophobic products was evaluated by the following tests: water absorption by capillarity, water absorption under low pressure using of *Karsten* pipe, drying, water vapor permeability, biological colonization and contact angle between water droplet and the support. Three samples of each hydrophobic product were used for each test and for each support, i.e., a total of nine samples.

Water absorption by capillarity was determined based on the European standard EN 1015-18 [6] for the stone and render specimens. For the ETICS samples, ETAG 004 was used [7]. The water absorption at low pressure with the *Karsten* pipe followed the test No. II.4 of RILEM [8] for 60 min, and it is evaluated by the coefficient of water absorption at this time. The drying test was performed according to test No. II.5 of RILEM [9], which assesses the drying capacity of the untreated and treated samples, through the initial drying speed and the drying index calculated from the resulting drying curve. The water vapor permeability, expressed by the water vapor diffusion resistance coefficient, was carried out according to the European standard NP EN 1015-19 [10]. The test of biological colonization was carried out according to ASTM D5590 [11] and was carried out on both the untreated and the treated renders, in order to evaluate the effect of the hydrophobic products on biologic growth. The contact angle was measured under a microscope with the fall of a 4 ml micro water droplet in accordance with European standard EN 15802 [12]. Table 3 summarizes the tests performed, their standards and the total number of measurements.

Table 3. Summary table of the tests performed, where: - = not tested; n.a. = not applicable, and x/y = untreated/treated samples.

Tests	Code/specification	Stone	Render	ETICS	Number of tests
Water absorption by capillarity	EN 1015-18 (2002)	3/9	3/9	n.a.	24
	ETAG 004 - ER3 (2013)	n.a.	n.a.	3/9	12
Drying	RILEM II.5 (1980)	3/9	3/9	n.a.	24
Liquid water permeability	RILEM II.4 (1980)	3/9	3/9	3/9	36
Water vapor permeability	EN 1015-19 (2008)	3/9	3/9	n.a.	24
	ETAG 004 - ER3 (2013)	n.a.	n.a.	3/9	12
Biological colonization	ASTM D5590-00 (2010)	-	3/9	-	12
Contact angle	EN 15802 (2009)	1/3	1/3	-	8

RESULTS AND DISCUSSION

Tables 4 and 5 show the average results of the water absorption coefficient by capillarity and the Karsten pipe, respectively, before and after treatments on the different supports. The results clearly show that the behaviour of all supports improved after application of any of the products by reducing their water absorption. Better results were obtained for the render (95 to 97%) because of its higher open porosity since the hydrophobic products penetrate into the pores of the support coating their walls. This leads to a reduction of the surface tension (lower tension than water) making the surface hydrophobic. In general, the product based on the nanostructured dispersion (H_{NST}) showed better results for stone and ETICS supports, while the hydrophobic product based on siloxane (H_{Silox}) showed better results for the render.

Table 4. Average results and standard deviation for the capillary water absorption coefficient before and after treatments.

Support	Capillary water absorption coefficient ($kg/m^2.min^{0,5}$)							
	Control		$H_{Sila/Silox}$		H_{Silox}		H_{NST}	
	C_{10-90}	SD	C_{10-90}	SD	C_{10-90}	SD	C_{10-90}	SD
Stone	0.234	0.031	0.175	0.012	0.116	0.052	0.040	0.000
Render	0.182	0.031	0.005	0.001	0.004	0.001	0.009	0.000
ETICS	0.044	0.008	0.002	0.001	0.002	0.000	0.001	0.001

Pinto [5] obtained capillary water absorption coefficients in the order of 0.53 and 0.04 $kg/m^2.min^{0,5}$ for a granite before and after treatment with a siloxane based hydrophobic product, respectively. This treatment shows a better initial performance on granite with hydrophobic product based on siloxanes (93% of reduction) than for limestone (moleanos) with hydrophobic product based on siloxanes (50% of reduction).

Table 5. Average results and standard deviation of the coefficient of water absorption at 60 min with the Karsten pipe before and after treatments.

Support	Coefficient of water absorption at 60 min ($kg/m^2.min^{0,5}$)							
	Control		$H_{Sila/Silox}$		H_{Silox}		H_{NST}	
	C_{60min}	SD	C_{60min}	SD	C_{60min}	SD	C_{60min}	Sd
Stone	0.672	0.178	0.170	0.072	0.049	0.030	0.053	0.043
Render	0.906	0.000	0.034	0.009	0.023	0.000	0.011	0.000
ETICS	0.109	0.042	0.062	0.028	0.034	0.028	0.023	0.009

From the values obtained by capillary water absorption testing, it was possible to trace the progress of water absorbed over time for both the stone (Fig. 1) and the render (Fig. 2). The latter figure shows that any of the hydrophobic products applied significantly reduced the initial capillary absorption of the render. This confirms that the hydrophobic products influence more the render than the stone, although the latter also shows an improvement of its water repellency.

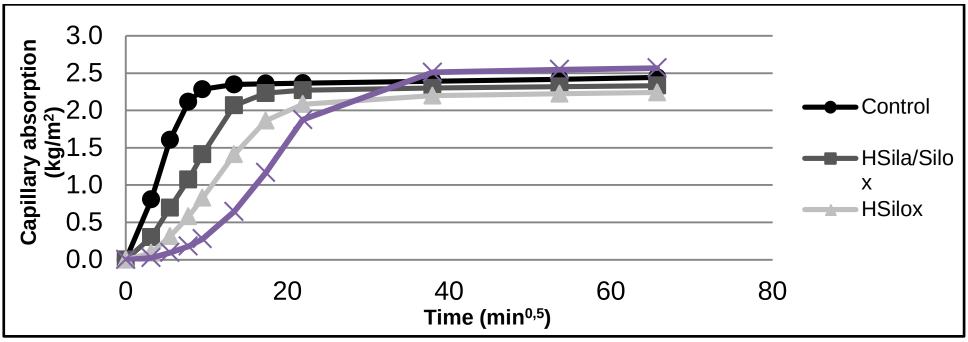


Figure 1. Evolution of the capillary water absorption by the stone.

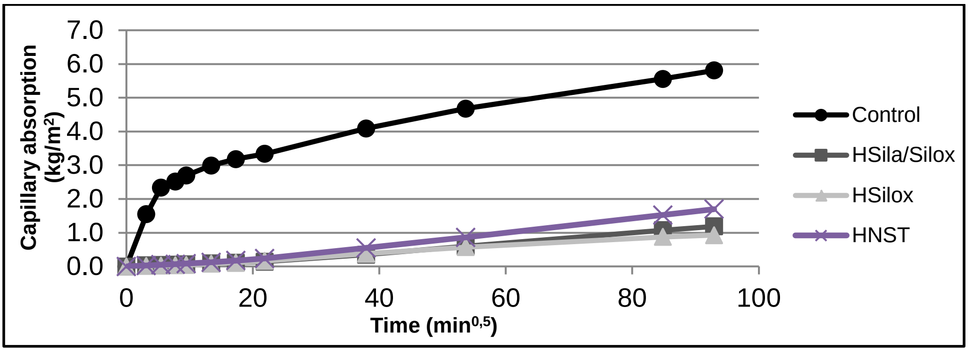


Figure 2. Evolution of the capillary water absorption by the render.

Tables 6 and 7 show the average results of the initial drying rate and the drying index, respectively, before and after treatment of the different supports studied.

Table 6. Average results of the initial drying rate before and after treatments.

Support	Initial drying rate ($kg/m^2.min^{0,5}$)			
	Control	$H_{Sila/Silox}$	H_{Silox}	H_{NST}
Stone	0.0152	0.0148	0.0134	0.0136
Render	0.0161	0.0140	0.0103	0.0152

Table 7. Average results of the drying index before and after treatments.

Support	Drying index			
	Control	$H_{Sila/Silox}$	H_{Silox}	H_{NST}
Stone	0.089	0.095	0.101	0.096
Render	0.191	0.117	0.133	0.207

The results obtained during the drying test served to plot the water loss (evaporated water) per unit area over time for the stone (Fig. 3), and the render (Fig. 4) samples.

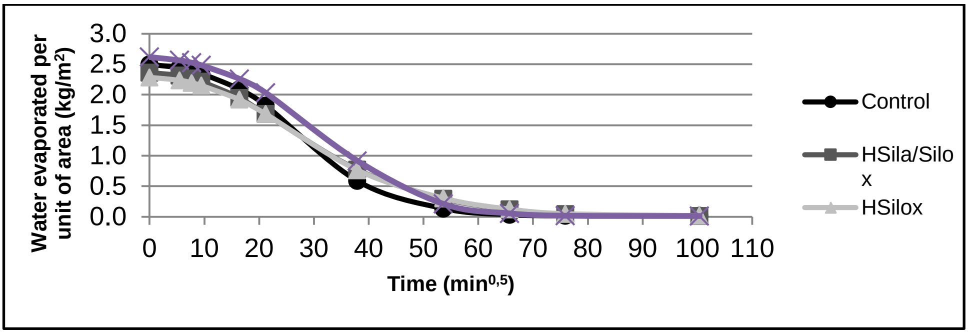


Figure 3. Drying curve for the untreated and treated stone.

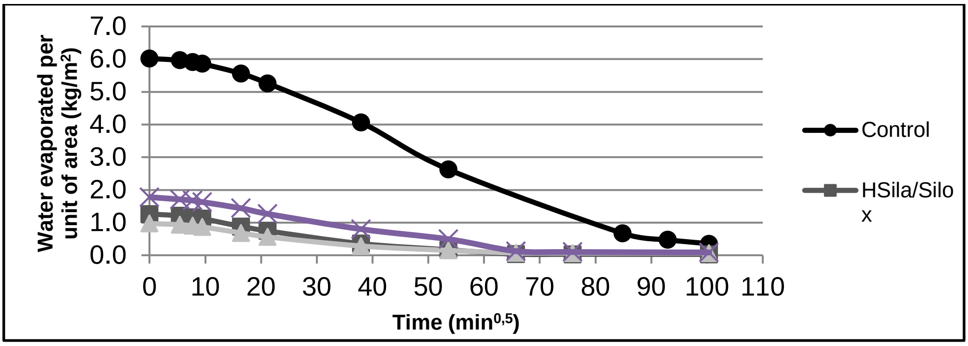


Figure 4. Drying curve for the untreated and treated render.

The initial drying rate for both materials when treated with the products was reduced. For the case of stone, the hydrophobic product based on silanes/siloxanes ($H_{Sila/Silox}$) had the lowest influence (3%) while H_{Silox} and H_{NST} had the worst effect by reducing the drying rate significantly (~11%). Contrariwise, for the render, the H_{NST} had a lowest influence (6%) while H_{Silox} reduced it significantly (36%).

For the case of the drying index, the $H_{Sila/Silox}$ increases slightly the overall drying of stone (6%) but reduces it significantly for the render (39%) mainly because it reduces the amount of absorbed water. Consequently, this product shows improvements for both stone and render supports. H_{Silox} showed the worst behavior by retarding the drying of stone (13%) while for the render, the H_{NST} product had a similar result (8%).

From the drying curves it can be seen that the application of each of the three products had only a slightly retarding effect on the drying behavior of the stone, with H_{NST} clearly differentiated from the other two hydrophobic products. For the case of the render, all three products significantly reduced the amount of water absorbed and therefore lees water needed to be eliminated. As discussed, the H_{NST} showed the worst performance taking longer to dry than the other two hydrophobic products.

All the tested products reduced the initial drying rate for the stone (3 to 12%) and the render (6 to 36%), and by examining the drying index, it was found that all products applied to stone increase its overall drying time (6 to 13%) while for mortar, only H_{NST} showed an increased drying time (8%), while H_{Silox} and $H_{Sila/Silox}$ showed a decreased drying time.

Table 8 shows the average results of the coefficient of water vapor diffusion coefficient obtained by the water vapor permeability test, for the different supports before and after treatment.

Table 8. Average results of the water vapor diffusion resistance coefficient before and after treatment.

Support	μ			
	Control	$H_{Sila/Silox}$	H_{Silox}	H_{NST}
Stone	3.20	10.46	7.30	5.57
Render	2.19	2.15	2.27	2.26
ETICS	51.22	54.34	53.37	61.80

From these results it can be seen that for stone, the $H_{Sila/Silox}$ is the product that increases most the resistance to diffusion of water vapor (227%), while it is least increased by H_{NST} (74%). For the render, none of the tested products significantly affect the water vapor resistance to diffusion (increase between 3 and 4%). In the case of ETICS, this parameter is not particularly affected by the application of the products, with H_{NST} increasing it most (21%), and H_{Silox}, increasing it less (4%).

It is evident that all the applied treatments resulted in an increase of the resistance to water vapor diffusion for all supports, but it was more relevant for the case of stone (74-227%). For the render, this increase was minimal (3 to 4%) while for ETICS it ranged between 4 and 21%. This means that the application of these products reduces the water vapor permeability of the supports, i.e., their "breathability."

Pinto [5] obtained a water vapor diffusion resistant coefficient of 2.88 and 4.49 for granite, before and after treatment with a siloxane based hydrophobic product, respectively. These values show that granite has a better initial performance than the tested moleanos limestone when treated with this type of hydrophobic product. The change in the measured coefficient from untreated to treated, was 56% and 129% for granite and moleanos, respectively.

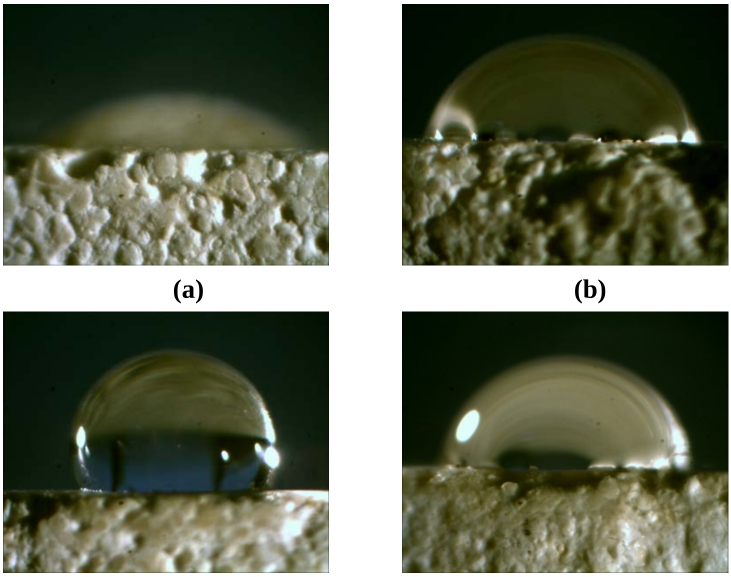
Table 9 shows the average results of the contact angle, before and after treatments on the stone and mortar supports.

Table 9. Average results of the contact angle, before and after treatments.

Support	Static contact angle water-support (º)			
	Control	$H_{Sila/Silox}$	H_{Silox}	H_{NST}
Stone	38	102	133	100
Render	47	90	180	102

From the results obtained, it is clear to see a large increase of the contact angle for all tested products, applied both to stone and to the render. Noteworthy are the high contact angle values obtained with the siloxane based hydrophobic product (H_{Silox}) for both stone and mortar. For stone, the H_{NST} is the one that shows the lowest increase of the contact angle relatively to the untreated sample (163%), while for the render, it is $H_{Sila/Silox}$ (90%). It can be seen that all the applied products increased the water-repellency of the treated supports.

Figure 5 shows the contact angle of the stone, where is notable that the best water-repellency is on the stone with siloxane based hydrophobic product (H_{Silox}).



(a) **(b)**

(c) **(d)**

Figure 5. Photos of water droplet on stone specimens: (a) untreated; and treated with (b) $H_{Sila/Silox}$; (c) H_{Silox}; (d)H_{NST}.

The biological colonization was assessed by the percentage of growth of microorganisms on the render. The results show that there was no great discrepancy in values between the three applied products and also between the untreated support or treated with any of the tested products. However, it was observed that the hydrophobic product that leads to a higher propensity for growth of microorganisms on the mortar was the one based on siloxane (H_{Silox}) even if the difference was not very significant.

CONCLUSIONS

This study aimed to evaluate the performance of the three hydrophobic products (one consisting of a silica and titania based nanostructured dispersion; one based on silane/oligomeric siloxane and another based on siloxane) applied to three different supports, a limestone (11% porosity), a cementitious rendering mortar (30% porosity), and an ETICS cladding. From the evaluation of the applied products on the supports under study, it was clear that all products introduced changes in terms of initial water absorption by capillarity, initial drying rate, drying index, resistance to water vapor diffusion and water-repellency assessed through the contact angle.

In terms of water absorption, the product based on a silica and titania nanostructured dispersion (H_{NST}) showed the greatest improvements to the stone (83%) while $H_{Sila/Silox}$ exhibited minor ones (25%). For the render, the difference between the improvements of the best and worst performing product, H_{Silox} (98%) and H_{NST} (95%), show a minimal difference. A similar minimal difference was observed for the case of ETICS, both H_{Silox} and H_{NST} presented the best values in reducing water absorption (97%) while $H_{Sila/Silox}$ showed the lowest improvements (95%). Through these values, it can also be concluded that treatment of the either the render or ETICS with any of the tested products, reduces the water ingress (95-97% and 95-97%, respectively) and are more effective than when applied the moleanoes limestone (25-83 %). The open porosity of the mortar used is three times higher than that of the stone.

The results from the drying test suggest that all the products applied to stone negatively influence the initial drying speed (3 to 12%) and their drying resistance index (6 to 13%). In the case of mortar, the products improve the drying resistance index (31 to 39%)—except for the H_{NST} (8%)—and decrease the initial drying speed (6 to 36%). In terms of water vapor permeability, the results showed, in general, an increase in the water vapor resistance coefficient, these values being much higher for stone (74-227%). The results suggest that with the application of hydrophobic products, drying of the substrate can be significantly reduced, as in the case of stone. Through the contact angle test it was possible to see a clear improvement of the water repellency of the stone and render for all tested products. The specimens of stone and render treated with the siloxane based hydrophobic product (H_{Silox}) present significantly higher values compared with the remaining hydrophobic products, especially in the case of the render (180º).

The hydrophobic product based on siloxane (H_{Silox}) proved to be the most suitable for use on render, since it had better initial performance concerning water absorption by capillarity, drying, water vapor permeability and contact angle. This may be due to the fact that the molecular structure of siloxanes is more adequate to the higher porosity of this support. In its place the $H_{NST,}$ proved to be less suitable for use in mortar, since it is the hydrophobic product that show less favorable results in the water vapor permeability and drying.

The hydrophobic product based on a silica and titania nanostructured dispersion (H_{NST}) proved to be the most suitable for use on stone and presented the best initial performance of the three tested products. This may be a result of the combination of low porosity and pore size of the stone with the surface deposition of a nanostructured layer. On the other hand, $H_{Sila/Silox}$ appeared to be less suitable for use on stone, as it shows the worst results in terms of water absorption, water-repellency, water vapor permeability and drying.

Among the supports assessed, the render appears to have the best initial performance after the application of any of the tested products, independently of their composition, possibly due to its high porosity (30%) and to the size of its pores. In short, the success of hydrophobic products depends not only on the product itself, but is the result of its "reaction" with the substrate to which it is applied. Therefore, it is critical to match the hydrophobic product to the substrate in question to obtain the best possible result. Ongoing research is being carried out to evaluate the durability of hydrophobic products applied to different substrates by subjecting specimens to artificial accelerated aging.

ACKNOWLEDGMENTS

To the researchers Dr. Dória Costa and Dr. Lina Nunes from LNEC for the possibility to perform the contact angle and biological colonization tests, respectively. And also to Ing. Sofia Malanho and the technicians from the *Laboratório de Revestimentos de Paredes* from LNEC for the support in the experimental work.

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