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How Environmental Circumstances Influence Water Repellent Treatment Efficiency – A Case Study of Bentheim Sandstone

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Abstract. *Water repellents are widely applied directly to the outer surfaces of existing buildings to increase the hydrophobicity and durability of historic facades. Nevertheless, there is little control over the initial moisture content inside masonry and boundary conditions in practice. The fluctuating temperature and moisture content of a wall are inevitable when applying the treatment under natural climatic conditions, therefore the efficiency under realistic application approaches may not be as optimal as in the lab. This research aims to understand how application circumstances affect the hydrophobic efficiency of a siloxane-based water-repellent solvent on sandstone. Samples with different moisture content were treated and cured under different temperature to simulate various field climate conditions in practical applications. The experiments indicate that application temperature and relative humidity have little effect on water-repellent efficiency. The water-repellent product is also effective on saturated walls.*

Keywords: *Hydrophobic Efficiency, Siloxanes-based Water Repellent Agent, Sandstone, Practical Application Conditions.*

1 Introduction

In order to reduce energy demands and CO2 emissions from the building stock, existing buildings are being renovated to make them more energy efficient. Hydrophobic treatments are frequently applied to prevent moisture ingress from external rain loads, allowing to reduce the moisture content, reduce the number of critical freeze-thaw cycles, maintain the dry thermal conductivity, reduce heat flux, and mitigate moisture risks.

A number of studies have been conducted to determine the factors that affect hydrophobic efficiency, such as concentration (Bao et al., 2020; Feng & Janssen, 2021; Zhao & Meissener, 2017), contact time and curing time. A lot of research confirmed that samples treated with a brush demonstrate a lower polymerization amount compared to those treated with capillary absorption(Feng & Janssen, 2021; Hansen et al., 2018; Soulios et al., 2019). Van Besien et al. (Van Besien et al., 2020) found that samples treated with a brush yield a lower efficiency in performance and less agent impregnation in the substrate due to the short contact time.

In contrast to typical application approaches in practice, most research was conducted in laboratory and material samples were treated with brush or capillary absorption to obtain homogeneity (Aktas et al., 2021; Bao et al., 2020; Cultrone & Madkour, 2013; Soulios et al., 2019). The comparison of performance efficiency in lab tests and realistic walls remains uncertain. Because the on-site treatment application was always subjected to temperature and relative humidity fluctuations, the efficiency under realistic application approaches may not be as optimal as in the lab. Due to a scarcity of research, it is hard to confirm that lab tests accurately represent the onsite performance.

This research aims to examine the hydrophobic efficiency of a siloxane-based water-repellent solvent under a practical application approach and to determine how environmental circumstances impact the efficiency of the water-repellent agent. In this study, a vertical setup was designed to mimic a real wall on the field and treat the samples with spray-flow as in practice. A series of tests were conducted to investigate the hygric performance of hydrophobized Bentheim Sandstone treated with spray flow under different temperature and moisture content.

2 Material and Methods

2.1 Bentheim Sandstone

Bentheim Sandstone (BS) is a quartz arenite, deposited during the Valanginian (Early Cretaceous) near the western edge of the Saxony basin, Germany (Klein et al., 2001). The sandstone outcrops near the Dutch-German border in region of Bad Bentheim and Gildehaus(Wim Dubelaar & Nijland, 2015). Bentheim Sandstone is often used in the Netherlands as building stone since the 15th century. In Flanders, Belgium, Bentheim is used as a building stone since the 15th century (e.g. Sint Rombaut Tower in Mechelen) and was in the 19th and 20th century used as replacement stone.

2.2 Water Repellent Treatment

The water repellent treatment is carried out with the agent REDISIL S, a modified oligomeric siloxane of the methyl-ethoxy type, dissolved in aliphatic solvents. REDISIL S is a commercial product which contains 10% siloxanes as active ingredient, and is typically applied on porous materials like brick, natural stone and lime mortar (*N.V Rewah, F.Oleofur, 2018.*). Despite the fact that this commercial product is technically approved, there are few studies about the efficiency of this solvent based siloxanes product.

2.2.1 Vertical setup

In this research a vertical setup is designed to mimic a real wall, in which cylinder samples were placed for water repellent treatment. Fifteen Ø 5cm*5cm cylinder samples were treated on the setup. Samples were treated with a typical backpack spray (0.25-1 L/m²) according to the product info sheet(*N.V Rewah, F.Oleofur, 2018.*) in order to reflect standard installation practice. The specimens were treated by spraying over the surface with constant low pressure in case of atomization of the product. After 30 minutes, the samples were given a second wet-on-wet treatment by spraying from bottom to top. Following the second treatment, samples were placed in a climate chamber, still wrapped in parafilm, to allow polymerization to work out at 50% RH and 20°C. It should be noted that the samples were wrapped around the sides to ensure a one-dimensional uptake of the agent and to avoid any other effects from placing the samples in the setup. Afterwards, the samples have been inspected visually to verify that no local infiltrations between the substrate and parafilm compromised this approach. Fifteen

Bentheim Sandstone samples were treated as a group on the vertical setup and tested to study the hydrophobicity efficiency (Fig 1). After treatment, the samples were placed in climate chamber to let polymerization work out until all samples reach a stable weight.

Figure 1. Cylinder samples of Bentheim Sandstone were placed on the vertical setup for spray treatment. The samples were left in the setup for 30 minutes until a second treatment from bottom to top was performed by spraying

2.2.2 Environmental circumstance investigation

Considering the various climate conditions when applying the treatment, a series of tests was conducted to further understand the influence of different temperature and water content on the efficiency of the water repellent treatment. As indicated in the agent's product info sheet, the REISIL agent should be applied between 5-30 °C on a dry surface (*N.V Rewah, F.Oleofur, Technical Data Sheet, 1-3, 2018*.). Three groups of spray-treated samples were cured at 5°C, 20°C, 30 °C respectively after treatment to simulate different temperatures in practice, as shown in Table.3. Sets of partly (50%) saturated and effective saturated (100%) samples were also treated and polymerized in a 20°C, 50% RH climate chamber to simulate the situation of a wet wall. Each group consisted of 3 samples.

Table 1. Determination tests of environmental circumstances

1	Vertical set up + polymerization in 20°C, 50% RH
2	Vertical set up + polymerization in 5°C, 50% RH
3	Vertical set up + polymerization in 30°C, 50% RH
4	Vertical set up + polymerization in 20°C, 50% RH - Fully saturated samples
5	Vertical set up + polymerization in 20°C, 50% RH - Partly saturated (50%) samples

2.3 Tests

To better understand the wetting and drying process, four main factors that characterize the hydrophobicity of the substrate were selected and tested in this study: (i) change in open porosity (φ) (moisture storage), (ii) contact angle, (iii) reduction of the water absorption coefficient (Acap) (moisture transport), and (iv) change in vapor diffusion resistance factor (µ) (moisture transport).

The open porosity were tested in accordance with EN 1936 (*EN 1936: 2006.*) when the samples acquired a steady weight after treatment. All of the specimens were completely immersed.

$$\varphi = \frac{m_s - m_d}{m_s - m_h} * 100\ \% \tag{1}$$

where m_s is the mass of the saturation specimen [g], m_d is the mass of dry specimen [g], and m_h is the mass of specimen immersed in water [g].

The contact angle(*ASTM D5946-17, 2017.*) and Karsten tube test (*RILEM II.4 Test Method*, 2006) determine the instant effect in waterproofing for hydrophobized samples. Moisture absorption was assessed at regular intervals in the Karsten tube test: 5 min, 10 min, 15 min and 24 h. The 5-minute measurement ensures that there are no obvious cracks or leakage in the setup. And the difference in absorption between 5 minutes and 15 minutes was measured as Wa-K15 [ml], to evaluate the effectiveness of the hydrophobicity in a short period of time.

$$Wa\text{-}K15=V(15min)-V(5min) \tag{2}$$

Where V(15min) means absorbed amount of water after 15 minutes; V(5min) means absorbed amount of water after 5 minutes. The water absorption at 24 h was also examined in order to better understand the treatment efficiency over a long period of time, as was the case in research by Lubelli (Lubelli, 2011). The result can be expressed as Wa-K 24 [ml] :

$$Wa\text{-}K24=V(24h)-V(5min) \tag{3}$$

The capillary absorption coefficient (Acap, kg/m².$s^{0.5}$) is measured to test the efficiency of water repellent in water absorption (*EN 1925, 1999.*).

$$Acap = \frac{Mi-Md}{A\sqrt{t}} \tag{4}$$

Where M_i [kg] is the successive masses of the specimen during test, M_d [kg]is the mass of the dry specimen, A [m^2] is the area of the side immersed in water and t means time elapsed from the beginning of the test until the times at which the successive masses M_i were measured.
The vapor diffusivity of hydrophobized material was measured according to EN 12572 (*ISO 12572:2016*), and determines the vapor diffusion resistance factor (µ) of the material derived from the weight change over time. Saturated salt solutions are used to impose two RH conditions: 0%-50% and 50%-100%.

3 Results & Discussion

3.1 Hygric Properties

After treatment, the Bentheim Sandstone samples absorbed an average of 134g/m^2 of the active agent in this study. The spray-treated Sandstone samples only demonstrate a slight decrease in open porosity after hydrophobization. Significant reductions in water absorption coefficient were observed, with little alteration in pore structure and vapor permeability.

Table 2. Hygric properties of untreated and treated Benteim samples

Bentheim Sandstone	Untreated (Std)	Treated (Std)
Open Porosity [%] (Std)	**21.79 (0.82)**	**20.23 (0.68)**
Bulk Density [kg/m3] (Std)	**1986.29 (71.00)**	**2002.24 (9.97)**
Contact angle	**-**	**115.72 (9.59)**
Water absorption coefficient [kg/m².s$^{0.5}$] (Std)	**0.560 (0.0114)**	**0.001 (0.00001)**
Wa-K15 [ml] (Std)	**50.175 (39.53)**	**0**
24h water uptake[kg/m²] (Std)	**0.278 (0.162)**	**0.123 (0.012)**
Vapor diffusion resistance (0-50% RH) (Std)	**19.96 (1.45)**	**28.02 (2.44)**
Vapor diffusion resistance (50-100% RH) (Std)	**9.04 (1.04)**	**9.43 (2.46)**

As a non-hygroscopic material, Bentheim Sandstone is not dominated with small pores. the siloxanes molecular attached on pore walls (Van Besien et al., 2020), and inevitably reduce the pore radius. Some siloxanes molecular size are larger than the pore throat and clogged the pores, converting open pores to closed pores, as well reducing pore connectivity. The small percentage of clogged small pores reduces the connectivity in pore structure, as well increases vapor permeability resistance.

The Bentheim Sandstone samples exhibit almost perfect hydrophobicity after treatment. The treated samples absorbed almost no water after treatment, with a over 90 degrees contact angle on the surface. The samples absorb very little water after 24 hours of water contact, indicating that the siloxanes based water repellent is very effective on sandstone samples, even under continuous water content. However, there are still some inhomogeneous treated samples after spray, and could cause the ineffectiveness of the agent. The water repellent migrates into the substrate via capillary, increasing the surface tension, promoting surface run-off, reducing contact time with moisture and forming the first barrier (a strong hydrophobized layer near the surface) to moisture absorption as a consequence of increasing contact angle. As the hydrophobic impregnates, the siloxane molecules attached to the pore walls and formed a partially hydrophobized region inside, which is the second moisture absorption barrier.

3.2 Environmental Circumstances

Figure 2 shows that environmental factors have a minor impact on open porosity after treatment. The minor difference is in the allowed range of material variation, with a deviation between 1.2%. The capillarity of samples polymerized under different environment circumstance was similar, less than 0.002kg/m^2.s$^{0.5}$ after the treatment, indicating the same efficiency regardless of environmental circumstance.

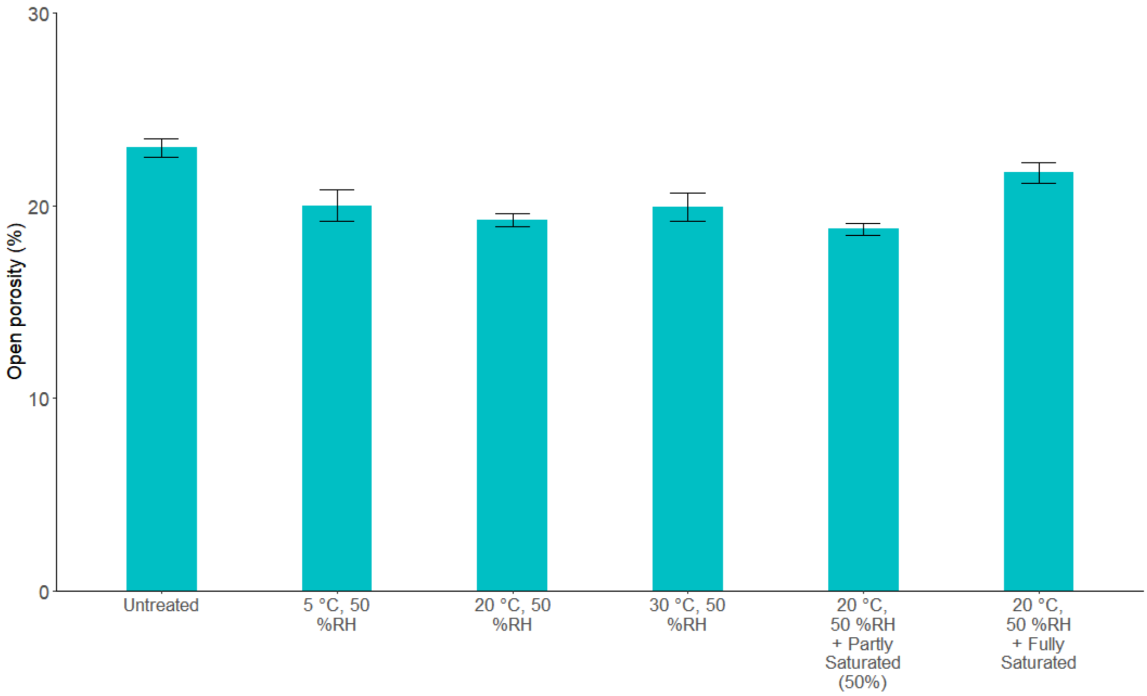

Figure 2. The open porosity of Bentheim Sandstone samples under different environmental circumstances and saturation degrees

Table 3. The capillary rate and 24h water absorption under different environmental circumstances and saturation

Treatment	Open Porosity [%]	Acap [kg/m².s$^{0.5}$]	24h Uptake [kg/m²]
Untreated	21.79 (0.47)	0.560 (0.0114)	0.278 (0.162)
20°C 50% RH	19.25 (0.34)	0.0012 (0.0003)	0,127 (0.022)
5°C, 50% RH	20.00 (0.83)	0.0015 (0.0001)	0,113 (0.015)
30°C, 50% RH	19.93 (0.74)	0.0018 (0.0001)	0,129 (0.018)
Fully saturated +20°C, 50% RH	21.71 (0.53)	0.0018 (0.0003)	0.136 (0.042)
Partly saturated (50%) +20°C, 50% RH	18.76 (0.31)	0.0015 (0.0004)	0,115 (0.016)

In practice, the fluctuating temperature and moisture content of a wall are unavoidable, when applying the treatment under natural climatic conditions. In this study, sandstone samples were treated after complete saturation to simulate a practical wall after rain. Only a minor increase in capillarity was observed when compared to the application on a dry material, suggesting that the moisture content of the wall has little impact on the hydrophobicity. This implies that the water repellent product is also effective on wet walls. Furthermore, the water repellent agent works effectively within the temperature range specified in the product's info sheet.

4 Conclusion

The present study examined the effectiveness of water repellent treatment on the hygric properties of Bentheim Sandstone through a practical application approach. The spray-treated samples illustrate almost perfect hydrophobicity after treatment. The water repellent increases the surface tension and prevents moisture invasion from surface without significantly changing the vapor permeability. Actually, various environmental circumstances exhibit little influence on the water repellent efficiency, indicating that application is also effective on a cold day or after rain. The practical application doesn't have to be conducted in perfect circumstances.

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