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Janz, Mårten

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LUND UNIVERSITY

PO Box 117
221 00 Lund
+46 46-222 00 00

Moisture transport in a sedimentary calcareous sandstone and its effect on different degradation processes

Mårten Janz
Division of Building Materials, Lund University, Box 118, SE-22100 Lund, Sweden

ABSTRACT

Degradation of historical buildings and monuments is an increasing problem in most countries around the world. The cost of preserving these buildings is substantial. In order to take the correct measures to preserve the buildings, the process causing the degradation must be identified. One technique for excluding or identifying probable causes of damage to porous materials is demonstrated in this paper. The technique is based on moisture transport calculations and is exemplified using sedimentary calcareous sandstone from the island of Gotland, Sweden. The calculations are made at very high moisture levels, both under and over capillary saturation. A traditional Fickian approach is used for the calculations up to capillary saturation. Above capillary saturation, a mathematical method based on the solubility of air is used. The calculations then lead into a discussion of which different degradation processes, such as frost action and leaching, are affected by the calculated moisture movements and moisture levels.

Keywords: Moisture transport, capillary transport, degradation processes, frost action, leaching, sandstone.

1 INTRODUCTION

Gotland sandstone, a sedimentary calcareous sandstone, is one of the predominant materials in historical buildings in the Baltic region (Lindborg 1990). Since Gotland sandstone is soft and easy to work, it was often used in sculptural decorations and facades on churches, castles, and private dwellings during the seventeenth and eighteenth centuries. Thus, the stone was often used in buildings that are now considered an important part of our cultural heritage.

Increasing degradation of this Gotland sandstone can be observed. One of the more famous buildings affected by this degradation is the Royal Palace in Stockholm, where the facade and balustrade are of Gotland sandstone.

The main explanation for this gradual degradation of Gotland sandstone is that air pollution such as acid rain is breaking down the stone. The increasing air pollution this century has possibly resulted in an accelerating rate of degradation.

Natural physical degradation of the stone (physical weathering) is, however, also taking place. The most important agent of physical weathering in the Nordic climate is probably frost action. Wessman (1997) has shown that the frost resistance of the stone studied here is rather low. Furthermore, Wessman has shown that a very low external salt concentration has an aggravating effect on the frost scaling. A concentration of 0.25% of NaCl in the surrounding solution is enough to cause severe scaling of the sandstone. Consequently the gradual deposit of air pollutants on the surface of the sandstone might also decrease its frost resistance. The scaling that Wessman observed in the laboratory is similar to what is seen in nature.

A third possible explanation for the degradation is leaching of calcium carbonate (the cement that binds the sand grains together) which is soluble in water (Fagerlund 1994). The buildings and sculptural decorations with damaged sandstone are often over 300 years old. During this time, large quantities of water have been transported in and out of the stone due to the very high capillarity of the stone. Frogner (1998) has modelled carbonate chemistry in the pores of Gotland sandstone and has shown that pore water becomes saturated with calcite within 100 seconds. Thus the degradation due to leaching is controlled by the transport of these ions from the pores. The leaching is accelerating due to air pollution since the pollution lower the pH of the water transported into the stone.

There is currently a lack of knowledge of which degradation process is dominant. Probably several different mechanisms have a synergistic effect. However, water and water transport play a decisive role in all the mechanisms described. Water is a transport medium for the different chemical substances that have a negative affect on the sandstone. It is also the direct cause of frost damage and leaching. Therefore, it is very important to be able to calculate moisture movement in the stone at the very high moisture levels at which degradation takes place. This article presents calculations of the water transport and discusses durability problems resulting from the calculated water transport and the moisture levels obtained. Water transport is calculated for levels both under and over capillary saturation. The gradual absorption above capillary saturation is very important since several durability problems occur at moisture levels above capillary saturation. In order to calculate the water transport, a traditional Fickian approach is used for the calculations up to capillary saturation. Above capillary saturation a mathematical method suggested by Fagerlund (1993) is used.

2 MATERIAL

The material studied is a sedimentary calcareous sandstone with a homogeneously light grey colour. The porosity of the sandstone is approximately 23% and the density 2059 kg/m^3 (Janz 1997). Thin section microscopy of the stone shows that it consists almost entirely of quartz grains glued together by calcareous cement with empty spaces constituting the porosity. According to Wessman and Carlsson

(1995) the size of the grains is 0.1–0.2 mm. Measurement of the pore size distribution using a pressure plate extractor shows that the pores of the stone are quite coarse. The volume of approximately 50% of the pores is larger than 0.01 mm (Janz 1997).

To calculate the water movement up to capillary saturation it is necessary to know the moisture storage capacity, the relation between Kirchhoff's flow potential ψ [kg/(m·s)] (or the moisture diffusivity) and the moisture content mass by volume w [kg/m^3]. These relations are given in Janz (1997) see Figure 1 and Figure 2. The Kirchhoff's flow potential reported in Janz (1997) was calculated from a series of sorption coefficients that in turn were obtained from capillary absorption tests. The moisture storage capacity in the superhygroscopic range was measured using pressure plate and pressure membrane extractors. In the hygroscopic range different climate chambers with saturated salt solutions were used.

In order to calculate the long-term water absorption above capillary saturation, a measure of this uptake must be used (Fagerlund 1993). Such a measurement is presented in Janz (1997), see Figure 3.

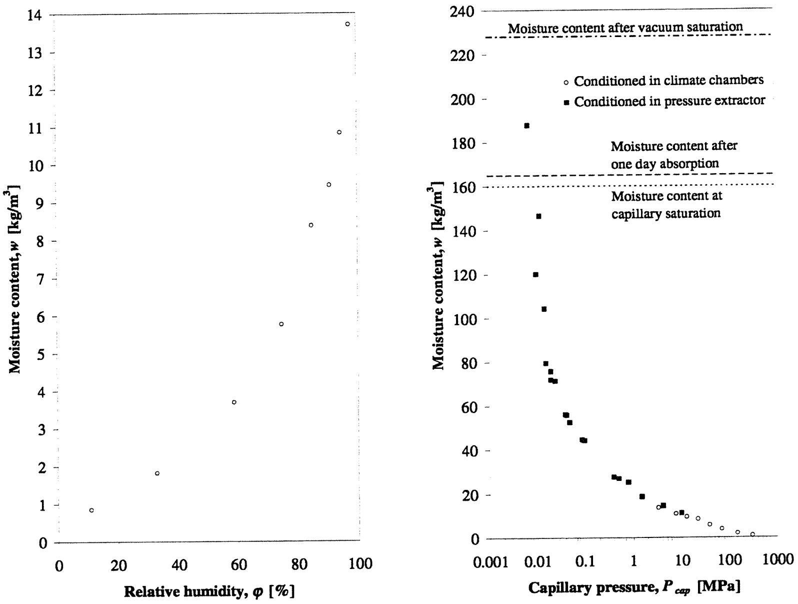

Figure 1 *Moisture storage capacity of Gotland sandstone described by a sorption isotherm (left) and by a water retention curve (right) (Janz 1997). For the sorption isotherm, only measured values within the hygroscopic range are used. The water retention curve shows values from both the hygroscopic and the superhygroscopic range.*

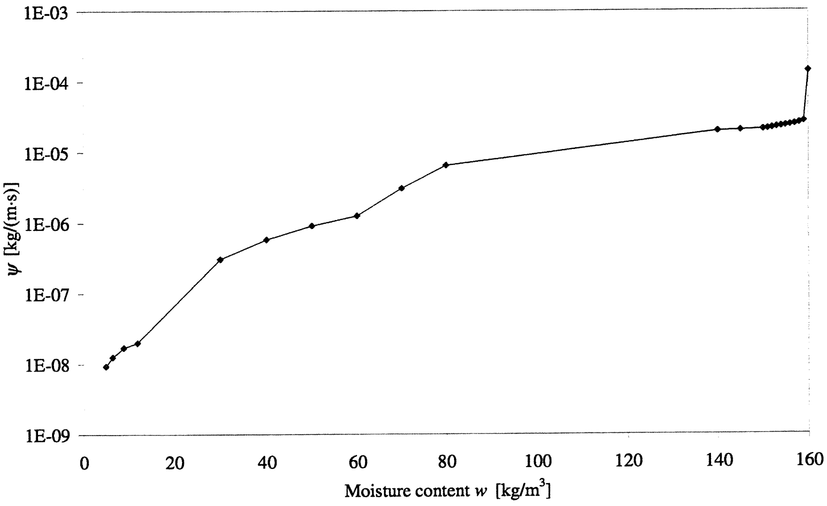

Figure 2 The relation ψ(w) of Gotland sandstone (Janz 1997)

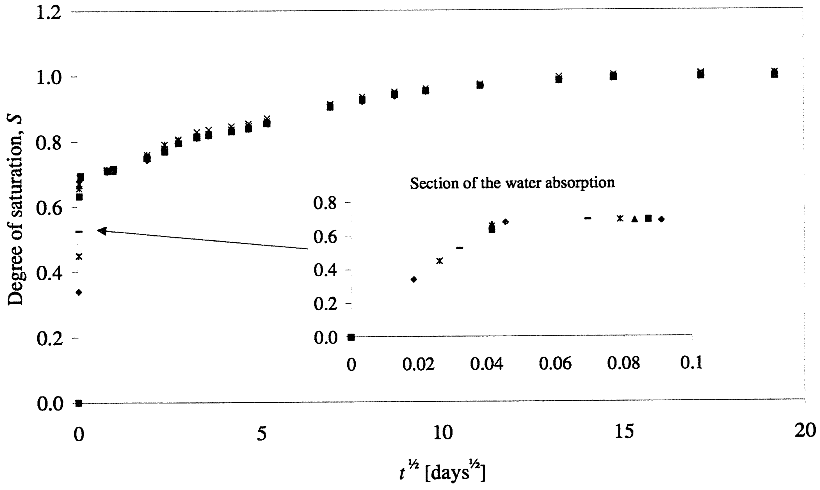

Figure 3 *Long-term water absorption in Gotland sandstone (Janz 1997). The test was performed on specimens dried at 105°C and then exposed to water on one side during the absorption. To prevent evaporation from the specimen, the test was performed in a small chamber with approximately 100% relative humidity.*

3 MOISTURE CALCULATIONS

3.1 Method of calculating water uptake up to capillary saturation

Calculations of the moisture flow are based on Fick's law in one dimension on a thick construction under isothermal conditions. At isothermal conditions it is possible to choose a flow potential for which the moisture diffusivity is always equal to 1. This potential is called Kirchhoff's flow potential ψ [kg/(m·s)], and is advantageous when numerical calculations with the explicit forward difference method are considered (Arfvidsson 1998). Kirchhoff's flow potential ψ is defined by:

$$\psi = \psi_{ref} + \int_{w_{ref}}^{w} D_{\varphi}\, d\varphi \tag{1}$$

that is,

$$\frac{d\psi}{d\varphi} = D_{\varphi} \tag{2}$$

where ψ_{ref} is the reference value at a moisture content $w = w_{ref}$ which can be chosen arbitrarily; D_{φ} is the moisture diffusivity, which is dependent on the moisture content; and φ is any of the following moisture state variables: vapor content, vapor pressure, relative humidity, pore water pressure (or suction), moisture content mass by mass or mass by volume. Moisture diffusivities dependent on any of the moisture state variables listed above can be recalculated to the Kirchhoff's flow potential.

Because of the numerical advantages, the Kirchhoff's flow potential has been used here. When using the Kirchhoff's flow potential, the one-dimensional moisture flow g [kg/(m^2·s] is described by:

$$g = -\frac{\partial \psi}{\partial x} \tag{3}$$

The moisture balance equation expressed with Kirchhoff's flow potential in one dimension becomes:

$$\frac{\partial w}{\partial t} = \frac{\partial^2 \psi}{\partial x^2} \tag{4}$$

The numerical method used is explicit forward differences. Using equation 3 and the notations shown in Figure 4, the flow between two cells can be approximated as:

$$g_{i-½} = \frac{\psi_{i-1} - \psi_i}{\frac{\Delta x_{i-1}}{2} + \frac{\Delta x_i}{2}} \tag{5}$$

In the calculations performed here, the dimensions of all the cells are equal, that is, $\Delta x_{i-1} = \Delta x_i = \Delta x_{i+1} = \Delta x$. Moisture balance then gives the new moisture content $w_{i,n+1}$ in cell i:

$$w_{i,n+1} = \left(\psi_{i-1,n} + \psi_{i+1,n} - 2\psi_{i,n}\right) \cdot \frac{\Delta t}{\Delta x^2} + w_{i,n} \tag{6}$$

where n is the present time step. Equation 6 is valid for all cells except cell 1 at the boundary, where moisture balance gives:

$$w_{1,n+1} = \left(2\psi_{bound,n} + \psi_{2,n} - 3\psi_{1,n}\right) \cdot \frac{\Delta t}{\Delta x^2} + w_{1,n} \tag{7}$$

where $\psi_{bound,n}$ is the ψ-value at the boundary at time step n. The time step Δt is given by:

$$\Delta t = \left(\frac{dw}{d\psi}\right)_{\min} \frac{\Delta x^2}{3} = \frac{1}{D_{w,\max}} \frac{\Delta x^2}{3} \tag{8}$$

$D_{w,\max}$ is the highest value of D_w in the interval $0 < w < w_{\max}$. The calculations up to capillary saturation are made using the above described theory and numerical method.

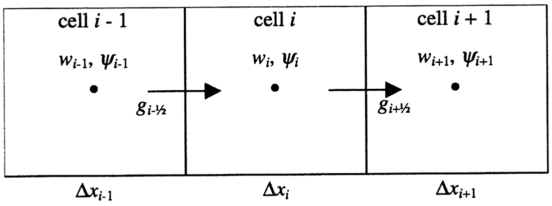

Figure 4. The cells i–1, i, and i+1 with notations

3.2 Method of calculating gradual water uptake above capillary saturation

When actual capillary absorption comes to an end, there is always a certain amount of enclosed air in the pore system. Thus the material is not completely saturated. This enclosed air will, however, gradually dissolve in the water and diffuse to larger pores or out of the pore system to the surface of the material, which will lead to slow water absorption above capillary saturation. The rate of this dissolution is proportional to the solubility of air in water, which is in turn proportional to the internal overpressure ΔP [Pa] in the enclosed air. Thus, the rate of gradual water absorption will be proportional to the dissolution rate of air in water and the diffusivity of air in the pore water. An air bubble that becomes enclosed in a coarse pore during the capillary absorption process is exposed to an internal overpressure that is inversely proportional to its radius. The overpressure is given by Laplace equation:

$$\Delta P = \frac{2\sigma}{r} \tag{9}$$

In equation 9, σ is the surface tension [N/m] and r is the radius of the bubble [m]. The diffusivity of air in the pore water is a function of the pore structures.

According to Fagerlund (1993) and Fagerlund (1995) one can calculate the water volume V [m^3] needed in order to completely dissolve an air bubble of radius r:

$$V = \frac{4\pi}{3}\frac{\rho_0}{s}\left(1 + \frac{P_0}{\Delta P}\right) r^3 \tag{10}$$

where ρ_0 is the density of air [kg/m^3], s is the solubility of air [kg/(m^3 Pa)] and P_0 is the atmospheric pressure.

Using equations 9 and 10 one can calculate that a water sphere with a radius of 69 μm is needed to completely dissolve an air bubble of the radius 10 μm (Fagerlund 1995). Shortly after the end of the capillary process will therefore small air bubbles become water filled. Large air bubbles, on the other hand, will take considerable time to dissolve.

This gradual absorption above capillary saturation is very important since several durability problems such as frost scaling often occur at moisture levels above capillary saturation. Little is known about this slow water absorption process, and consequently the available computer programs can only calculate water absorption up to capillary saturation. The fields of application of these computer programs are therefore limited. However, Fagerlund (1993) examines long-term water absorption above capillary saturation theoretically and experimentally. Fagerlund also proposes a way of modelling this absorption. For reasonably large specimens, water absorption is assumed to take place simultaneously over the specimen body. The degree of saturation over capillary saturation S_a [m^3/m^3] is then defined by:

$$S_a = \frac{V_w - V_{w,cap}}{V_P - V_{w,cap}} \tag{11}$$

where V_w is the volume of water in the specimen [m^3]; $V_{w,cap}$ is the volume of water in the specimen at capillary saturation [m^3]; and V_P is total pore volume [m^3].

The degree of saturation over capillary saturation can, according to Fagerlund (1993), possibly be modelled by:

$$S_a = a t^b \tag{12}$$

where a and b are constants and t [s] is the time, measured from when capillary saturation is reached. The constant a is a is function of the diffusivity of air in the pore system and b is a function of pore size distribution.

By using equation 11 and 12 the constants *a* and *b* are calculated from a long-term water absorption test. For Gotland sandstone, the values of the constants have been calculated from the absorption test shown in Figure 3. These constants are shown in Table 1. Since the long-term water absorption is measured on the sandstone until vacuum saturation is reached, there is no actual need to use equation 11. But in the normal case, the measurement of the long-term water absorption is not measured until the absorption comes to a complete end. For some materials such as concrete, it will take a considerable time, perhaps years, until the absorption stops. Thus, in the normal case the gradual absorption must be extrapolated, e.g. with equation 11. For some concrete, the constants *a* and *b* were approximately $6.7 \cdot 10^{-3}$ and 0.26 respectively (Fagerlund 1993).

Table 1 The constants a and b calculated from Figure 3 (Gotland sandstone)

Specimen	*A*	*B*
1	$6.953 \cdot 10^{-4}$	0.4430
2	$6.074 \cdot 10^{-4}$	0.4509
3	$6.391 \cdot 10^{-4}$	0.4495
4	$8.121 \cdot 10^{-4}$	0.4359
5	$9.024 \cdot 10^{-4}$	0.4278
6	$6.434 \cdot 10^{-4}$	0.4502
Mean value	$7.166 \cdot 10^{-4}$	0.4429
Standard deviation	$1.161 \cdot 10^{-4}$	0.0094

3.3 Results

3.3.1 Moisture calculations up to capillary saturation

The calculations were made in one dimension on a 0.5 m thick construction under isothermal conditions. Such a construction could be a homogenous wall or a sculpture. Three different calculations were done, each using 25 cells. At the boundary different concentrations are prescribed, i.e. essential boundary conditions are used. The relation between Kirchhoff's flow potential and moisture content used is prescribed in Figure 2. This relation is only valid for absorption, but is here used for both the wetting and drying process.

Example 1 – One day's capillary absorption

The initial moisture content is 7 kg/m^3, which according to Figure 1 corresponds to 80% relative humidity. The boundary is subjected to rain for 24 hours. The result of the calculation is shown in Figure 5, which shows that capillary saturation reaches a depth of approximately 0.35 m after 24 hours. After only one hour, capillary saturation reaches a depth of 5 cm (i.e. the outer surface is affected after a brief period of rain).

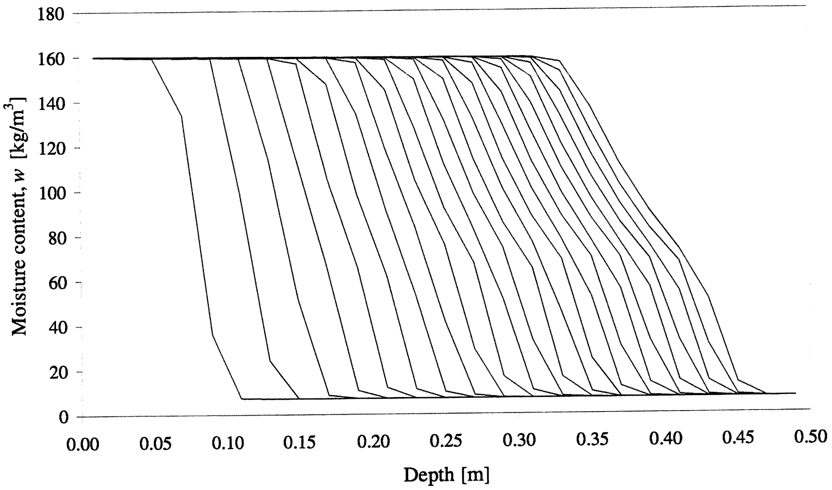

Figure 5. Example 1: Moisture distributions drawn at hourly intervals

Example 2 – Cyclically repeated rain

The initial moisture content is 7 kg/m^3 (80% relative humidity). The boundary is subjected to rain for 1 hour, followed by 23 hours drying out at 80% relative humidity. This cycle is repeated 8 times. Figure 6 shows the moisture distribution directly after rain and directly after a drying-out period. Capillary saturation is obtained to a maximum depth of roughly 7 cm. However, the depth influenced by the rain gradually increases. After 8 days (192 hours), the moisture content has increased to a total depth of 45 cm. The gradually increasing moisture penetration can be clearly seen in Figure 7.

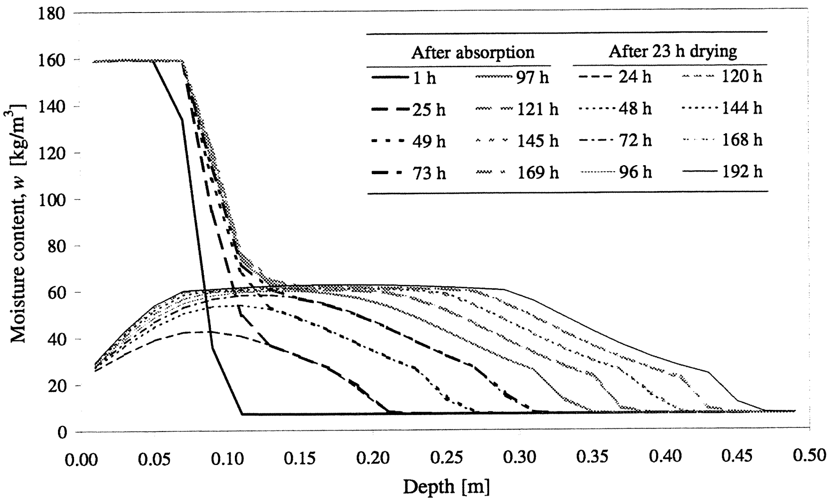

Figure 6. Example 2: Moisture profiles drawn at different times

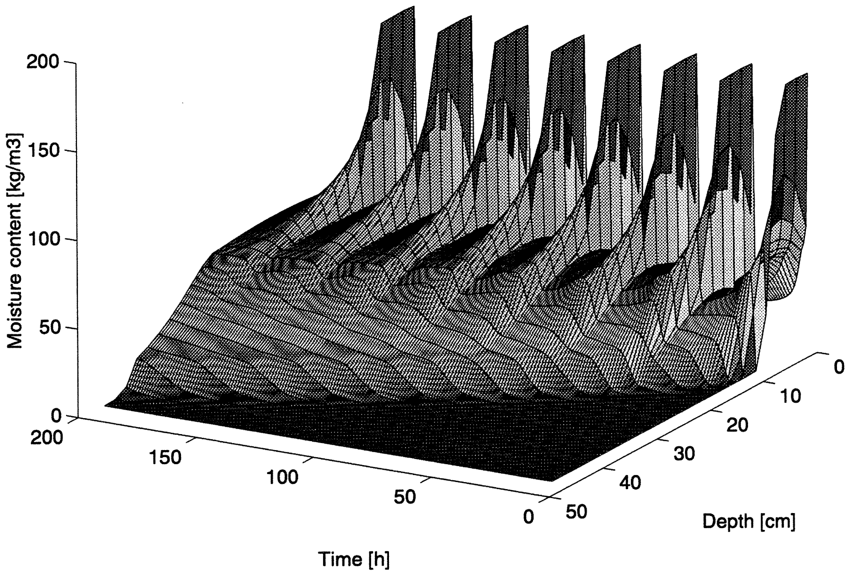

Figure 7. Example 2: Moisture penetration during cyclically repeated rain

Example 3 – One hour's rain followed by a drying-out period

The initial moisture content is 7 kg/m^3 (80% relative humidity). The boundary is subjected to rain for 1 hour, followed by 720 hours drying out at 80% relative humidity. The total time is one month. As shown in Figure 8, the moisture content decreases rapidly directly after the rain. But, the drying time becomes considerably longer at the end of the sequence. Figure 9 shows the moisture distribution after 1 hour, 15 days, and 30 days. The difference between 1 hour and 15 days is very large, but the difference between 15 days and 30 days is almost negligible.

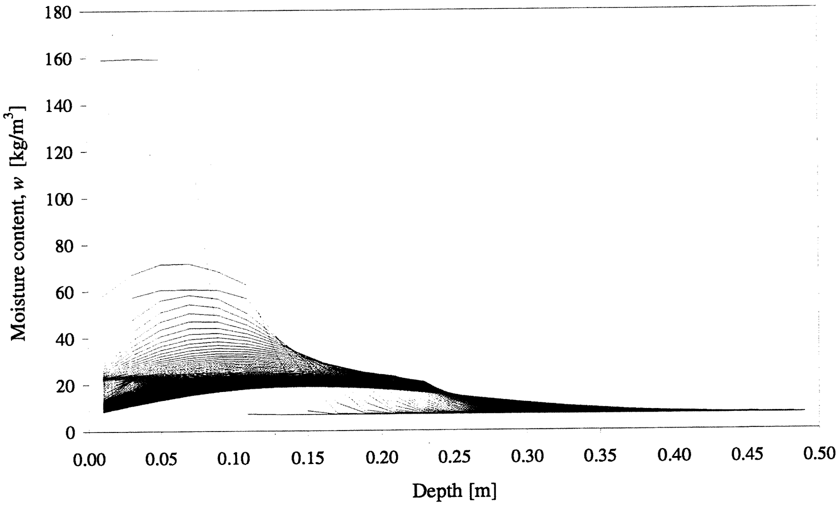

Figure 8. Example 3: The first moisture profile is drawn after 1 hour, thereafter every 3 hours.

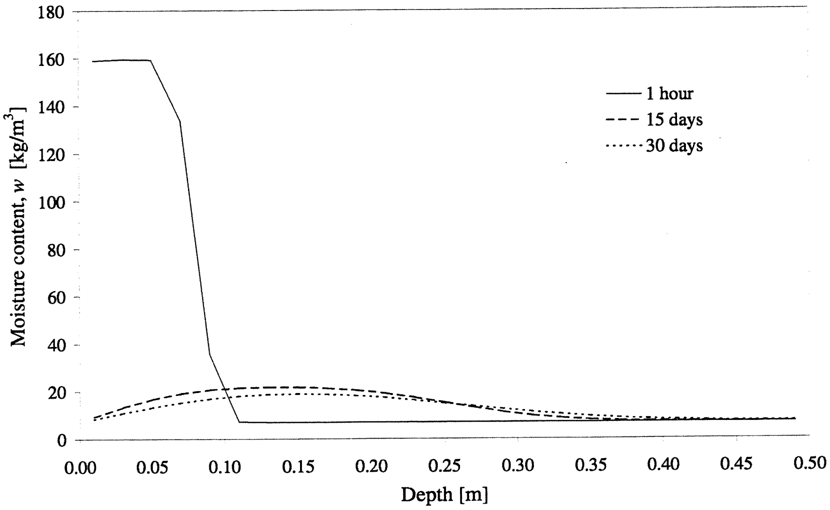

Figure 9. Example 3: Moisture profiles drawn after 1 hour, 15 days, and 30 days

3.3.2 *Moisture calculation above capillary saturation*

Wessman (1997) has measured critical degrees of water saturation S_{crit} (i.e. the degree of saturation above which deterioration due to frost action is likely to occur) for different natural stones. She states that the sandstone studied here has a S_{crit} of about 0.89–0.96. The degree of saturation is defined as the quotient between the volume of water in the specimen and the total pore volume. The critical degree of saturation over capillary saturation will consequently be (see equation 11):

$$S_{a,crit} = \frac{S_{crit} - S_{cap}}{1 - S_{cap}} \tag{13}$$

where S_{cap} is the degree of saturation when the capillary absorption comes to an end. According to Janz (1997), S_{cap} is 0.70. The value of $S_{a,crit}$ will consequently be 0.63–0.87. Equation 12 gives the time until the critical degree of saturation is obtained:

$$t = \sqrt[b]{\frac{S_{a,crit}}{a}} \tag{14}$$

With a and b values of $7.166 \cdot 10^{-4}$ and 0.4429 respectively (see Table 1) and equation 14, the time before there is actual risk of degradation due to frost action will be somewhere between 51 and 106 days. Thus, it must be noted that the coefficients a and b are based upon the hypothesis that the long-term absorption shown in Figure 3 occurs simultaneously across the entire cross-section.

4 CONCLUSIONS

The type of calculation done here is a powerful tool for identifying which degradation processes may be affecting different structures and different positions in structures. Combining information about degradation processes, the location of a damaged structure, and the surrounding environment facilitates excluding some processes and singling out others as probable causes of damage.

For example, it can now be stated that, providing the constants *a* and *b* in equation 13 are generally applicable, degradation due to frost action on Gotland sandstone can only occur when the stone is exposed to water for 50 days or more. In other words, the stone must be part of a structure or sculptural decoration that receives a continuous supply of water or has limited possibility of drying out. Examples of parts of structures that would meet these requirements include a facade at a damaged drainpipe or a part of a structure that is exposed to groundwater.

The calculation of the effects of cyclically repeated rain in example 2 shows that a very large quantity of water is absorbed and thereafter dried out, especially in the 7 cm nearest to the surface. This cyclical absorption and desorption corresponds to conditions such as repeated driving rain. Reiterated water absorption and desorption will result in a convective transport of calcite ions out of the stone. According to Fagerlund (1994) and Frogner (1998), this will lead to a leaching of the calcium carbonate that glues the sand grains together in the stone.

The water may also transport chemical substances derived from air pollution, which can damage the stone. Frost action can, however, be excluded as a contributor to the degradation process in structures exposed to a moisture load similar to that in example 2 (cyclical repeated rain) because the moisture levels achieved are not sufficient for degradation due to frost action.

Before far-reaching conclusions can be drawn from these calculations, the mathematical models used must be improved and the calculations calibrated against measurements. Particular attention needs to be paid to the calculations above capillary saturation. The transport mechanisms for above and under capillary saturation also need to be combined in a single mathematical model. There is also an obvious lack of knowledge about transport properties in different materials at high moisture levels, both above and under capillary saturation. Here, too, much work is called for. Finally, there must be further study of the degradation processes acting on different materials.

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