Moisture in Self-levelling Flooring Compounds. Part II. Sorption Isotherms

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ABSTRACT

Moisture sorption in self-levelling flooring compounds was investigated by using a sorption balance. Results are presented as sorption isotherms. Influence of temperature, age, water to binder ratio and carbonation are also presented. An increased temperature and increased water to binder ratio gives lower moisture sorption. Results from scanning curves indicate that, when changing sorption mode, even a minute change in moisture content may result in a significant change in relative humidity in the material.

Key words: sorption isotherm, water vapour, self-levelling flooring compound, sorption balance.

1. INTRODUCTION

Recently, the connection between the indoor environment and occupants health has gained an increased interest. Moisture has been shown to be an important factor as it promotes chemical reactions and biological growth. This is further discussed in the first paper [1] of this series of two papers dealing with moisture properties of self-levelling flooring compounds.

Moisture in materials has different properties and is of different interest depending on how hard the water molecules are bound to the surface and the structure of the material. This is important as not all water inside materials contribute to the processes that may influence the indoor environment. The weakest bound water is here the most interesting as it supports biological growth, transport of substances, etc. [2].

The strongest bound water in a cementitious material is chemically bound as reaction products in the hardened cement paste and is part of the solid structure of the material. As this water is
strongly bound in the structure and does not leave the material under normal circumstances it is of no interest concerning indoor environmental aspects.

Physically bound water stands in equilibrium with the water vapour in the surrounding atmosphere. The amount of water in a material, excluding chemically bound water, is normally given as a sorption isotherm, i.e., the relation between the moisture content (the mass of water per mass of dry material) and the relative humidity (RH) at equilibrium.

Physically bound water can either be adsorbed to surfaces, absorbed in the structure of the material or capillary condensed in pores. The first layer of adsorbed water is the strongest bound, most immobile and remains non-frozen at -55 °C [3]. This water behaves as part of the solid and corresponds to the monolayer moisture content [2]. A monolayer can be seen as when all of the dry matter is covered with one layer of water molecules [4]. Further added water is more mobile. When sufficiently abundant part of this water may allow for chemical reactions and microbial growth.

In porous materials water is bound not only due to adsorption, but also due to capillary condensation. Water molecules then condense on concave water menisci. Pores with radii less than a certain critical radius are then completely water filled. The curvature of the menisci corresponds to a certain equilibrium RH. A smaller radius corresponds to a lower equilibrium RH. When a material is in equilibrium with low RH, only small water menisci can exist, i.e., only small pores can be completely water filled. At higher RH, larger water menisci can exist and pores with larger pore radii can thus be water filled. In a fine porous material, capillary condensed water is the dominant contributor to transport processes, chemical reactions and microbial growth.

A consequence of capillary condensation is sorption hysteresis [4], i.e., a material will contain different amounts of water in equilibrium with a certain RH, depending on whether the material is drying or taking up moisture, Fig. 1. A material taking up moisture never contains more water than a drying material at the same RH.

As can be seen in Fig. 1, a sorption isotherm contains two main curves, one for desorption and one for absorption. If a previously desorbing material starts to absorb moisture it follows a scanning curve from the desorption isotherm towards the absorption isotherm and vice versa. A result of this is that a small increase in moisture content may lead to a large increase in RH. The consequence of this for moisture related processes is not fully known, but it is probable that RH is not the only parameter describing the rate of chemical and biological processes [2] and that moisture content also may be important, which has been seen for microbial growth on food stuffs [5].
The sorption isotherm is, as the name indicates, only valid at a certain temperature. An increase in temperature at constant moisture content results in an increase in RH [2, 6], opposite to the behaviour of air, where an increase in temperature results in lower RH. For concrete, variations of the sorption isotherm within normal temperature ranges are generally neglected, although studies by for example Nilsson [7] show a temperature dependence of about 0-0.4 % RH per ºC depending on moisture condition. An increase in temperature normally increases the rate of chemical reactions [8].

Chemical degradation reactions are dependent on transport properties, as reactants have to come in contact with each other. The critical RH limits at which processes start to take place are somewhat diffuse. A practical value often used in Sweden for concrete slabs with bonded PVC-flooring is 85 % RH. Generally an RH of at least 80 % is needed for biological growth, although some species can grow at lower RH [2, 9].

This paper deals with physically bound water in the RH-range of 10-95 %. Measuring moisture at higher RH levels than 98 %, requires other measurement techniques, see e.g. [10-13]. Measuring moisture down to 0 % RH gives incorrect values for self-levelling flooring compounds (SLC) as chemically bound hydrate water from ettringite is partly non-reversibly released. This has been observed during vacuum drying and drying over desiccants [14] and drying at temperatures above 60 °C [15]. We have noted a significant decrease in mass when drying specimens from 10 % to 0 % RH (not shown in this paper). This mass decrease corresponded to more than 5 % moisture content, i.e., more than the total sorption capacity between 10 % and 95 % RH.
2. MATERIALS AND METHOD

2.1 Materials

Sorption isotherms of three different types of commercial SLC were tested. The three tested commercial products were based on Portland cement, calcium aluminate cement, calcium sulphate and small amount of silica fume. The three tested product were:

A. Normal SLC with water to binder ratio of about 1.0. This product is generally used in non-industrial constructions.
B. Rapid-drying SLC, which is similar to the above product, but is used with lower water to binder ratio (about 0.7) to give the product a higher degree of self-desiccation. This product is mainly used in renovation as it allows foot traffic in 1-2 hours and final covering in 1 day.
C. SLC for industrial floors, also containing slag as a binder and a higher amount of polymer to increase the abrasion resistance [16]. The water to binder ratio is about 0.6. The final product allows medium heavy rolling equipment.

The products were mixed according to the manufacturers guidelines and left to hydrate, the first 24 hours in open air, approximately 20 °C and 50 % RH, and after that in sealed glass jars. Small amounts of water were added to the specimens in the jars to keep the RH at 100 %. Test specimens were cut from the centre of cast cylinders to avoid edge effects and carbonation. Specimens had hydrated for a minimum of 12 months prior to testing, if not otherwise stated. The side lengths of the specimens were in the order of 4x4x3 mm with masses in the order of 100 mg. Carbonated specimens were exposed to atmospheric conditions for three months. Validation of carbonation was made in parallel measurements (not on the tested specimens).

2.2 Method

A sorption balance (DVS 1000, Surface Measurements Systems, London, UK) was used to determine sorption isotherms and scanning curves. This type of instrument has previously been used in the study of sorption on various materials, for example, food stuffs [17], plant material [18] and inorganic building materials [19]. With this instrument it is possible to measure sorption when the RH is changed in steps and as ramps. A flow of dry nitrogen gas is divided into two gas streams of which one is saturated with water vapour and the other left dry, see Fig. 2. By mixing different proportions of the gases, RH between 0 and 100 % can be generated, although for practical reasons, the upper limit is 98 % RH.
Figure 2 – Schematic picture of the DVS 1000 sorption balance. A flow of dry nitrogen gas is divided into two gas streams where one is saturated with water vapour, by bubbling through liquid water (S) and one is kept dry. The flow rates are controlled by two mass flow regulators (Rw and Rd). By mixing different proportions of the two gases, the desired RH can be generated.

Test cycles with several measurement steps and ramps have been used in the present measurements, Fig. 3. The test specimen is exposed to these relative humidities and the mass change of the specimen is continuously measured. A normal test cycle for one specimen lasted 8-12 days in the present study. The environment in the sorption balance was free from carbon dioxide during the measurements. The trueness (precision) was ±1.0 % RH and the balance resolution was 0.1 μg.

Figure 3 – Typical RH test cycle used in the present study. The test cycle starts and ends at 95 % RH and includes two ramps for scanning curves, one from desorption to absorption and one from absorption to desorption.

Scanning ramp rates were about 100 minutes per % RH. After each scanning ramp, the relative humidity was kept at the final ramp value for 120 minutes validating that each scan was close to equilibrium. The change in mass during these 120 minutes corresponded to less than 0.03 % moisture content.
2.3 Performed tests

Sorption isotherms for the three self-levelling flooring compounds described above were measured with the sorption balance. The influence of different amounts of mixing water, different temperatures, age and carbonation were also measured.

2.4 Evaluation

Steady state measurements were made until the specimens had almost reached equilibrium. The final part of the curve was then curve-fitted and extrapolated with the equation

\[ m(t) = m_0 + (m_f - m_0)e^{-k(t-t_0)} \]  

(1)

where \( m(t) \) is the mass at time \( t \), \( m_0 \) is the initial mass at the start of the curve-fitting, \( m_f \) is the final mass, \( k \) is a constant and \( t_0 \) is the initial time at the start of the curve-fitting, see Fig. 4. The curve fitting was done in MATLAB 6.5 with a least square method. The correction never corresponded to more than 6 % of the total mass change in each time step. The final sorption isotherms shown in the figures are curve fitted between the measurement points with a general spline function.

![Figure 4](image-url)

*Figure 4 – Extrapolation of the final part of a measurement step in the sorption balance. Solid line is measured values, dotted line curved fitting of measured values and dashed horizontal line extrapolated equilibrium value used in evaluations.*

For one material, SLC A, two measurements were performed with specimens casted at different occasions, but with material from the same batch. The mean relative difference in moisture content between the two measurements was 5 %.
3 RESULTS AND DISCUSSION

3.1 Sorption isotherms

Results of measurements of SLC A-C are presented in Figs. 5-7. Results are given as moisture content (relative to mass at 10 % RH, here called dry weight) as a function of RH. SLC B has a higher moisture sorption than SLC A. This may be an effect of the lower water to binder ratio of SLC B (see subsection 3.2), but also due to the larger amount of paste in SLC B. The sorption isotherm of SLC C for industrial floors has a different appearance than the sorption isotherms for SLC A and B. This might be an effect of the slag content and the lower water to binder ratio.

![Figure 5 – Sorption isotherm of flooring compound A. Stars are measured values.](image-url)
Figure 6 – Sorption isotherm of flooring compound B. Stars are measured values.

Figure 7 – Sorption isotherm of flooring compound C. Stars are measured values.
3.2 Influence of water to binder ratio

The influence of water to binder ratio was investigated by casting specimens of product A with 20 % less and 10 % more mixing water than recommended. The result indicates that a higher water to binder ratio results in lower sorption, although no difference was seen for normal and +10 % mixing water in absorption. A decrease in water to binder ratio results in lower pore volume and a different pore size distribution with a larger amount of smaller pores. A smaller pore has a relatively larger surface area and will be water filled at lower RH than larger pores. As a result specimens casted with lower water to binder ratio will contain more moisture at the same RH in the hygroscopic range.

![Sorption isotherms of SLC A casted with normal amount of mixing water (stars), 10 % more mixing water than recommended (x-marks) and 20 % less mixing water than recommended (triangles). Measurement values below 40 % RH showed only small differences.](image)

3.3 Influence of temperature

The influence of temperature on moisture sorption was studied by measurements at 10, 20 and 40 ºC. As can be seen in Fig. 9, the moisture sorption capacity is lower at higher temperature, that is, an increase in temperature results in an increase in RH at constant moisture content. This behaviour is in qualitative agreement with the Clausius-Capeyron equation [20].
3.4 Influence of age

The structure of the material gradually develops with time until hydration has stopped. As the measurements of sorption isotherms in the sorption balance are relatively fast, it is a suitable instrument for determining sorption isotherms on not completely hydrated materials. Sorption isotherms for SLC A presented in Fig. 10 were determined at 1, 3 and 12 months of age. A difference due to hydration can be seen, but the major part of the sorption isotherm was already developed after one month. The RH in a young material will thus decrease, even if moisture cannot leave the material. Note that specimens in this investigation hydrated at 100 % RH.
Figure 10 – Sorption isotherm of SLC A at 1 month (stars), 3 months (triangles) and 12 months (x-marks) of age. Measurement values below 40 % RH showed only small differences.

3.5 Scanning curves

Knowledge of scanning curves makes it possible to do more accurate predictions of RH in materials changing sorption modes. As seen in Figs. 11 and 12 a small change in moisture content results in a large change in RH. The consequences of this for chemical reactions and biological growth is however somewhat unclear, as discussed in section 1.

The following scanning curves for SLC A, are presented in Figs. 11 and Fig. 12.

- absorption 70-90 % RH after desorption (Fig. 11)
- desorption 90-70 % RH after absorption (Fig. 11)
- absorption 60-95 % RH after desorption (Fig. 12)
- desorption 95-70 % RH after absorption (Fig. 12)

The results show, e.g., that a sample that has dried to 70 % RH and then absorbs moisture corresponding to a moisture content increase of 0.1 %, will have a final RH of about 80 %. If a calculation had been made with only one curve of the isotherm the final RH would have been about 73 % RH, i.e., only slightly higher than 70 %. The scanning effect will therefore act as a lever, giving large increases in RH for small increases in moisture content.
Figure 11 – Scanning curve for SLC A, absorption from desorption mode (70 to 90 % RH) and desorption from absorption mode (90 to 70 % RH).

Figure 12 – Scanning curve for SLC A (three months old), absorption from desorption mode (60 to 95 % RH) and desorption from absorption mode (95 to 70 % RH)
3.6 Carbonation

Carbonation is the process where carbon dioxide from air dissolves in the pore solution and reacts with components in the material. Carbonation of ettringite [21] and calcium aluminate cement based materials [22] leads to a slightly more open material, while carbonation of Portland cement based materials results in a denser material [23]. Measurements on carbonated and non-carbonated specimens of SLC A are presented in Fig. 13. Only minor differences were seen.

![Figure 13](image)

*Figure 13 – Measurements performed on carbonated (triangles) and non-carbonated (stars) specimens of SLC A.*

4 CONCLUSIONS

This study shows that self-levelling flooring compounds have a pronounced sorption hysteresis. Measured scanning curves indicate that when changing sorption mode, for example when adding water to a drying material, a small change in moisture content may result in a large change in RH. Normally RH is believed to be the index best describing critical levels for moisture related processes, such as chemical degradation and biological growth. However, for materials showing large hysteresis it may be necessary to also consider the influence of moisture content.
REFERENCES
