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# STUDIES OF MOISTURE AND ALKALINITY IN SELF-LEVELLING FLOORING COMPOUNDS

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## Abstract

In recent decades there has been an increasing interest in the indoor environment and its connections with public health. One important topic discussed has been the relation between the moisture in buildings and health.

Cementitious materials, when cast, contain excess water since this is necessary for workability and for flow properties. The pore solutions in cementitious materials, besides being high in water content, often have a high pH. High-moisture conditions can cause biological growth. A combination of high moisture conditions and high pH can also result in chemical degradation of other materials, affecting both indoor environment and the technical functioning and appearance of the materials involved.

Self-levelling flooring compounds (SLCs) that provide smooth horizontal surfaces are used to level the substrates (mainly concrete slabs) before floor coverings are applied. Although such compounds are used extensively, only limited research on their moisture and alkali properties and their functioning in floor constructions has thus far been conducted. The aim of the present study was to investigate the moisture and alkali properties of SLCs and gain a better understanding of their interaction with other materials in floor constructions.

Many traditional methods of determining the moisture properties of building materials are time-consuming and are unsuitable for SLCs. Within the project, new methods, such as a rapid method for the simultaneous determination of both the moisture sorption isotherms and of diffusion coefficients, were developed.

Measurements were performed of such moisture properties as the chemical binding of water, the physical binding of it (the moisture sorption isotherm), moisture transport (diffusion), and the rate of surface evaporation and of moisture transport to a concrete substrate, with the aim of better understanding the drying behaviour involved and enabling predictions of the long-term moisture state of SLCs to be obtained. The results served as the basis for a drying model that was developed into a computer-based simulation programme for predicting drying times and long-term moisture states of SLCs.

Only very low rates for the transport of hydroxide ions from the concrete substrates to the SLCs and within SLCs were found to occur. This implies that SLCs can be used as barriers for protecting floor coverings and floor adhesives from the high pH of concrete.

The alkaline degradation of floor adhesives is dependent upon the pH (hydroxide ion concentration) in the zone of contact of the adhesives with the substrate. Secondary emissions from a floor construction due to alkaline degradation are thus dependent

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upon the pH-level and the transport of hydroxide ions in the substrate. For highly alkaline substrates, such as concrete, carbonation is essential for avoiding degradation of sensitive materials. This is not the case for SLCs that have considerably lower pH. The carbonated layer at the concrete surface may, however, due to its limited thickness, be unable to serve as a long-term protection in terms of secondary emissions. This is better provided by an SLC of lower alkalinity.

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## Sammanfattning

Under de senaste decennierna har inomhusmiljön och dess relation till hälsa väckt ett allt större intresse. En viktig aspekt i detta sammanhang har varit kopplingen mellan fukt i byggnader och hälsa.

Cementbaserade material innehåller ofta överskottsvatten ("byggfukt") när de gjuts då detta är nödvändigt för deras arbetbarhet och flytegenskaper. Förutom det höga vatteninnehållet är porlösningarna i dessa material oftast högalkaliska, d.v.s. de har höga pH. Höga fuktillstånd gynnar biologisk tillväxt och höga fuktillstånd i kombination med höga pH kan leda till kemiska nedbrytningsprocesser i andra material. Båda dessa fenomen kan påverka såväl inomhusmiljön som den tekniska funktionen och utseendet av ett material.

Avjämningsmassor används för att skapa horisontella och jämna ytor i golvkonstruktioner innan ytbeläggningen påförs. Trots att avjämningsmassor används i stor omfattning finns det knappt några publicerade resultat som beskriver deras fuktegenskaper och alkaliska egenskaper. Denna studie syftar till att bestämma sådana egenskaper samt att öka förståelsen för hur avjämningsmassor fungerar i samverkan med andra material i golvkonstruktioner.

Många av de idag förekommande metoderna för bestämning av fuktegenskaper hos byggnadsmaterial är tidsödande och ej avsedda för avjämningsmassor. Nya metoder har därför tagits fram inom detta projekt, bl.a. en snabb mätmetod i vilken både sorptionsisotermen och diffusionskoefficienten bestäms.

Mätningar av kemisk bindning av vatten och fysikalisk bindning av vatten (sorptionsisotermen) samt mätningar av diffusionskoefficienter, ytavdunstninghastigheter och fuktflöden till en underliggande betong har utförts för att kunna studera uttorkningsförloppet hos avjämningsmassor samt för att kunna bedöma deras fuktnivåer i ett längre tidsperspektiv. Resultaten från dessa mätningar har utgjort grunden till en uttorkningsmodell vilken sedan vidareutvecklades till ett datorbaserat simuleringsprogram som bedömer uttorkningstider och fuktnivåer på längre sikt i avjämningsmassor.

Mätningar visar att transport av hydroxidjoner mellan betong och avjämningsmassor och i avjämningsmassor sker mycket långsamt. Detta innebär att avjämningsmassor kan skydda golvbeläggningar och golvlim från en betongs höga alkalinitet.

Alkalisk nedbrytning av golvlim beror av pH (hydroxidjonkoncentrationen). Sekundära emissioner från golvkonstruktioner som uppkommer genom alkalisk hydrolys är därför direkt beroende av underlagsytans pH och transportegenskaper för hydroxidjoner. För

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högalkaliska underlagsytor som betong är därför karbonatisering nödvändig för att undvika nedbrytning av känsliga golvmaterial. Detta gäller inte för avjämningsmassor som har en betydligt lägre pH. Karbonatisering ger vanligen endast ett tunt skikt av lägre pH i betongens yta. Det är därför osäkert huruvida detta skikt kan utgöra ett långsiktigt skydd för känsliga golvmaterial mot betongens i övrigt högalkaliska fukt. En avjämningsmassa får därför anses vara ett bättre alternativ i detta fall.

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## Key words

self-levelling flooring compound, floor screed, SLC, moisture transport, sorption isotherm, diffusion coefficient, alkalinity, secondary emissions



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## The papers included

**Paper I Moisture properties of self-levelling flooring compounds. Part I. Diffusion coefficients.** A. Anderberg and L. Wadsö, *Nordic Concrete Research*, Publication 32, 2/2004, pp. 3-15.

**Paper II Moisture properties of self-levelling flooring compounds. Part II. Sorption isotherms.** A. Anderberg and L. Wadsö, *Nordic Concrete Research*, Publication 32, 2/2004, pp. 16-30.

**Paper III Method for simultaneous determination of the sorption isotherm and the diffusivity of cement-based materials.** A. Anderberg and L. Wadsö, (*Submitted*)

**Paper IV Drying and hydration of cement-based self-levelling flooring compounds** A. Anderberg and L. Wadsö, (*Submitted*)

**Paper V Hydroxide ion concentration in an interface between a concrete and a screed with different alkalinities** A. Anderberg and L. Wadsö, (*Submitted*)

**Paper VI Secondary emissions from a floor adhesive as a function of pH and RH.** A. Anderberg and L. Wadsö, (*Draft*)

**Paper VII Influence of additives on physical properties of self-levelling flooring compounds.** A. Anderberg and L. Wadsö, (*Submitted*)

**Paper VIII Influence of storing temperature on ageing of raw materials** A. Anderberg and L. Wadsö, (*Accepted for publication in Proceedings of the 12th Congress on the Chemistry of Cement, Montreal, 2007.*)

# Preface

The work on which this doctoral thesis is based was carried out at the Division of Building Materials, Lund Institute of Technology. The project is part of the industrial research school The Building and Its Indoor Environment. The financial support that Maxit Group AB and The Knowledge Foundation (KK-stiftelsen) provided is gratefully acknowledged.

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Lund, March 2007  
*Anders Anderberg*

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*“To explain all nature is too difficult a task for any one man or even for any one age. Tis much better to do a little with certainty & leave the rest for others that come after you“.*

Isaac Newton

Cited in [http://en.wikiquote.org/wiki/Isaac\\_Newton](http://en.wikiquote.org/wiki/Isaac_Newton).

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# Chapter 1

## Introduction

The relationship between the indoor environment and the health of the inhabitants has been studied for centuries. One of the issues discussed throughout the years has been the relationship between dampness in buildings and the health of those living or working there. For example did an health inspector in Stockholm in 1913 write about health consequences such as headaches and mucous membrane irritation caused by dampness in buildings and his worries about the fact that the building hygiene issues had received less attention during the preceding decades [1]. Although the dwellings of today have little in common with those habited a century ago, the connection found then between dampness and health is still valid. During recent decades, the attention directed at emissions of volatile compounds released from various materials has increased, these being suspected of reducing indoor air quality and of producing deleterious health effects.

Since the walls, ceilings and floors of buildings posses a large surface area, they can have a strong effect on the indoor environment. The most complex of the surface constructions involved is that of the floor, which has to resist abrasion from for example foot traffic through the building and from furniture, as well as to withstand wet cleaning. A large part of the floor constructions of today are levelled by self-levelling flooring compounds (SLCs) that create smooth horizontal surfaces.

The present study aims at describing the moisture and alkali properties of SLCs. A further aim is to increase the understanding of the functioning of this type of material in a floor construction, how SLCs can interact with other materials and what consequences this can have for the indoor environment.

The study is divided into five main parts. The first part concerns the indoor environment and its relation to health. This was studied through a literature survey, conducted in order to gain a better understanding of the relation between the indoor environment and public health, thus placing the project in a broader context. The

second part concerns SLCs as materials, their constituents and the properties of the final product. The third and major part of the study concerns the moisture in materials and measurements of the moisture properties of SLCs. In this part, all the important moisture properties of SLCs are quantified and a drying model is presented, one which form the basis for a computer-based programme enabling the drying and long-term moisture states of SLCs to be simulated. The fourth part deals with the alkalinity of cementitious materials, how concrete and SLCs differs in this respect, and the transport of hydroxide ions in a structure consisting of a combination of concrete and SLC. The fifth and last part finally, deals with emissions, in particular secondary emissions from floor adhesives and floor coverings that are degraded in contact with cementitious materials. These five parts are taken up in chapters 2-6.

Eight papers are included in the thesis. They are summarized and discussed in chapters 3-6, the summary of each paper being presented in the context in which it is most relevant.

# Chapter 2

## Buildings and health

### 2.1 Background

During the 19th century increasing attention was directed at the housing conditions of the poor people in Sweden. Connections between housing conditions and mortality were observed [2]. Many people at the time lived in overcrowded, dirty and insanitary dwellings that left no space for other indoor activities than sleeping and eating. Conditions throughout much of Europe generally were similar to this [3].

Political and technical developments led to the construction of better buildings. At the end of the 1960s, a decade in which a million dwellings were constructed, housing conditions in Sweden improved substantially. At present, a few decades later, a new situation is being encountered, housing conditions being so much improved and life having changed so much for what would superficially seem the better that people tend to spend most of their time indoors. Today we spend much of our time at home or in other indoor environments such as offices, schools, day-care centres and vehicles. Only a fraction of our time is spent outdoors. A new discussion of the sedentary nature of our indoor activities in modern buildings and the health-related effects of this has replaced earlier discussions of insanitary conditions in buildings.

Our buildings have primarily been designed to provide a climate shield against precipitation, wind and coldness. During the 20th century this function was developed increasingly, partly because of the demands of the users themselves and of authorities and partly through new building materials and construction solutions having been developed and adopted. A specific indoor environment, almost completely separated from the outdoor environment, has been created. Since we spend most of our time indoors, it is primarily the indoor environment to which the human body is exposed. In some buildings, new types of indoor-related health problems have appeared. Although



these may have long been present, in fact, such indoor-related health problems were first recognised around 1965 in the US [4], during the 1970s in Germany [5] and in the late 1970s in Sweden [6]. The increased costs of heating have led to tighter buildings being constructed with lower ventilation rates and greater amounts of recirculated air. This, together with the use of new building materials and construction solutions, are suspected of having contributed to deterioration of the indoor environment and to an increase in the health complaints resulting.

## 2.2 Sick-Building Syndrome, SBS

The health effects related to the indoor environment can be divided into a number of different categories, see [7], for example. The most widely known of these is the Sick-Building Syndrome, SBS, which includes irritation of the eyes, nose and throat, headaches and fatigue, skin disorders, and unpleasant odour and taste perceptions. These are symptoms that cannot be linked with some single pollution source. They tend to increase with the time spent in the building and to decrease when leaving the building. SBS is a somewhat diffuse term since it is not the buildings in question that are sick but the individuals living or working in them. It should be pointed out that a fraction of the individuals living or working in a building normally report health problems at any given time. According to Jones [8], as many as 20% of those working in a "healthy" office building may report symptoms of the SBS type. Significantly higher levels of complaints need to be reported before a building can be classified as being "sick".

Another type of disorder, one that should not be confused with SBS, is that of Building Related Illness, BRI. Such illness has a known etiology and specific symptoms that can be connected directly with a particular factor present in the building, Legionnaire's disease being an example of an illness of this kind [7]. However, the distinction between BRI and SBS is not always clear [9].

## 2.3 Indoor environment quality

Various attempts have been made to explain the negative health effects evident in indoor environments. Although extensive research has been carried out, with few exceptions, such as in the case of environmental tobacco smoke, house-dust mites and radon [10], it is not known what agents in the indoor environment that cause the negative health effects. Several large studies have been performed in efforts to relate agents found in the indoor environment to health. However, since the sampling and analysis technique presently available fail to register all the potential agents the indoor air contains, certain important agents may be missing in the studies conducted.

Jones [8] divides factors contributing to problems in the quality of indoor air into four categories: chemical, physical, biological and psychological. These categories are discussed below.

### **Chemical factors**

Chemical factors are mainly those of the emission of molecules from such sources as furnishings, personal care products, building materials, cleaning products and house hold or office equipment. Some of these emissions smell, the odours perceived influencing our sense of well-being. The health effects of some emissions, such as those of benzene, toluene and formaldehyde, are known, but only for high levels of exposure, not for the typical levels present in the indoor environments of today. Questions have been raised of whether all the relevant pollutants have been measured [11]. Some potential irritants such as radicals from gas-phase reactions, are not easily detected by the measuring techniques presently available. Ozone, a strong oxidant that can react with various compounds in the indoor air to form irritants, is one possible source of such irritants [12]. Emissions from building materials are discussed further in chapter 6.

### **Physical factors**

Physical factors include those of temperature, light, noise and humidity. Humidity is an often discussed subject, not because of its direct impact on human health, but because of its indirect influence, as it promotes degradation processes and microbial growth. Dampness and humidity have been found to increase the risk of negative health effects on airways, and to produce headache and fatigue [13]. This appears to be true, irrespective of whether the dampness is measured as condensation on windowpanes, as water damage or as smell or odour. However, except for house-dust mites, it is not known what connections there are between dampness and negative health effects. Possibilities that have been discussed include emissions from degraded building materials [14], emissions from microbial growth [15] and viruses [16]. Both chemical degradation processes and microbial growth increase with increasing moisture content since this leads to an increase in the molecular mobility of the chemical reactants and the microbial nutrients involved [17].

### **Biological factors**

Biological factors include mould and bacteria, and also emissions from humans, pets and indoor plants. The key factor for microbial growth indoors is that of humidity. Other factors of importance, such as temperature, nutrients and oxygen, are nearly always at sufficient levels. Potential agents of biological origin that are suspected of contributing to

negative health effects are those of proteins, mycotoxins and MVOCs, for example [18]. MVOCs are volatile emissions of alcohols, ketones, esters, etc., released during microbial growth, whereas mycotoxins are toxic metabolites produced during the growth of mould. Mycotoxins can be deposited in the airways when spores are inhaled [19]. Although it is not known which agents are responsible for the negative health effects here [20], it has been found that repeated exposure to high levels of biological agents represents a risk factor for the development of specific allergic reactions [19]. Once the immune system has been triggered, the allergic reaction can be started by exposure to low levels of allergens (substances that induces allergy).

## Psychological factors

In the literature, psychological factors are linked mainly with office environments. Factors such as labour relations and office culture are mentioned, and also stress in general [21], which can influence individuals in environments of all types.

## 2.4 Indoor air quality, IAQ

Indoor air quality (IAQ) is influenced by chemical, physical and biological factors, the perceived IAQ being influenced by psychological factors as well. A general way of improving the IAQ is to exchange the polluted air for "fresh" outdoor air through ventilation. Several studies in non-industrial environments have shown an increase in the supply of outdoor air to decrease the risk of health symptoms, and to increase both satisfaction with the IAQ [22, 23] and productivity in office work [24]. Another approach to improving the IAQ is to remove the pollution source, so called source control. Two Scandinavian studies in this area [25, 26] concerned the effects of introducing a pollution source, an old carpet from a building with an SBS history. Both studies found an increase in SBS symptoms and a negative impact on perceived air quality and productivity in office work when such a pollution source was introduced. These studies all indicate that source control or removing pollution influence both productivity and the perceived IAQ.

Although there is only limited scientific evidence for the impact on health of organic compounds present in indoor air, these are suspected of contributing to poor indoor environments [27, 28]. It has been shown that secondary emissions (emissions formed by reactions within the materials involved) are important for indoor air quality [29]. Secondary emissions from floor constructions with bonded floor coverings atop concrete are suspected of causing health problems [14]. Such emissions can occur when floor adhesives and floor coverings are degraded, forming volatile products that are emitted to the indoor air. The degradation (alkaline hydrolysis) occurs under the

influence of moisture and high pH. Concrete substrates with bonded PVC and linoleum flooring are floor constructions commonly subjected to degradation. The degradation products include 2-ethyl-1-hexanol and butanol [30, 31, 32]. Both are strongly odorous but neither of them has been proved conclusively to have any negative impact on health, although correlations between increased levels of 2-ethyl-1-hexanol and adverse health effects have been found in a number of studies [33, 34, 35].

According to the World Health Organization (WHO), "health is a state of complete physical, mental and social well-being and not merely the absence of disease or infirmity" [36]. The WHO-document "The right to healthy air" also states in the section "Principles" under the heading "Principles" that "Under the precautionary principle, where there is a risk of harmful indoor air exposure the presence of uncertainty shall not be used as a reason for postponing cost-effective measures to prevent such exposure" [37], meaning that possible sources should be limited or removed, even if only suspected of being harmful.



# Chapter 3

## Self-levelling flooring compounds

### 3.1 History

The first pumpable self-levelling flooring compound (SLC) was developed by Nils Johansson, Oy Partek Ab, Finland, and was launched on the market in 1977. Portland cement was used as a binder and casein-based flowing agents gave the product its excellent flow properties. The aim of the product was to create an easy and fast way of levelling off concrete floors before applying a floor covering. Instead of having to post-treat the concrete surface, which often is hard work, it became possible to produce smooth horizontal surfaces with use of a pumpable cement-based mortar. A capacity of up to 400 m<sup>2</sup>/h [38] was achieved. The product rapidly gained popularity on the Nordic market. At the beginning of the 1980s, the product was used on about 90% of the concrete floors produced in Sweden [39].

However, after some time complaints began to appear. Oak parquets and cork floorings sometimes became discoloured. Also people living or working in buildings in which there were concrete slabs sometimes claimed that they did not feel well while in these buildings. The symptoms were found to be ones related to SBS, see section 2.2. Several large-scale investigations were undertaken in Sweden and complaints of this sort were found in a small percent of the buildings [39]. Discolouration of the oak parquets and of the cork floorings were the most frequent complaints, ammonia from the casein flowing agent being found to be the cause of this discolouration [40]. Casein was also suspected of contributing to the negative health symptoms related to SBS that were reported in these buildings, although this was never proven to be the case [6]. The suspicion was based on the fact that when casein is degraded, odourous volatile products like ammonia and amines are formed. Some other suspected reasons for the complaints were those of inadequate ventilation and of emissions resulting from the degradation of the plasticisers contained in vinyl floorings [40]. Moisture was found to play a central

### A generic SLC recipe

Paper VII presents a generic recipe that was formulated for enabling studies to be carried out of the properties of SLCs as a function of different additives and different concentrations of additives. Measurements of the influence of three different polymers and of one thickener on the moisture properties, density, strength and dynamic modulus of elasticity of SLCs were made using the recipe. It was found to be possible to study the influence additives had on most properties by use of the recipe. No tendencies toward separation in the mortars were observed during the experiments.

role in the degradation, both of casein and of plasticisers. High moisture levels were found in various of the concrete slabs [39].

Casein is a protein prepared by the precipitation of milk, by rennet, for example. Similar proteins can also be obtained from other animal or plant species [39]. Casein products had been used in building materials for decades, but were found here to degrade under the moist alkaline conditions often present in concrete slabs [14]. Some of the SLCs that contained casein could emit ammonia at an RH of less than 85% [41].

Since casein was suspected of contributing to a poor indoor environment, Nordic producers of SLCs developed new formulations that were without casein. In these new formulations, casein was replaced by synthetic flowing agents, and the binders were also changed. The new binder systems were mixtures of different types of cements and calcium sulphates. These binder systems are of lower pH, making the final product less aggressive toward other materials.

## 3.2 SLCs today

Today SLCs are still widely used in Sweden, a rough estimate being that about 70% of all floors produced in Sweden are levelled by use of SLCs. In central Europe and in southern Europe in particular this figure is considerably lower.

Alongside SLCs using synthetic flowing agents, SLCs using casein as flowing agent are still manufactured outside the Nordic countries. There are also SLCs based on use of calcium sulphates as binders. However, the SLCs investigated in the present study are mainly cement-based and make use of synthetic flowing agents.

Table 3.1: Contents of a typical SLC

Component	Approximate quantity / %	Main function
Calcium aluminate cement	13	Binder
Portland cement	2	Binder
Calcium sulphate	5	Binder
Limestone filler	30	Aggregate
Quartz sand	50	Aggregate
Redispersible polymer		Improvement of flow, abrasion resistance, flexural strength and bond strength
Thickener		Prevents bleeding and segregation
Flowing agent		Improves self-levelling properties and reduces water demand
Retarder		Increases open time
Accelerator		Increases rapidness of early strength development
Defoamer		Reduces air entrainment during mixing and application
Water	100 mass-% of binder	

SLCs consist of binders, aggregates and additives. A typical composition including the main additives is shown in Table 3.1. The binders are normally a mixture of calcium aluminate cement (CAC), Portland cement (PC) and calcium sulphates (CS) in the form of anhydrite and hemihydrate. This binder system makes it possible to formulate rapid-hardening, rapid-drying and shrinkage-compensating SLCs having relatively high water-to-binder ratios. The main reaction product when mixtures of CAC and CS are employed is ettringite,  $[\text{Ca}_3\text{Al}(\text{OH})_6]_2(\text{SO}_4)_3 \cdot 26\text{H}_2\text{O}$ . Ettringite consists to nearly 50 mass-% of water and is formed very quickly if sufficient quantities of sulphates are available [42]. The production of ettringite can provide shrinkage compensation since the formation ettringite can be accompanied by expansion [43]. Ettringite formed in the hardened state in concretes based on Portland cement can create problems of durability due to expansion and cracking brought on by sulphate attack or by elevated temperatures during hydration [44]. If all the CS reacts in the course of the initial reactions (as they do in modern SLCs), no such harmful expansion occurs [42]. Some of the hydrate water in ettringite is lost, to some extent irreversibly, when drying occurs at temperatures above some 60 °C [45] or when it occurs over desiccants [46]. The harsher



### Low temperature storage of inorganic binders

Paper VIII presents measurements of the influence of storage time and storage temperature on the rate of reaction of inorganic binders and SLCs and on the flow of SLCs. Three types of binders, - a Portland cement, an aluminate cement and a calcium sulphate - and three SLCs were studied. The materials were stored for four years at two different temperatures, room temperature and in a freezer (-25 °C). The effects of time on the properties of the materials were found to be less at the lower temperature. Storing dry cement-based materials at low temperature is thus desirable when measurements are performed over a long period of time.

the drying is, the more hydrate water is lost. For this reason, SLCs are dried with use of "gentle" methods when the moisture content is being determined, see section 4.4.

An important additive is the redispersible polymer powder that improves the flow, the surface abrasion resistance, the flexural strength and the bond strength. The polymer particles coalesce into films or membranes in the final product, which bind the cement hydrates together into a network [47]. Sand and finely ground mineral materials such as limestone serve as aggregates. The other additives control such matters as setting time, curing time, flow characteristics, air entrainment and separation. A normal SLC contains about 15 different components, making it a very complex product.

Whereas the first SLCs were meant to be laid in thin layers, layers up to 100 mm are used today. Such thick layers are generally divided into two parts, a fine smoothing SLC being laid on top of a "base" SLC. SLCs are meant to be applied indoors. Because of their low viscosity, one needs to be careful in connection with joints and the like in the substrate to avoid flow of the fresh product into other parts of the construction.

The widespread use and the lower pH (as compared to concrete) makes the product interesting, not only for increasing production efficiency and the working environment, but also in terms of IAQ. The lower emission levels in floor constructions when PVC-floorings are bonded on SLCs as compared with those bonded on concrete, [30, 31, 48], leads to a better IAQ at a given level of humidity in the floor construction. It would also be possible to obtain similar emission levels as for the case of concrete, but at higher moisture levels, by the use of SLCs in floor constructions. Whereas the former would provide a higher safety margin in the avoidance of undesired levels of VOCs in the indoor air, the latter would provide possibilities of reducing the construction time through a reduction in drying time.

Although modern SLCs are quite different from the original ones, the by far most important factors regarding their possible influence on the indoor environment are still those of moisture and of alkalinity.



# Chapter 4

## Moisture

### 4.1 Introduction

Moisture is not only related to indoor environmental issues, but is also of interest in connection with other processes occurring in materials, such as the corrosion of steel, shrinkage and swelling generally, frost damage and the hydration of inorganic binders.

Moisture content is the mass of water in a material divided by the mass of the dry material. This is a convenient measure of the moisture state since it is easy to measure. The moisture content is calculated from the measured mass of a sample, before and after drying it.

The availability of moisture in materials is expressed as the water activity ( $a_w$ ), defined as the ratio of the fugacity of the water in the specimen to the fugacity of pure water, where fugacity is the tendency for water to escape [49]. At atmospheric pressures, the ratio of the vapour pressure of the water above a sample to the vapour pressure of pure water can be used as a measure of water activity at an inaccuracy of less than 1% [17]. The relative humidity (RH), which concerns water in the vapour phase, is the water vapour pressure divided by the water vapour pressure at saturation at the temperature of interest. A ratio of vapour contents can also be used since at a constant temperature the vapour content is proportional to the vapour pressure. The RH is often expressed as a percentage. Whereas RH is a measurable quantity,  $a_w$  is not, but can be deduced from RH measurements under equilibrium conditions, as described above. The terms  $a_w$  and RH are used interchangeably in some areas of science, such as in building materials science, where the moisture state of materials often is expressed as RH.

Water activity is a measure of the availability of water. The rates of processes involving water should be dependent, therefore, on the water activity. As an example,

an  $a_w$  of 0.8 is required for most building materials in order for the growth of mould to occur, although some species of mould are able to grow at a lower  $a_w$  [18, 50]. However, chemical reactions are also dependent on transport properties, since reactants have to come in contact with each other, so the moisture content may also be of relevance for chemical processes. Two other factors influencing the biological and chemical processes that occur are those of temperature and of alkalinity.

The moisture in materials is often divided into different types depending upon how hard the water molecules are bound to the surface or to the structure of the material. This is important since not all water inside materials contributes to biological growth, to the transport of substances, to chemical reactions and the like. The water, which is most weakly bound is of greatest interest here [17].

The water in a cementitious material, which is most strongly bound is chemically bound as reaction products in the hardened cement paste, it is thus being part of the solid structure of the material. Since this water is strongly bound in the structure and under normal circumstances does not leave the material, it is of no direct interest in connection with indoor environmental matters. It is nevertheless of interest, however, since chemical binding consumes mixing water (self-desiccation) and therefore contributes to the drying process. The water which is less strongly bound is discussed in the sections that follows.

Note that there are often no natural borders between the different categories of water in a material. For example, there is no clear distinction between the chemically bound and the physically bound water that cementitious materials contain. Different measurement methods used to measure this water often give different results. Accordingly, in stating the result obtained for a measurement of physically or of chemically bound water, one should always state the method employed. Another example of this is the distinction between hygroscopic water (water that a material can absorb from the air) and superhygroscopic water. Here, values in the range of 95% to 98% RH are often presented in the literature.

## 4.2 Moisture sorption

The amount of water in a material, in addition to chemically bound water, is normally given as the sorption isotherm, i.e., the relation between the physically bound water (moisture content) and the water activity, at constant temperature and constant total pressure. Physically bound water can either be adsorbed to the surfaces of the material, absorbed into the structure of the material (at binding sites) or condensed in capillaries on water menisci formed in the pores. The first layer of adsorbed water is the most

### Measurements of sorption isotherms

Paper II presents measurements of the moisture sorption of three commercial SLCs. The materials were cured at 20 °C and 100% RH prior to the measurements being made. Only small differences were found between sorption isotherms measured after 1, 3 and 12 months, the sorption capacity increasing over time only slightly after the first month. A temperature dependence of about 0.2% RH per °C in the RH range of 50 to 95% could be observed in measurements performed at 10, 20 and 40 °C. A clear effect of the water-binder ratio (w/b) ratio was noted, a higher w/b resulting in a lower sorption capacity, whereas carbonation had only a minor effect. Various scanning curves were also obtained. These showed that in changing the sorption mode minor changes in the moisture content could lead to large changes in the RH.

strongly bound and the most immobile, and is unfreezable at -55 °C [51]. This water behaves as though it were part of the solid and it corresponds to the moisture content of the monolayer (a fictive state in which the internal surface of the material is covered by a layer of water molecules) [52]. Further water which is added is more mobile. When sufficiently abundant, this water allows microbial growth, ion transport and chemical reactions to occur in solution.

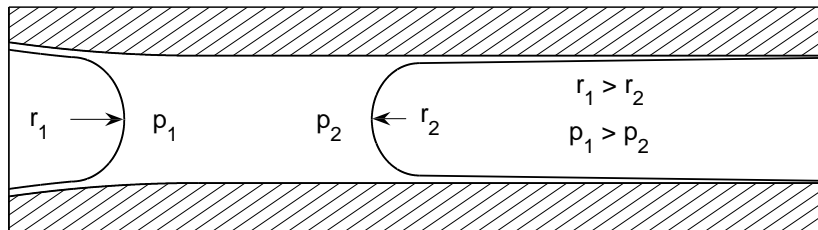


Figure 4.1: Water menisci in a pore. The meniscus having the smaller radius corresponds to a lower RH. The water tends to move towards the lower RH since the pressure is lower there due to the smaller radii.

In fine-porous materials such as hardened cement paste, water is bound not only by adsorption but also by capillary condensation in the pores. Water molecules then condense on concave water menisci, Figure 4.1. The curvature of the meniscus corresponds to a particular equilibrium pressure of the water. The smaller the radius, the lower the equilibrium pressure is. This relation can be expressed by means of the

Kelvin equation [52]:

$$RT \ln\left(\frac{P}{P_0}\right) = -\frac{2\gamma V}{r_m} \quad (4.1)$$

where  $R$  is the gas constant ( $8.314 \text{ JK}^{-1}\text{mol}^{-1}$ ),  $T$  is the temperature (K),  $P$  is the vapour pressure over the meniscus ( $\text{Nm}^{-2}$ ),  $P_0$  is the vapour pressure of pure liquid ( $\text{Nm}^{-2}$ ),  $\gamma$  is the surface tension ( $\text{Nm}^{-1}$ ),  $V$  is the molar volume ( $\text{m}^3\text{mole}^{-1}$ ) and  $r_m$  is the mean radius of curvature of the meniscus (m). When a material is in equilibrium at low RH, only small water menisci can exist, i.e., only narrow pores can be filled with water. At higher RH levels, larger water menisci can form and pores with a larger pore radius can be filled with water, see Table 4.1. In a fine-porous material, condensed water in capillaries is probably the dominant contributor to microbial growth, transport processes and chemical reactions.

Table 4.1: Thickness of the adsorbate, radii of the water menisci and radii of the pores that hold capillary water in cylindrical pores in both absorption and desorption. The adsorbate was calculated using the Halsey equation, equation 9 in [53], assuming the constant  $r=3$ . The Halsey equation was solved by using the monolayer content calculated using the BET-equation, equation 28 in [54], assuming the constant  $c=20$ . This gave the monolayer at RH=18%. The radii of the menisci were calculated using equation 4.1. It was assumed that menisci with radii smaller than 1.4 nm could not exist. The thickness of a layer of water molecules is 0.29 nm.

RH / %	Adsorbate / nm	Radius meniscus desorption / nm	Radius meniscus absorption / nm	Adsorbate + radius desorption / nm	Adsorbate + radius absorption / nm
20	0.30	-	-	-	-
40	0.36	-	-	-	-
60	0.43	2.1	-	2.5	-
80	0.57	4.8	2.4	5.4	3.0
90	0.73	10.2	5.1	11	5.9
95	0.93	21.0	10.5	22	11
99	1.6	107.3	53.6	109	55

Most materials show sorption hysteresis, that is, they will contain differing amounts of water in equilibrium with a particular RH, depending on whether the material is drying or is taking up moisture. This can be seen in Figure 4.2, where the sorption isotherm has one main loop for desorption and another main loop for absorption. If a drying material is rewetted, it follows a scanning curve from the desorption isotherm towards the absorption isotherm. This means is that a small increase in moisture

content can lead to a large increase in water activity (RH). A consequence this has for moisture-related processes is that a single parameter, such as water activity, is unable to completely describe the effect of moisture on the rate of biological and chemical processes [17]. One may need to take into consideration not only the water activity, but also the moisture content as has been noted for microbial growth on foodstuffs [55].

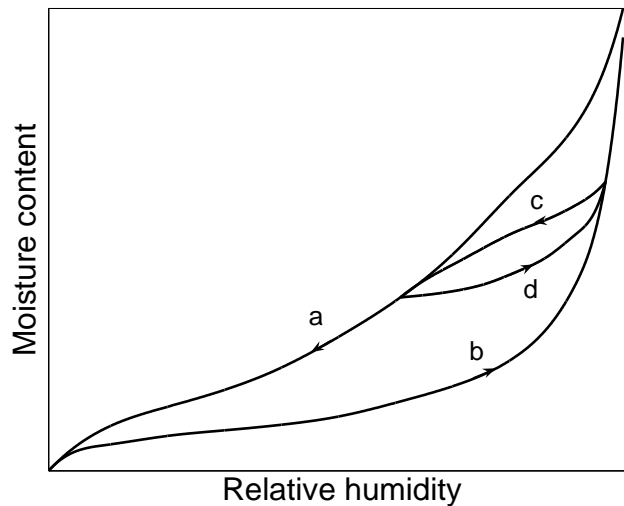


Figure 4.2: An example of a sorption isotherm having two main loops, a (desorption) and b (absorption), and two scanning curves c and d.

Sorption hysteresis is often temperature-dependent and may occur for any of several different reasons [52, 56]. The following are various explanations of hysteresis:

- The ink-bottle theory. In ink-bottle pores, larger spaces are connected by narrow channels. The menisci in the narrow channels retard the desorption of the larger spaces.
- The differing conditions found (for example, different contact angles) when water menisci are forming or are breaking resulting in this occurring at different RH values.
- Irreversible changes in the pore structure during sorption.
- Time-dependent hysteresis caused by relaxation phenomena.

A rather complete model describing sorption hysteresis occurring in Portland-cement-based materials has recently been presented by Espinosa and Franke [57, 58]. The model includes the effects of ink-bottles and the different conditions for the forming and the



### Measurements of water diffusion coefficients

Paper I presents measurements of moisture transport performed by use of the cup method. The measurements were made on three commercial self-levelling flooring compounds (SLC), differing in their properties, a normal SLC and a rapid drying SLC - both for non-industrial applications - and an SLC for industrial floors. The evaluation was based on Fick's first law of diffusion. The effects of the amount of mixing water, the temperature and the initial moisture content were investigated. A lower water-to-binder ratio, a lower temperature and a lower initial moisture content all resulted in lower diffusion coefficients.

breaking of water menisci as well as the effects of the degree of hydration and the effects of drying on the pore structure.

The sorption isotherm, as the name indicates, only applies to a given temperature. An increase in the temperature results in an increase in water activity at a constant moisture content [17, 59] (opposite to air, in which an increase in temperature results in a lower RH). For concrete, a temperature dependence of 0-0.4% RH per °C, dependent on moisture conditions, has been reported [60]. Other substances, for example some foodstuffs, may show a stronger temperature dependence [61]. An increase in temperature normally increases the rate of physical and chemical processes [49].

## 4.3 Moisture transport

Moisture transport occurs in porous media when a driving force is present. The driving force may be either a concentration gradient or an external pressure gradient. If there is an external pressure gradient through a material, moisture, either in the liquid phase (water) or in the vapour phase (convection by air), is forced through the material. Since this type of moisture transport is not relevant in the present study, it will not be discussed here further.

Moisture transport due to a concentration gradient can be divided into two types: vapour transport and capillary transport. Vapour transport occurs in non-water-filled pores due to diffusion, see Figure 4.3. Diffusion is a net flow of molecules due to random motion from a region of higher concentration to a region of lower concentration of molecules. Diffusion in very narrow pores where the collisions between the molecules and the pore walls have a considerable effect on the rate of diffusion is sometimes called Knudsen diffusion [62]. Another type of diffusion that has been discussed is

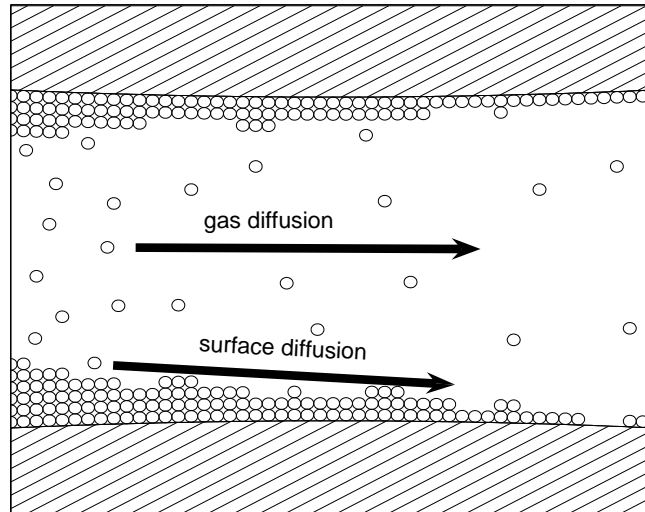


Figure 4.3: A schematic diagram of the diffusion in a pore, where molecules in random motion tend to move towards areas of lower concentration. Note that there is always a rapid exchange of molecules between the gas phase and the condensed phase.

movement of the bound adsorbate, or surface diffusion [63, 64], which can better explain measurements of higher moisture flow in narrow pores than can be expected from gas diffusion. Capillary transport occurs in water-filled pores due to pressure differences in the water menisci, Figure 4.1. Since it normally is not possible to separate the different transport mechanisms, it is usually the total moisture flow which is described. At a steady state, this is given by Fick's law [65]:

$$q_m = -D \frac{dC}{dx} \quad (4.2)$$

where  $q_m$  ( $\text{kg m}^{-2}\text{s}^{-1}$ ) is the flux,  $D$  ( $\text{m}^2\text{s}^{-1}$ ) is the diffusion coefficient, and  $dC$  ( $\text{kg m}^{-3}$ ) is the difference in concentration over the distance  $dx$  (m). Other formulations of Fick's law use vapour pressure, relative humidity or other moisture potentials. Moisture transport is normally dependent on the moisture state and the temperature of the material. A typical relation between the diffusion coefficient and the water activity for a cementitious material is shown in Figure 4.4, where it can be seen that the moisture transport is rather constant at lower levels of water activity, but rapidly increases at high levels of water activity.

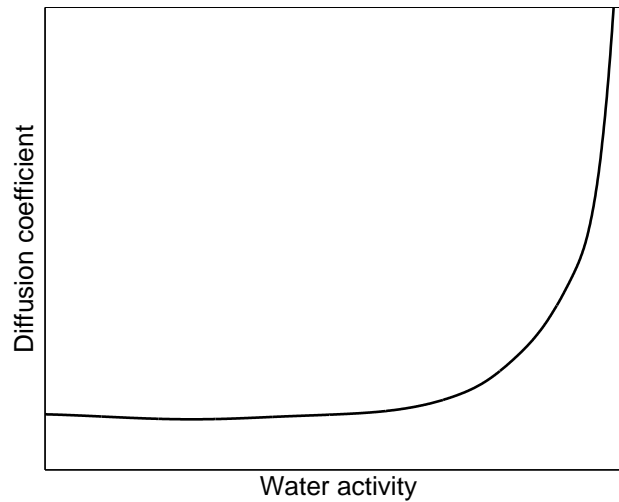


Figure 4.4: Schematic diffusion coefficient for a cementitious material as a function the water activity,  $a_w$ .

## 4.4 Drying of self-levelling flooring compounds

SLCs dry as a result of four different processes: chemical binding of water, physical binding of water, surface evaporation and moisture flow to the substrate. When water reacts with the binder, it is chemically bound in the reaction products and a fine-porous matrix is created in which excess water is physically bound. The process of chemical and physical binding of water is called self-desiccation. Self-desiccation occurs in all materials in which water reacts with a binder and becomes part of the solid, as occurs for example in all cement-based and calcium-sulphate-based materials. However, only for some few products in certain applications, drying by only self-desiccation is sufficient to meet practical requirements regarding moisture levels. In most applications, drying by surface evaporation is needed as well in order for these requirements to be met.

SLCs normally have high water-to-binder ratios (w/b) due to the self-levelling properties that are desired. A low w/b tends to give the fresh mortar too stiff a consistency. A consequence of high w/b may be that part of the water separates from the mortar (bleeding) and forms a water film on the surface. The separated water includes such additives as the redispersible polymer and other fine particles. As the surface water evaporates, the polymers are left in the surface region. Besides affecting the mechanical properties, the higher amounts of polymers in the surface may influence the moisture transport properties by adding an extra resistance to the moisture flow to the surface.

### A method for rapid determination of the sorption isotherm and the diffusion coefficient

Paper III describes a rapid method enabling both the sorption isotherm and the diffusion coefficient to be determined in one measurement. The test specimens are inserted in a sorption balance and exposed to step changes in the RH. The equilibrium value at each measurement step is used to evaluate the sorption isotherm, and the sorption kinetics in the first part of each measurement step is used to evaluate the diffusion coefficient. To enable measurements to be performed rapidly, the last part of each measurement step is extrapolated towards a final equilibrium value. Corrections are made for the measurement steps not reaching equilibrium and for the surface mass transfer resistance. Measurements in 8 steps in both the desorption and the absorption mode take about 10 days to complete for a given specimen.

## Chemical binding of water

Chemical binding of water occurs when water reacts with the binder (hydration), e.g. cements and calcium sulphates in the case of SLCs, and becomes chemically bound in the reaction products. The rate of hydration is a function of temperature, degree of hydration and moisture state. The amount of chemically bound water in cement-based materials is often determined by drying at 105 °C or by other methods, such as drying over desiccants or over saturated salt solutions, to release water that is physically bound, and then heating the material to around 1100 °C so as to release the chemically bound water. In the present study, drying to release physically bound water was carried out at 10% RH and 20 °C. Harsher drying can result in mass loss due to the release, partly irreversible, of hydrate water from ettringite, which is defined as chemically bound water [45, 46]. In practice, oven drying at 40 °C is often used for drying SLCs. Under conditions in which the humidity level in the surrounding climate is not too low (about 15% RH at 20 °C), the values obtained in drying in an oven at 40 °C correlates well with values obtained in drying at 10% RH at 20 °C.

## Physical binding of water

In addition to the chemical binding of water, water is also physically bound to surfaces and to water menisci in the developing pore system. Cement-based materials have large inner surface areas, the pores generally having radii in the nm-range. Thus according to the Kelvin equation, equation 4.1, these are capable of holding capillary water also at a low relative humidity. The amount of water that is physically bound in a material which

### The drying of SLCs

Paper IV describes measurement methods for, and measurements of the four different drying processes that contribute to the drying of SLCs: evaporation, chemical binding, physical binding and absorption into the substrate. These measurements were performed on three commercial SLCs. The importance of the different processes is discussed and a model describing the total drying behaviour is proposed. The model relates all the properties of the SLC to the hydration state as measured by isothermal calorimetry (produced heat).

is in equilibrium with a particular RH and a certain temperature is given by the sorption isotherm, as discussed above. The sorption isotherm changes as the hydration reactions in the material proceed. A higher degree of hydration results in finer pores and generally in a greater sorption capacity.

### Surface evaporation

Surface evaporation starts immediately upon mixing. When cast, bleeding (the transport of water to the surface) occurs, forming a water film on the surface. During this brief stage, the rate of surface evaporation depends mainly on the temperature, the ambient RH and the air speed over the surface. After about one to two hours, the main reaction starts and the material turns from a particle suspension into a solid. The surface then gradually dries and a moisture gradient through the material develops. The rate of surface evaporation depends then to a large extent on the moisture transport from within the material to the surface, which is a function of the moisture transport properties of the material. An example of the rate of surface evaporation during the first day after casting is given in Figure 4.5.

### Moisture transport to the substrate

SLCs are generally cast on concrete, but sometimes on other materials or "inert" substrates as well. If cast on concrete, water from SLC is transported down into the concrete. Using a primer significantly decreases the initial moisture transport to the substrate. In the long run, however, moisture redistributes between SLC and the substrate, even if a primer is used. The moisture state of the substrate may thus be important for the long-term moisture state of the SLC and the floor covering.

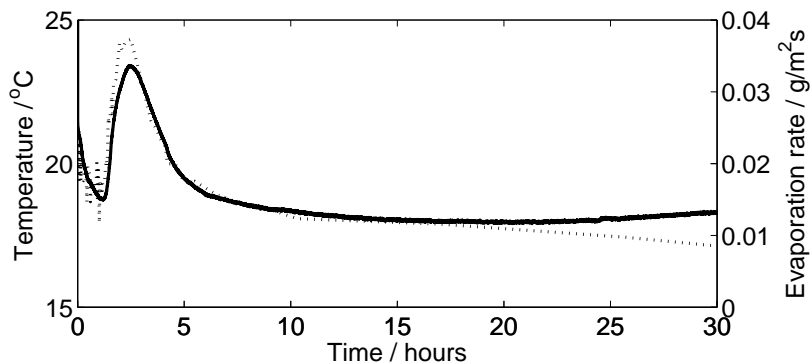


Figure 4.5: Temperature (solid line, left y-axis) and rate of surface evaporation (dotted line, right y-axis) as functions of time for an SLC stored in a climate room at 20 °C and 55% RH.

## Total drying

Before floor coverings are laid on SLCs and concretes, one normally wants the RH of these materials to be below a certain level, for example 85%, so as to minimise the risk of degradation problems and secondary emissions. Figure 4.6 shows the state of the water in the three SLCs dealt with in this study, after three months of hydration at 20 °C under sealed conditions. It can be noted that since self-desiccation (chemical and physical binding) of product A is not sufficient to bring its RH down to the level needed for practical applications, surface evaporation is an important drying mechanism for this product. The rapid-drying product B self-desiccates down to a level that corresponds to approximately 97% RH. Certain surface evaporation is also needed, therefore, in order for this product to meet the requirements for general applications. Since product C self-desiccates down to 85% RH, it can be regarded in this context as a truly self-desiccating material.

## A computer-based drying model

The moisture properties measured and the drying model outlined in this study were used to develop a computer-based programme that can be used to predict both the drying and the long-term moisture states in SLCs as a function of the drying climate and the moisture permeability of the floor covering. The computer-based simulation programme includes three of the four drying processes discussed above (moisture flow to the substrate is not included). It relates each of the properties of the SLC to the hydration state as measured by isothermal calorimetry. The programme is described further in [66]. A typical result for a simulation is shown in Figure 4.7.

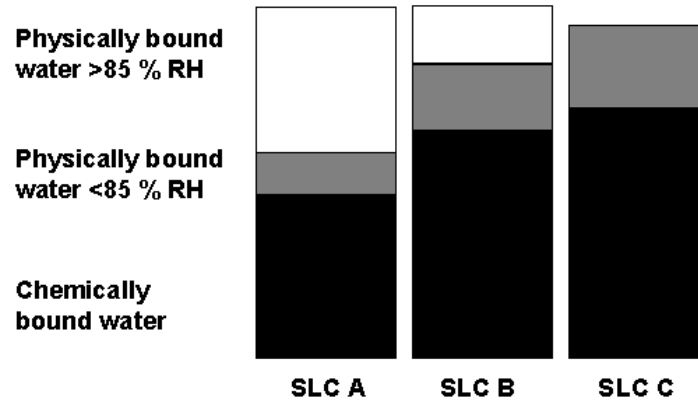


Figure 4.6: Water in three typical SLCs after three months of hydration: SLC A normal drying, SLC B rapid drying and SLC C for industrial floors. The black boxes represent chemically bound water, the grey boxes physically bound water up to 85% RH and the white boxes physically bound water above 85% RH.

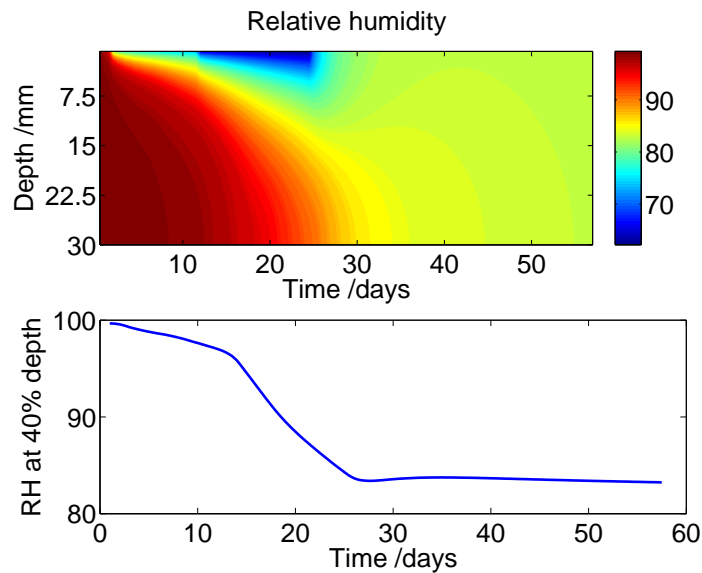


Figure 4.7: The simulated drying of a 30 mm normal drying SLC at 20 °C using a surface mass transfer resistance of 160 s/m. The RH in the ambient air was 90% during the first two days, followed by ten days at 70% and another 13 days at 60%. After these 25 days, an impermeable floor covering was applied, allowing the residual moisture to redistribute.

# Chapter 5

## Alkalinity

A pH above 7 denotes alkalinity, that is an increase in hydroxide ion concentration compared with the neutral state, pH 7 (at 25 °C). Most inorganic binders have an alkaline pore solution (high pH) with  $\text{OH}^-$  and such cations as  $\text{Ca}^{2+}$ ,  $\text{K}^+$ , and  $\text{Na}^+$  that are products of the hydration reactions. The pH of a concrete based on Portland cement is 12.6-13.9 [67], whereas the pH of the SLCs treated in the present study is lower, about 11 [68, 69]. Since some polymers are sensitive to a high pH, the combination of moisture and high pH may lead to the degradation of these polymers.

When exposed to air, the pH of cementitious materials decreases due to carbonation (the process by which carbon dioxide from the air is dissolved in the pore solution forming carbonic acid that lowers the pH). The rate of penetration of this process into a cementitious material is mainly dependent upon the pore structure of the material (a denser structure gives lower rates) and the moisture state (the diffusion of gases is several orders of magnitude faster in air than in a liquid). Since the reactions take place in the pore solution, there is a particular moisture level at which the rate of carbonation is highest. Under constant conditions the carbonation depth is considered to be proportional to the square root of time [67]. In a hydrating and drying material this is not the case, however, since the pore structure and the moisture state change in the course of time.

When two materials of differing alkalinity are combined, ion transport between the materials may occur. There are five main transport mechanisms for ion transport [70], although here only convection and diffusion will be considered.

A qualitative account of the development of the moisture state and the pH in a combination of concrete and SLC is provided in Figure 5.1. The newly cast concrete has pH 13 and a high moisture state (A), but immediately starts to dry and to carbonate at the surface (B). After some time, an SLC at pH 11 is cast on top of the concrete



## Transport of hydroxide ions in an interface between concrete and an SLC

Paper V describes measurements of the distribution of hydroxide ions in an interface between a concrete substrate and an SLC, pH profiles measured after 6 and 15 months being presented. The measurements show only little transport of hydroxide ions in the specimens, even after 15 months of storage at a moisture state above 95% RH. This indicates that SLCs can be used as alkaline barriers between concrete and flooring materials, also on a long-term basis.

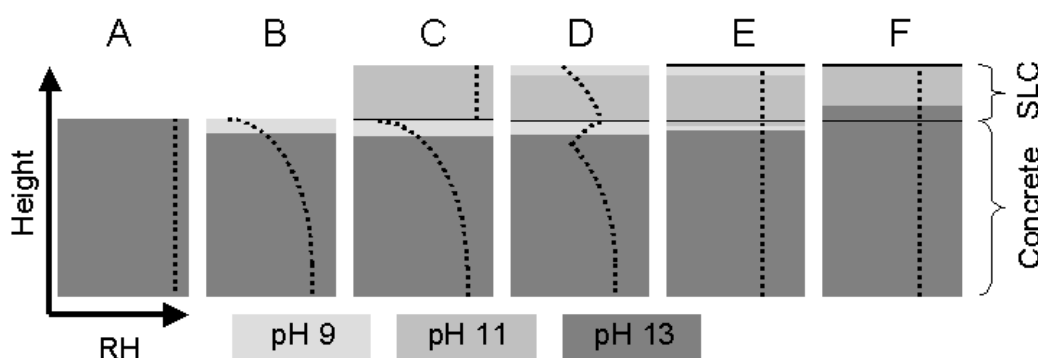


Figure 5.1: Schematic diagram of the distribution of moisture (dotted line) and alkali (different shades of grey) in a floor construction during drying and after a vapour-tight floor covering (thick black horizontal line) has been applied.

(C). The moisture from the SLC both evaporates and is absorbed by the concrete, tending to result in a rather complicated moisture profile (D). The SLC then carbonates at the surface before the vapour-tight floor covering is applied. The moisture is then redistributed from the more humid parts at the bottom of the construction towards the drier parts at the top, bringing ions in convective flow (E). When the moisture is redistributed, the transport of hydroxide ions continues by diffusion from the concrete to, and in the long run possibly through the SLC (F).

Two previous studies have reported on the transport of ions by moisture convection in concrete [71, 72]. However, another earlier investigation found little or no transport of hydroxide ions from concrete to SLC at RH levels estimated to be 85-95% [68]. The results in the present study confirm only limited transport of hydroxide ions occurring in the hygroscopic range, see Figure 5.2. Although the magnitude of

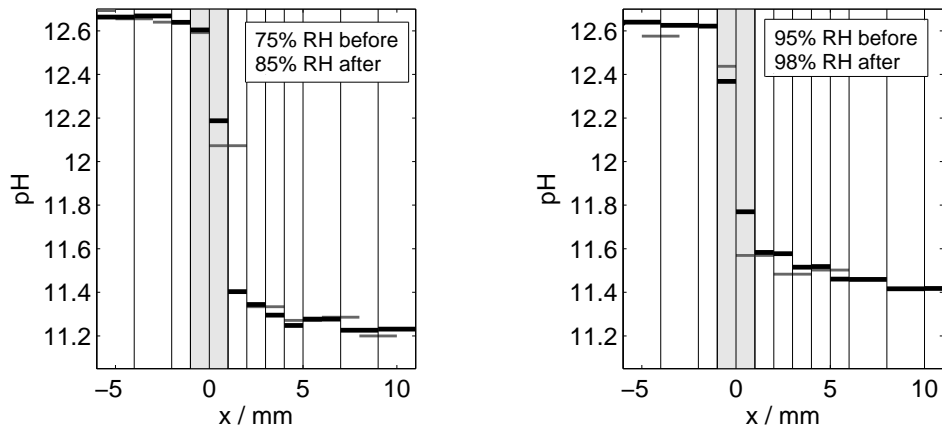


Figure 5.2: Measurements of pH in 1 or 2 mm layers at an interface between concrete and SLC. The horizontal grey and thick black lines show measurements 6 and 15 months after SLC was cast, respectively. The vertical lines show the layers measured after 15 months. In both diagrams, concrete is on the left and SLC on the right and the interface between concrete and SLC is in the grey zone. The legends show the storage RH before and after the casting of the SLC.

the hydroxide ion transport is low, it may be of importance since hydroxide ions are consumed in alkaline hydrolysis, so that new ions are required for the degradation process to continue. Carbonation often occurs prior to application of the floor covering. This thin layer provides initial protection against the higher alkalinity in the non-carbonated material, but may be unable to serve as a long-term protective barrier due to its limited thickness. In Figure 5.3, pH profiles are modelled using the error function solution to Fick’s law of diffusion, employing the estimated maximum apparent diffusion coefficient obtained in the present study.

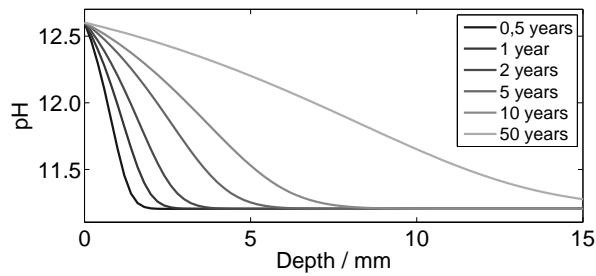


Figure 5.3: Simulated pH profiles in an SLC cast on a concrete substrate as a function of time, using an apparent diffusivity of  $10^{-14}$  m<sup>2</sup>/s. Note that the hydroxide ion concentration has been recalculated into pH on the y-axis.



# Chapter 6

## Emissions

An emission is something that is discharged or released (emitted), such as heat, light, sound, gas, or radiation. In the present report, however, the term emission refers only to vapours with the exception of water vapour. This is often called chemical emission.

Organic compounds constitute the dominant form of emissions. Such emissions are normally divided into different groups, depending on their volatility (as measured by their boiling point). The most common group associated with the indoor environment is that of VOCs (volatile organic compounds) that have boiling points in the range of 50-100 to 240-260 °C [73]. VOC in building materials can be measured, for example by a FLEC (Field and Laboratory Emission Cell) [74] or by chamber methods [75], the emissions being collected on a sorbent and later being analysed in the laboratory. The results of measurements are normally presented as emission rates,  $\mu\text{g m}^{-2}\text{h}^{-1}$  or concentrations,  $\text{mg m}^{-3}$ . Besides VOC, more volatile compounds such as formaldehyde (VVOC, very volatile organic compounds) and ammonia are frequently measured. Larger molecules such as the plasticizers and co-solvents in paints are classified as SVOC (semi-volatile organic compounds).

VOC concentration can be measured either individually or as a total VOC (TVOC). Although VOCs appear to have negative health effects and to be a cause of discomfort in indoor environments [27, 28], and various VOCs appear to differ in their health impact, neither individual VOCs nor TVOC have been found to be relevant risk indices for indoor air quality [28]. However, due to limitations in measurement techniques and in knowledge, TVOC is still commonly used in measuring indoor air quality. Lately a proposal to subdivide VOCs into different categories depending on their chemical and biological reactivity and toxicity has been presented [76].

Emissions in the indoor environment may come from furniture, building materials, microbial growth, human activities and the like or through exchange with the

## Degradation of floor adhesives

Paper VI describes the measurement of degradation for two different floor adhesives, one based on acrylate co-polymers known to degrade in humid alkaline environments forming volatile products and the other, based on ethylene-vinyl acetate (EVA) co-polymers and considered not to degrade forming volatile products in humid alkaline environments. Both adhesives were exposed to different concentrations (pH) of NaOH in aqueous solution, the rate of degradation being measured in an isothermal calorimeter in terms of thermal power. The thermal power of both adhesives was found to be clearly influenced and to a similar extent in both cases by pH. Head-space measurements of emissions from the acrylate based floor adhesive applied to three different substrates, one cement mortar and two SLCs (all of the specimens "non-carbonated") were also made. The materials were cast in small ampoules and were conditioned to different humidity levels: 75, 85, 91, 95 and 98% RH. After 10 months, headspace measurements of VOCs were performed. Clear differences between the substrates of differing pH could be noted, but the moisture state appeared to have no effect on the results.

outdoor air. Emissions in buildings are normally highest when the buildings are newly constructed or have just been renovated. According to Brown [77], concentrations of VOCs are normally about an order of magnitude (tenfold) higher in new buildings. These initial emissions come from construction materials and the contents of buildings [77], such as from paints, adhesives and furniture. There is a strong decrease in emissions during the first six months thereafter [78]. An investigation of primary emissions (see below) from flooring materials presented in [79] reports about a 2/3 decrease between one and six months of age.

Emissions from materials are normally divided into two main categories: primary emissions and secondary emissions. Primary emissions are emissions of compounds present in a material. In principle, primary emissions decay over time under constant conditions, but in reality emission rates may vary with the temperature and moisture content, for example. Examples of primary emissions are formaldehyde from chipboard, solvents from adhesives and paints, ammonia from Portland-cement-based materials and terpenes from wood. Today measurements of primary emissions are made for most indoor surface materials and there are several standards for the measurement of primary emissions from building materials. Production developments, new measurement techniques and new labelling systems have led to the development of low emitting materials and to a general decrease in primary emissions from building materials [80].

Secondary emissions stem from degrading processes such as oxidation and hydrolysis [29] which give rise to volatile compounds. These processes can occur both in single materials and in combinations of materials that in some way interact, e.g., flooring adhesives [31] and PVC-flooring [48] on moist concrete. Secondary emissions are suspected of having greater impact on health than primary emissions do [81]. The measurement of secondary emissions is complex in nature since it can involve several different materials reacting over long periods of time. Completely different results for measurements made under differing conditions or at different times may be obtained.

Inorganic binders in the pure state do not produce any significant primary emissions, but additives such as grinding aids for Portland cement and admixtures to concretes or mortars may give rise to measurable amounts of emissions. However, secondary emissions from materials being degraded while in contact with moist cement-based materials, Figure 6.1, are of greater importance in indoor environments. Concrete is cast with use of water. It forms a fine and dense porous material containing large amounts of highly alkaline pore water. If the concrete is not dried sufficiently, the pore solution comes in contact with the adhesive and the flooring material, possibly degrading it by alkaline hydrolysis. A common degradation process in an alkaline environment is

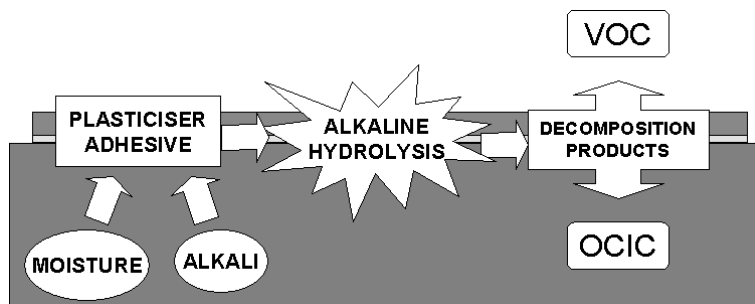
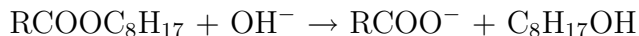


Figure 6.1: A model of the degradation process adapted from Sjöberg [31]. Alkali is transported at high moisture levels through a concrete slab up to the floor adhesive and the flooring, where alkaline hydrolysis can occur. The reaction products may be emitted to the indoor air as VOC or stored in the concrete as OCIC (organic compound in concrete).

the hydrolysis of ester groups to produce alcohols, for example, 2-ethyl 1-hexanol [32]:



where R is an organic group. Both the alkalinity (pH) and the humidity level influence the rate of hydrolysis [31, 82]. The rate of hydrolysis of dissolved compounds in a solution is generally proportional to the hydroxide ion concentration [83]. For adhesives

on alkaline substrates the trend is probably the same, even if the situation is more complex. Moisture is essential since it acts as a transport medium for hydroxide ions from cement-based materials that react with components of other materials and since it allows reactions to occur in solution. Not only the moisture state and the pH are relevant here, but an increase in temperature has also been found to increase the rate of alkaline hydrolysis [84].

Just as production times in the building sector have decreased in recent decades, drying times have shortened considerably as well. Various investigations of the drying of concrete and limits to when other materials can be applied to concrete and to other types of cementitious materials have been carried out, concerned both with moisture and alkali; see for example [32, 48, 85, 82].

In the present study the effect of pH on the rate of alkaline hydrolysis was investigated for floor adhesives by using two methods. Figure 6.2 shows the results of measurements in a calorimeter in which two floor adhesives, based on acrylate copolymer and EVA polymer, respectively, were subjected to solutions of differing pH. The results

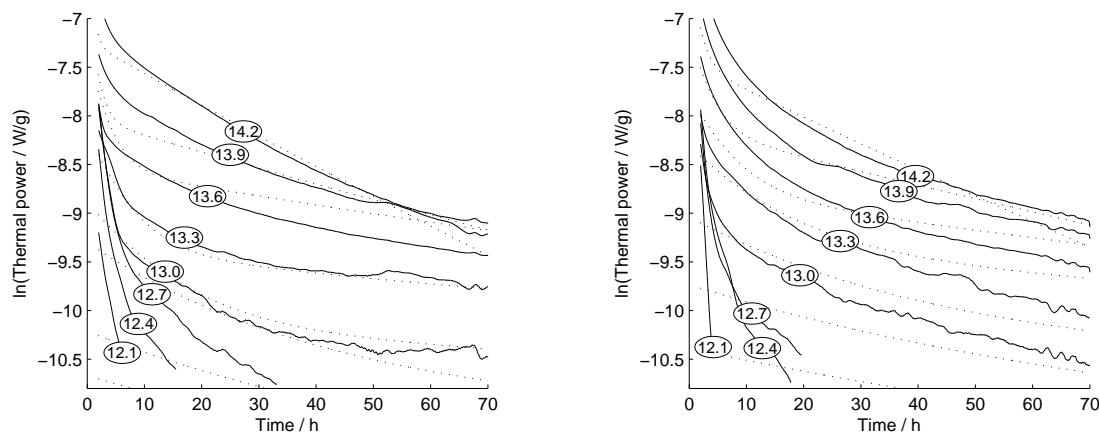


Figure 6.2: The heat production rate as a function of time for two floor adhesives exposed to differing concentrations of an NaOH solution (given as pH) in water. The solid lines are measured values and the dotted lines represent simulated values.

obtained show similar effects of pH for the two adhesives. VOC measurements on adhesives adhered to different cementitious materials showed basically the same results.

# Chapter 7

## Conclusions

In the present study, measurements of the moisture properties of SLCs were made. The drying of an SLC was divided up into four processes: the chemical binding of water, the physical binding of water, surface evaporation and moisture flow to a substrate. The importance of these four processes as well as related properties were measured for three typical SLCs. The chemical binding of water was generally found to represent the main drying process, the drying by surface evaporation that was required in order for the practical moisture requirements to be met also being found to differ widely for different types of products. A model for the drying of SLCs which relates all of the properties of the SLC to the hydration state as measured by isothermal calorimetry (produced heat) was outlined. The model was developed into a computer based-programme able to simulate the drying of SLCs and suitable for assessing the long-term moisture state of SLCs in a floor construction.

Results of measurements of the transport of hydroxide ions from a concrete to an SLC, as well as within an SLC, show this only to occur at low levels, in the hygroscopic range. The low levels of transport of hydroxide ions implies that a layer of SLC can act as an alkaline barrier between a sensitive floor covering and a concrete slab.

The rate of degradation of two common floor adhesives based on acrylate co-polymers and on EVA was found to be clearly related to the concentration of hydroxide ions (pH). High rates of degradation were obtained at pH levels above 12 in the solution. One typical degradation product was detected when the acrylate-based adhesive was applied to a Portland-cement-based substrate, but not when applied on an SLC. Carbonation is thus essential for avoiding high levels of emissions when sensitive floor coverings and sensitive adhesives are applied to a concrete slab, but not when an SLC of significantly lower alkalinity is employed. The carbonated layer at the concrete surface may, however, due to its limited thickness, be unable to serve as a long-term protection in terms of secondary emissions. This is better provided by an SLC of lower alkalinity.





# Chapter 8

## Future investigations

This study has mainly been concerned with the moisture properties of SLCs and with methods for determining them. The transport of hydroxide ions ( $\text{OH}^-$ ) and the consequences this has was also investigated. The study has clarified various issues, but has also raised a number of questions of interest in future work in this field.

- A large part of the water in SLCs is initially within to the superhygroscopic range. Measuring such moisture properties as moisture sorption and moisture transport in the superhygroscopic range requires other techniques than those used in the present investigation. Measurements of these moisture properties were not performed here, but are of definite relevance to obtaining a better understanding of what occurs in the initial phase of SLC hydration. It is also of interest for the simulation of the initial moisture transport in SLCs.
- Laboratory results showed that a high percentage of the water in a fresh SLC evaporates during the first hours. Whereas climate rooms in the laboratory provide stable climatic conditions and high air exchange rates, the climate at construction sites can vary considerably, especially during the first hours and days after casting when large amount of water evaporate to the indoor air. Field studies monitoring the climate when SLCs are laid would thus be of interest to be able to relate the results of laboratory studies to field conditions.
- Moisture transport is temperature-dependent, but only little has been done to investigate this for cement-based materials. Further investigations within this area are needed therefore, in order to learn more about moisture flows both at different constant temperatures and under non-isothermal conditions.
- Ettringite can release chemically bound water at low RH. Since ettringite is the main reaction product of SLC, it would be of interest to investigate further the behaviour of ettringite at low RH and different temperatures.

- The studies carried out showed only limited transport of hydroxide ions to occur in concrete and SLCs at moisture levels within the hygroscopic range. Further measurements performed under longer periods of time are needed to confirm these findings and to provide more accurate figures concerning the apparent diffusion coefficients and the ion transport by moisture convection during drying and the redistribution of moisture after floor coverings have been applied.
- Measurements of the degradation of sensitive materials in contact with cementitious materials and the resulting emissions from such degradation are of clear interest. Such measurements require a method using relevant substrates and an understanding of how pH and RH influence the degradation processes. No such testing method is available at present.
- Ettringite contains large amounts of water hydrate. However, when there is a shortage of water during the reaction due to surface evaporation and self-desiccation, monosulphates containing lesser amounts of water hydrates may form instead, or ettringite may reform into monosulphates, as is the case with sulphate shortage. This would be of interest to investigate since such behaviour will influence the drying process.
- The water activity, or the relative humidity, is generally considered to be the factor best describing the influence of the moisture present on the chemical and biological processes that occur in materials. Studies have showed, however, that hysteresis effects (moisture content) also influences these processes. It would be of interest, therefore, to investigate how the rate of the degradation processes is affected by the water activity and the moisture content.
- Chemical reactions continue for a long time after the casting of cementitious materials. Although they are of very low rate, these chemical reactions may influence the moisture state of an SLC by reducing the RH on a long-term basis. Since this could be of importance for predicting the long-term moisture states of various constructions, it would be of interest to investigate further.

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## Peer reviewed conference papers

**A method to study sorption phenomena** Åhs M., Sjöberg A. and Anderberg A., (*Proc. of the 10th International Conference on Indoor air Quality and Climate.*), Beijing, September 2005, pp. 1969-1973.

**Sorption hysteresis and scanning in a flooring screed - implication for moisture induced degradation.** Anderberg A. and Wadsö L., (*Proc. of the 10th International Conference on Indoor air Quality and Climate.*), Beijing, September 2005, pp. 3928-3932.

**A new method to evaluate compatibility of floor constructions** Sjöberg A. and Anderberg A., (*Proc. of the 10th International Conference on Indoor air Quality and Climate.*), Beijing, September 2005, pp. 3933-3937.

**Method for simultaneous determination of sorption isotherms and diffusivity of cement based materials.** Anderberg A. and Wadsö L., *Proc. of the 7th symposium on Building Physics in the Nordic Countries.*, Reykjavik, June 2005, pp. 103-112.

**A method for simultaneous measurements of heat of hydration and relative humidity.** Wadsö L. and Anderberg A., *Proc. of the Third International Research Seminar on Self-Desiccation and Its importance in Concrete Technology.*, Lund, June 2002, pp. 102-107

## Reports

**A computer based programme simulating drying of self-levelling flooring compounds** Anderberg A. and Wadsö L., Report TVBM-7191, Div. Building Materials, Lund Institute of Technology, 2007.

## Others

**Fukt i avjämningsmassor.** Anderberg A., (*Bygg & teknik*), 8 nov. 2004, pp. 18-20.

**Förbättrad mätmetod för limmade golvkonstruktioner.** Sjöberg A. and Anderberg A., (*Bygg & teknik*), 5 aug. 2003, pp. 62-63.

## Drafts

**A method to validate relative humidity generation in sorption balances.** Wadsö L. and Anderberg A., (*Draft of paper*).

**Moisture profiles during drying and redistribution of moisture in self-levelling flooring compounds. Results from experiments and simulations** Anderberg A., (*Draft of report*).

**Chemical binding of moisture, heat of hydration and rate of surface evaporation of self-levelling flooring compounds** Anderberg A., (*Draft of report*).

**Moisture transport properties of self-levelling flooring compounds and their dependence on curing conditions.** Anderberg A., (*Draft of report*).

# Dissertations from Building Materials, Lund University, Sweden

Mod�er M	A fracture mechanics approach to failure analyses of concrete materials. TVBM-1001	1979
Burstr�m P-G	Ageing and deformation properties of building joint sealants. TVBM-1002	1979
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