

# Hygro-Thermal Properties of Sheep Wool Insulation



**T.M. Tuzcu** Civil Engineering Faculty Delft University of Technology



T.M. Tuzcu Bazuinlaan 50 2287 EG Rijswijk tmtuzcu@gmail.com



### **Thesis Commission**

Prof. ir. J.J.M. Cauberg TU Delft, Faculty of Architecture

Ir. A.C. van der Linden TU Delft, Faculty of Architecture

Dr. Ir W.H. van der Spoel TU Delft, Faculty of Architecture

Ir. B.L.H. Hasselaar TU Delft, Faculty of Architecture

Dr. Ir. A.L.A. Fraaij TU Delft, Civil Engineering Faculty



# Foreword

This report is a part of my graduation research on Civil Engineering faculty, Delft University of Technology. The research was done at Building Physics department of the civil engineering faculty of TU Delft and named "Hygro-Thermal Properties of Sheep Wool Insulation". After a detailed study on wool the properties of sheep wool, the study focused on the water vapor sorption properties and moisture transfer through wool insulation. Secondly, the effect of sheep wool insulation in building constructions on its moisture behavior was analyzed.

With this thesis, I want to present my special thanks to my thesis commission for the understanding and patience they have showed during my research.

Furthermore, I want to thank my mother, my brother and Mr. Vercouteren for their support; and everybody who has contributed somehow to this study.

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# 1 Introduction

The primary energy use for house heating has increased strongly in the second half of the previous century, especially between 1965 and 1975. The environmental and economical impact of this high energy consumption has augmented the use of affordable insulation materials which have moderate environmental impact. With the introduction of different kinds of insulation materials onto the construction market, the energy use has begun to decrease from the beginning of the 80's.

From the viewpoint of sustainability, renewable natural insulation materials are good alternatives for common (inorganic) insulation materials. All natural insulation materials are made from plant or animal sources; their manufacturing processes use very little energy; they are safe for installers and pleasant to handle; and they are re-usable or biodegradable at the end of a building's life.

A variety of plant fibers and animal hair have been used in the past as a material in constructions; but these traditional materials have fallen into disuse. But with the increasing demand on sustainable construction materials, the natural materials have called attention again. Sheep wool is one of these natural products which are becoming more popular in the construction industry. The subject of this thesis concerns the use of sheep wool in building construction with the focus on its moisture transfer properties.



Central Asian nomads have built felt homes in order to move with the herding season since approximately the fifth century B.C. A wood frame composed of long pieces and collapsible trellis provide the skeleton, which is covered in felt and then roped down with a functional, attractive interweaving; Figure 1-1 [1].

Figure 1-1: Old type of wool felted house

# **1.1 Moisture in Insulated Constructions**

It is well known that the presence of an insulation layer in the building envelope affects the moisture transfer and storage in the construction layers. When positioned between an inner and outer leaf, the temperature of the outer leaf of the wall will be quite close the outside temperature. In wintertime, condensation may therefore occur in this cold layer due to vapor transfer through the wall which originates from inside. Interstitial condensation due to vapor diffusion may cause decay of the structure itself. Sometimes, it may visually appear as mold growth on the interior finish, or as ceiling discoloring, ceiling leaks or paint peeling. In many cases, the use of a vapor barrier between insulation and inner leaf offers sufficient reduction of the moisture transfer rate such that interstitial condensation is prevented.



On the other hand, besides using a vapor barrier, one can also make use of hygroscopic properties of (insulation) materials. It is known that nearly all (construction) materials absorb moisture from the surroundings in their environment, but the amount of absorbed moisture depends on the hygroscopic moisture capacity. As results of the hygroscopic moisture capacity of materials, the adverse effects of condensation can be reduced. As the (insulation) material acts as a moisture reservoir; the relative humidity inside the material is reduced.

Besides absorption, all (insulation) materials provide resistance to water vapor diffusion. The  $\mu$ -factor is a measure of this characteristic, indicating the resistance relative to that of still air. If the materials, which the building envelope consists of, have high resistance (high  $\mu$ -factor) the flow of vapor will be retarded. The term  $\mu d$  ( $\mu d = \mu$ \*thickness of the material layer) refers to its vapor resistance and is usually used in calculations. If the insulation layer has a higher resistance, less moisture is transferred to the cold outer leaf. In many cases the use of vapor barriers (retarders) with a moderate diffusion resistance (µd approx. 5m.) offers sufficient control of interstitial condensation during the heating period. In that case, the resistance of the insulation doesn't play a significant role. But in absence of a vapor barrier, the resistance of the insulation (in combination with that of the inner leaf) does determine the rate of diffusion towards the cold out leaf. A vapor barrier may then be omitted only if the inner leaf has substantial resistance and if the outer leaf in combination with the insulation layer provides sufficient capacity to absorb water without adverse effects. For analyzing the performance of a building construction with respect to moisture diffusion, it is thus important to know the moisture absorption and vapor resistance properties of the respective materials.

# 1.2 This Study

Because sheep wool insulation, as a product, is very new on the market and also produced at this moment on relatively small scale, the general properties of sheep wool and especially the properties regarding moisture transfer have not been examined in great detail yet. Therefore, in first instance this study aimed at accurately measuring the moisture transfer properties of wool (samples) in the hygroscopic range.

## **1.2.1** Moisture transfer properties of wool

Traditionally, the vapor transfer inside a material is governed by sorption on one hand, and on the other hand by the resistance to vapor flow. For sheep wool, the (steady-state) sorption behavior of wool has been studied extensively: sorption-isotherms of wool under different circumstances can be found in the literature. Also the vapor resistance has been studied in some detail. But, the  $\mu$ -factor of two wool insulation products with identical mass density can lie far from each other, both being measured according to the German norm (DIN 52615). "Doschawol" in the Netherlands points out that the  $\mu$ -factor lies between 4 and 5; while "Sheep Wool Insulation Ltd.", in Ireland points out that the  $\mu$ -factor lies between 1 and 2. Because of these differences, it was decided to measure the  $\mu$ -factor of sheep wool insulation in this study using the well-known cup method. In addition, since wool samples may exhibit varying properties, the sorption isotherm of the wool was measured as well. And finally, besides these two experiments, a third



experiment was carried out which focused on the time and space-dependent vapor transfer in a column of wool. The data from this quite unique experiment may indicate whether the vapor transfer inside the wool can be described using a Fickian diffusion model (i.e. one effective differential equation for the vapor transfer) or that non-Fickian effects do occur.

### **1.2.2** Application of Sheep Wool Insulation in Building Constructions

Secondly, the effect of sheep wool insulation in building constructions on its moisture behavior has been analyzed. The results of the experiments are applied in a simulation program, as material property inputs, to analyze the behavior of sheep wool insulation in different climates and constructions.

Practical cases of thatched roofs are modeled and analyzed with the simulation program "WUFI". The interstitial condensation problem in constructions, which occurs when water vapor moves under temperature gradient -in hot and humid weather circumstances-to the colder side of the construction and forms interstitial condensation, was thought to be solved by using absorption property of sheep wool (insulation). Therefore, the sheep wool insulation (with and without a vapor barrier layer) was applied under both types of the thatched roofs (open-, and closed structure) in the simulations, and the results were analyzed.

### **1.2.3** The Thesis

The first chapter includes the information which is more interesting for the Material Science departments of the institutions. The properties of wool are discussed in second chapter which is relevant regarding general properties and regarding the use of wool in the indoor environment. After gathering information about wool, the theory of vapor transport in porous materials is discussed in chapter 3 with the focus on relevant properties of sheep wool in chapter 4. The experimental setups are presented in chapter 5 and their results are discussed in chapter 6, which concluded the first part of the study. The second part, on the application of sheep wool in several construction types, will be analyzed within chapter 7. In the last chapter (8), conclusions are drawn and recommendations for further research are given.







# 2 Properties of Wool

# 2.1 Wool

Sheep are the most adaptable of all domesticated animals and they are a provider of a natural fiber, wool. One of the first animals to be domesticated was sheep, some ten to twelve thousand years ago. This very early domestication was due to mainly two important factors. Sheep provided humans with two sources of food - meat and milk as well as producing a very useful fiber, plus a skin.

The adaptability has necessitated a variety of types being utilized in these environments. Today sheep are farmed in most countries around the world, from the hot regions of Australia, Asia and the Middle East through to cold, bleak conditions in Northern Europe, Russia, Iceland and Southern America with about 200 different breeds and crosses producing a great variety of grades and types of wool.

Wool is a valuable by-product of animal meat production. Wool had been a principal force in the economy of Western civilization, by providing basic essentials but also providing capital and luxury goods. Nowadays the consumption of wool is more limited in the end use due to the introduction of artificial fibers. In spite of many countries producing wool, the market is on the hands of the six main countries (France, Germany, Italy, Japan, UK and USA). There are, however, a number of other products made from wool and a few of the interesting ones include: Mattress and furniture stuffing, Tennis ball covers, Piano hammers, *Insulation*, Felt hats and toys.

World Production of Raw Wool (greasy wool equivalent)			
2002/2003	Million kg		
Australia	544		
China	302		
New Zealand	235		
(Former) Soviet Union	133		
Argentina	72		
Turkey	70		
UK	50		
South Africa	45		
Uruguay	43		
Pakistan	40		
World Total (including other countries)	2194		

Table 2-1: Source: http://www.britishwool.org.u
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Table 2-2: Industrial	Usage	of Wool,
Source Lit: 2		

Wool used in	Million kg
Menswear	101.1
Womenswear	150.1
Knitwear	89.7
Other Apperal	73.7
Carpets	165.9
Other Interior Textiles	70.6
Total	651.1

The reason of the difference in the "*totals*" of Table 2-2 and Table 2-1 is that the majority of the wool produced by the world's sheep is used locally and less than half becomes a on the international market [5].

Except for the countries Australia and New Zealand, nowadays sheep is usually farmed for meat production. This makes wool a by-product of sheep. Sheep provide wool at an acceptable price (production of one kg greasy wool needs the feeding equivalent of 70 kg

grain or a feeding cost of US\$10/kg wool, if fed with grain; while the price of raw wool is not more than US\$4/kg) [3].

### 2.1.1 Quality of Wool

Wool types are classified according to fiber diameter and length. Fineness is considered as the most important quality attribute because fine wool has more crimp. Crimp means the waving ability of the wool fibers and crimp helps individual fibers stick together during spinning and results in a stronger product. The finer the wool, the more waves and scales are per centimeter present, which can be called as the unit of the crimp (waves/centimeter). The crimp property of the fiber is also influenced by the humidity of the environment [7]. Fine wool often has 38-40 crimps per centimeter; coarser wool has less. Length of fiber is another quality factor; the longer the fiber, the better quality the wool has. The type of wool in greatest demand on the market today is merino because of the uniformity and the fineness of the fibers.

### 2.1.2 Harvesting and Processing Wool

Sheep are usually shorn in the spring/ summer months; this may be twice a year or three times in two years. The time and frequency of shearing has a great affect on the quality of harvested wool as it can have a major influence on important fiber characteristics.

The basic techniques of wool processing, regardless of the end product, are the same. Essentially the wool is washed in hot, soapy water (i.e. scoured) to remove dirt, grease and other impurities. It is then carded to separate the fibers from each other, take out any tangles and clumps, break up staples, and align the fibers so they are ready for the next process. The carded fiber is pin-drafted on machines to be able to get parallel fibers, which then goes into products.

**Scouring:** washing the wool before further processing. The scouring water is normally about 65°C, which is hot enough to dissolve the wax (i.e. lanolin), and detergent is added to help remove the dirt from the fibers and to emulsify the wax so it doesn't stick back onto them. This stage is also the stage where the chemical agents (according to the end usage) are applied to the wool.



Picture 2-1: Scouring process of Sheep Wool



**Carding:** is a process that passes the wool through a series of rollers covered with fine bristles or fine wire teeth. This separates the wool out from tangles, clumps and staples and lays the fibers parallel and formed into a fine web.





Picture 2-2: Carding Process after Scouring





Picture 2-3: Wool is combed into nearly parallel fibers [4]

### 2.1.3 Mechanical Properties of Wool

Clean wool contains 82% of the keratinous proteins, which contain high concentration of sulphur (3%). The amount of sulphur in the keratin determines the strength of wool because of strong disulfide bonds<sup>1</sup>. Keratin does not dissolve in cold or hot water; and does not breakdown into soluble substances (I APPENDIX WOOL, I.3). The outer surface of the wool fiber is responsible for wet ability, physical properties and felting<sup>2</sup> behavior.

Dry fiber breaks at about 30% elongation whereas a wet fiber (100% humidity) doesn't break until it extends 70%, whereas the fiber after stretching returns rapidly and completely to the original length. Elongations more than 70% cause cracks in wool [6,7].

The water (vapor) works as a plasticizer on the fiber [8]. The larger the moisture content of the fiber, the further it can be stretched. Furthermore; the temperature has the same effect as the moisture content. As the temperature rises, the fiber becomes weaker and

<sup>&</sup>lt;sup>1</sup> A disulfide bond (SS-bond), also called a disulfide bridge, is a very strong chemical bond between two sulphydryl group; and is very important to the folding, structure and function of proteins. The disulphide bonds are responsible for the relatively good strength of wool fibre.

<sup>&</sup>lt;sup>2</sup> Felting occurs when individual fibers move in one direction. Such movement occurs when the fiber assembly is agitated in water.

becomes easy to elongate. Both the effects of acid and alkali cause a decrease in the strength of the wool fiber.

### 2.1.4 Chemical Reactivity of Wool

Wool proteins have mainly a spiral structure with amino acid ends with the general formula of COOH-CHNH<sub>2</sub>-**R**. Different amino acids distinguish themselves from each other by the active **R**-rest group (See also Appendix I.5). The reactive nature of wool enables wool to react with many gases and metals in its environment. Gases and metals bond chemically to wool by making cross-links between two rest groups of amino acids. The chemical and physical structure of wool gives the ability to absorb many other types of molecules including SO<sub>2</sub>, NO<sub>x</sub>, O<sub>3</sub>, Aldehydes, and VOC. The sorption of some of these gases are reversible (General VOC, Phenol... see Appendix I.6) and some not (NO<sub>x</sub>, SO<sub>x</sub>, Formaldehyde...see Appendix I.6). The studies of Schmidt (et. al. 2003) about the sorption behavior of gases for wool have shown that wool has a high capability to absorb tobacco smoke components and VOC's. On the contrary, the desoprtion is very slow.

Because wool is heavily cross linked by disulphide bonds (See also Appendix I.3), it is insoluble in almost all solvents, except for alkalis. Alkalis, damage the wool fiber even in diluted solutions. Generally it's accepted that alkaline which is higher than pH=11 causes damage. The increasing temperature of the environment also accelerates the reaction. The wool fiber shows a strong absorptive behavior for alkalis. In alkaline fluids with pH higher than 11, the wool swells and finally dissolves in alkalis [7]. Soda work less aggressive and ammonia is almost harmless.

The effects of acids on the wool fiber are much less serious than that of alkalis. Diluted mineral acids cause almost no damage, or only a little, on wool. In very high dilution, the acids even work as protector for the fiber. Also swelling with acids is low in comparison with wool in an alkaline solution. The effects of diluted acids on the wool fiber are specific distinguished of those of alkalis. Swelling occurs less frequently than in alkaline solution [7] because the isoelectrical<sup>3</sup> point (pH value) of wool is pH = 4,8. A solution of pH 4-5 has less influence than pure water, en even works protective.

Soap doesn't cause any damage on wool. Soap can be split up a little, and therefore the soap particles are too large to penetrate into the wool fiber [7].

### 2.1.5 Moisture-Sorption Ability of Wool Fiber

Wool fibers exhibit hygroscopic behavior, which allows them to absorb up to 33-34% (kg/kg) of its dry weight. The experiments [35] which were done with different types of wool show that the general sorption properties of different wool types are almost the same. The adsorbed amount is inversely proportional to fiber diameter, which is probably because of the available surface area for the sorption (Morooka et al., 1991).

<sup>&</sup>lt;sup>3</sup> Isoelectrical Point of a protein is the pH at which the protein has an equal number of positive and negative charges. The isoelectric point is of significance in protein purification because it is the pH at which solubility is often minimal.

E.V. Truter [8] has found out that wool contains 33% water by weight at 100% RH. Truter mentions that the density of dry wool is 1,307 gr/cm<sup>3</sup> [8], while the density of wool at 100% RH is 1,266 gr/cm<sup>3</sup>. He reports that the swelling of the wool fiber rises up to 40% at cross- sectional direction. Scholl and van Rosmalen [7] have also found out that the wool fiber swells in width where the cross- section increases up to 40; while, on the other hand, the increase of the fiber length was only 1%.



Figure 2-1: Schematic bonding structure of Wool-water system, source: Doschawol

Swelling of the fibers occurs when water molecules enter between the hydrogenbonded wool molecules by breaking a large number of hydrogen bonds between and within the protein chains (Figure 2-1) [34]. The relation between wool and water is interdependent. The swelling of the fiber increases with the adsorption, and the more the fiber swells the more water it absorbs [7].

The kinetic behavior of sorption depends on the initial water concentration in the fibers, and also on the length of time that the fibers have been at that concentration. Entry and exit of water from keratin is partially governed by structural changes which occur during sorption. Watt mentions [49, 50, and 51] that the molecular organization of keratin changes with water if water is present within the keratin; and the extent of such changes depend on the water concentration.

### 2.1.6 Wool against Fungi and Insects

Bacteria and fungi can always be found on wool and as always under favorable conditions (temperature, relative humidity and pH) and; they grow and may attack the fibers.

Keratinaceous materials (feather, hair, **wool**, horn) are insoluble and resistant to degradation by common enzymes because of high degree of cross linking by disulfide bonds, hydrogen bonding and hydrophobic interactions [10]. One of the possible mechanisms of breakdown of keratin is reduction of disulfide bonds. From the many micro organisms that can be found on the wool, only four of them are well-known as causing degradation of wool, namely *Proteus vulgaris*, *Pseudomonas aeruginosa*, *Aspergillus and Penicillium* [22].

Wool can be attacked by the larvae of certain moths (Lepidoptera) and beetles (Coloeoptera) (APPENDIX I.7). The most important wool-digesting insect pests are: Tineola bissiella (common cloth moths), Tinea (case bearing cloth moth), Hofmannophila pseudosprettella (brown house moth), and Anthrnus (carpet beetle).

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### 2.2 Wool as an Insulation Material

The aim of insulating buildings is to slow down heat transfer. Insulation materials are required to keep buildings cooler in hot climates, or warmer in cold climates. As more insulation is installed, more comfort (thermal and soundproofing) is created; and operating costs are lowered.

Insulation materials are divided into three main classes; namely *a-biotic insulation materials* (foam glass, glass wool, and rock wool), *biotic insulation materials* (Cellulose, hemp, coconut fiber, cork, sheep wool and flax wool) and *chemical insulation materials* (polystyrene and polyurethane). The thermal properties and the durability of all insulation materials are more or less the same (Table 2-3). Because of these nearly equal properties, the attention is paid nowadays to the fire resistance of the materials; and the problems due to occurrence of moisture in the structure (inertial condensation and Indoor Air Quality), and how to avoid these problems using materials available.

 Table 2-3: Properties of Insulation materials (The chosen materials are compared with the most important properties regarding Building Physics)

Material	Hygroscopic	Density p	Thermal	µ-Factor	Life time	Fire	Combusti	Specific Heat
	Capacity [weight%]	[kg/m <sup>3</sup> ]	conduct., λ[W/mK]		[year]	Resistance Classification [DIN4102]	on point [ <sup>0</sup> C]	capacity c[J/kgK]
Sheep wool	33	25	0.033	1-2* 4-5**	>100	B2	600	1720
Flax wool	20	32	0.040	1-2	100	B2	570	1550
Glass wool	< 0.1	22	0.034	1-2	100	A1	600	799
EPS	1.1 (by volume)	15	0.040	20-90	70	B1	360	1500

\*Sheep wool insulation Ltd., Ireland (DIN52615), \*\*Doschawol, the Netherlands (DIN52615),

### 2.2.1 Environmental Impacts of Wool Insulation

Hasselaar (2004) reports that the environmental impacts of natural insulation products are less than common insulation products. Because natural insulation materials are made from renewable plant or animal sources; their manufacturing processes use very little energy; and they are re-usable or biodegradable at the end of a building's life.

Sheep wool is used as well for façade insulation as for roof insulation. There are two important health impacts regarding wool. At first; the farmers can get health problems as a result of plunging the sheep twice a year with pesticides. The second impact is in the stage of processing wool, while using insecticides (0.3% by weight Mitin FF is applied), or flame retardant treatments. (APPENDIX I.8)



Picture 2-4: Sheep wool insulation; Source: Doschawol



#### 2.2.2 Fire Resistance Properties of Wool

Resistance against fire is one of the most important properties of insulation materials. The resistances are classified and standardized by organizations (DIN4102, ISO13501-1, etc...). The materials are tested against combustibility<sup>4</sup>, heat of combustion<sup>5</sup>, single burning item<sup>6</sup>, ignitability<sup>7</sup> and smoke hazard.

The test results are classified in six classes; namely A, B, C, D, E, F. Class A products does not contribute in any stage of the fire including fully developed fire; therefore they are assumed to be fire resistant. One other next coming class has less strict requirements than previous class; and Class F for products show no resistance to fire. Two sheep wool insulation products (Doschawol, the Netherlands and Sheep Wool Insulation Ltd, Ireland) have been classified in B2 in accordance with DIN 4102.

The wool fiber may be regarded under most conditions as reasonably resistant to fire. The natural flame-resistant properties of wool are connected with its relatively high nitrogen content (16%), high moisture content (10-14%), high ignition temperature (570-600 0C), low heat of combustion (20.5 kJ/g), low flame temperature (680  $^{\circ}$ C) and relatively high limiting oxygen index (LOI<sup>8</sup>-value = 25-28%). This LOI-value implies that for a complete combustion of wool an oxygen amount of more than 25% is necessary. Our surrounding air contains however 21% oxygen.

The fire retardant treatments for wool were based on the impregnation of borates, phosphates and to a limited degree the organic phosphorus compounds. With the introduction of stricter flammability requirements, the new fire retardant treatments are based on zirconium or titanium salts. A suite of treatments, based on these two activities, was developed under the generic title of ZIRPRO.

Fire statistics reveal that toxic gases are responsible for more fire deaths than are burn injuries. Despite the recognition of CO as the primary toxicant produces during fires,  $HCN^9$  must not be overlooked especially since it is nearly 40 times more potent than CO and much faster acting than CO at levels above the minimum lethal levels [52].

To be able to understand the effects of smoke during a fire, the acute lethality of thermal decomposition products of various materials were investigated in a research [52] by using mice and rats. The amount of gases needed for 50% lethal causalities (LC50) and the time needed for 50% causalities (LT50) were introduced as comparison parameters. The most important gases of the smoke (CO, HCN, CO<sub>2</sub> and low O<sub>2</sub>) were exposed first as single



<sup>&</sup>lt;sup>4</sup> This test identifies products that will not, or not significantly, contribute to a fire regardless of the end use. <sup>5</sup> Determines the potential maximum total heat release of the product.

<sup>&</sup>lt;sup>6</sup> Evaluates the potential contribution of a product to the development of a fire.

<sup>&</sup>lt;sup>7</sup> Evaluates the ignitibility of a product under exposure to a small fire

<sup>&</sup>lt;sup>8</sup> LOI-value is the minimum amount of oxygen in oxygen-nitrogen mixture required to support complete combustion of a vertically held sample that burns downward from the top. The higher the LOI-value, the more resistant is the material against combustion.

<sup>&</sup>lt;sup>9</sup> HCN (Hydrogen Cyanide) is a chemical compound. The toxicity is caused by the cyanide ion, which prevents cellular respiration.

gases in to the test environment, and than as combination of two gases, to be able to understand the effects. Of the two gas mixtures tested (CO, HCN, and low O<sub>2</sub>), the most toxic environment was that of "HCN +low O<sub>2</sub>". The mean lethal blood concentrations of cyanide and COHb were lower during the combination exposures as compared to the single gases.

The LT50 value for wool was 23 minutes (for; Douglas Fir Wood 16min, Nylon 7min. Nylon Carpet 14min, Urea formaldehyde 23min). Decomposition of wool began at 100 0C with a steady mass loss throughout the duration of heating. HCN began to be generated at 286 <sup>0</sup>C (8 minutes), increasing steadily and reached maximum at 567 <sup>0</sup>C (21 minutes). The animals died shortly after the peak of HCN. Oxygen depletion and CO<sub>2</sub> production were minimal. Blood cyanide concentrations were 1.03 mg/L (Acute toxicity is associated with blood cyanide concentrations of 0.5 mg/L or more with a lethal level in a man beginning about 1 mg/L).

All the nitrogen containing polymers studied in that research released HCN. For some, such as Wool and Urea Formaldehyde, it was clear that cyanide was the main toxicant while others CO as well as low  $O_2$  played an important role.

A modified treatment leading to reduced smoke emission includes zirconium acetate. A variant of Zirpro treatment (low smoke Zirpro treatment) lets that treated wool products have very short or zero "after flaming" times by increasing LOI-value up to 27.

### 2.2.3 Problems in Buildings due to Moisture

It has been estimated that 75% of building failures are due to water [55]. Biological organisms such as fungi, bacteria and viruses can cause main problems in the indoor environments. It is well-known that the relative humidity is an important influencing factor for the development and the increase of these organisms. Fungi can cause as well visual embarrassment as damage to the area of the material or in the long run internally damage the constructions. But the biggest problem of biological organisms is the temporary or Picture 2-5: Mold due to inwardly driven moisture permanent effect on the health of people.



trapped by the vapor impermeability of the vinyl wall covering; Lit: 46

The most common indoor fungi are (Fenner, 2001, Adan et al, 2000): Cladasporium, Penicillium, Aspergillus, Alternaria, Eurotium, Wallemia and Mucor. Among these fungi, Cladasporium, Penicillium, Aspergillus, Alternaria and Mucor have been revealed by skin prick tests to be allergenic. It has been shown that building related moisture and microbial growth increases the risk of respiratory symptoms, respiratory infections allergy and asthma. The underlying mechanisms are irritation of mucous membranes,

allergic sensitization and non-specific inflammation [22]. Symptoms like headache, eye, nose and throat irritation or fatigue have been associated with volatile compounds produced by fungi (Samson, 1985).



 Table 2-4: Growth Environments of Various organisms (Concerning health)

In fact, getting relative humidity levels to around 50%, for most of the tome deals with most indoor air quality issues, providing that toxic materials are avoided as much as possible (Table 2-4). Interestingly, certain bacteria can also grow below 40%, which may cause health problems and building decay. Furthermore, Allergic irritations and respiratory infections increase also below 40% relative humidity.

There are numbers of factors which influence the development of fungi spores on a construction surface. The most influencing factors thereby are the surface temperature, the humidity of the air on the surface, pollution of the surface and the pH value of the surface. (For more information about influencing factors for development fungi, see Appendix I.10). It is commonly stated that there is a direct link with the relative humidity of the air and the increase of surface fungi. The other factors influence more to the speed of the fungal growth. Since renewable insulation materials are food sources, they are treated with fungus and moth repelling chemicals to prevent the growth of fungi. Dependent of the technical lifespan of these chemicals, renewable insulation materials form a greater risk to biological contamination than glass wool, which has no nutrition for fungi.

Fungal growth begins after only a short period of favorable conditions (Pasanen et al., 1991). On the other hand, spores may survive for a long time after contaminated materials dry. The required relative humidity range of building materials for fungal growth has been determined to be 76-96% depending on temperature, time, substrate and fungal species (Coppock& Cookson, 1951; Block, 1953; Grant et al. 1989).

The availability of water to fungi in a building material depends on the material's equilibrium relative humidity (RH) or water activity  $(a_w)$ . Water activity  $(a_w)$  is the relative availability of water in a substance and is defined as the ratio of the vapor pressure of water in a material (p) to the vapor pressure of pure water  $(p_o)$  at the same temperature. The term water availability refers to unbound water in the material which is not bound to the material molecules. So, the water activity of a material is not the same thing as its moisture content.

When vapor and temperature equilibrium are obtained, the water activity of the sample is equal to the relative humidity of air surrounding the sample in a sealed measurement chamber. Multiplication of water activity by 100 gives the equilibrium relative humidity (RH) in percent.  $\mathbf{a}_{w} = \mathbf{p}/\mathbf{p}_{o}$  or,

$$RH = a_w * 100\%$$
 (2.1)



Table 2-5: Source: http://www.foodscience.afisc.csiro.au/water\_fs.htm

In practice, it is generally not possible for the growth of fungi to the limit of 65% up to 70%; and this level is reached in all constructions in all insulation materials most of the year. As RH increases more species of fungi are able to grow and growth becomes more rapid. At given moisture content, the availability of the water to fungi may be different for different materials and constructions. Because of these differences it is difficult to design a standard solution, which will overcome all the problems of moisture and fungal growth.

Because all the influencing factors are not always in ideal circumstances, Building Decree (Bouwbesluit), in the Netherlands, has declared a maximum monthly marginal value of 80% based on ISO 13788 [58]. For shorter duration of the relative humidity, than a higher value has been permitted. This value is confirmed by an experiment [30] with the two common indoor fungi. See also Figure 2-2. The curves connect the points of equal growth rate per day (mm/day).



Figure 2-2: Growth of Penicillium (a) and Aspergillus (b), Source: Lit: 30

#### The Effects of Thermal Bridges on Fungi Growth

In building practice, fungal growth is associated with thermal bridges. The requirements of a minimum thermal quality of the building are adopted to reduce the risks of fungal growth (Anon., 1991a). The thermal quality of a cross-section of the building is expressed by the temperature ratio or temperature factor  $\mathbf{f}$ :

$$f = \frac{T_{i,sur} - T_{e,air}}{T_{i,air} - T_{e,air}}$$
(2.2)

 $T_{i,sur}$  = interior surface temperature  $T_{i,air}$  = indoor reference temperature  $T_{e,air}$  = outdoor reference temperature

Just like for condensation, fungi growth is not considered or hardly possible on a construction with a temperature factor of above 0.65 for houses.

Weather conditions affect indoor temperatures and humidity. Although it is suggested that the indoor fungal problems occur in autumn and spring, the simulations of Van den Bosch and Smolders (1994) have shown that in general the highest risks for the growth of fungi is the winter season, especially the month January. The simulations about the daily temperature fluctuations were also done by Van den Bosch and Smolders (1994). The growth of fungi deviated by 10% from the seasonal effects.



Figure 2-3: integrated growth as a function of the month of the year for the hydrophilic (a) and xerophilic fungi; Source: Lit: 30. In both figures, a thermal bridge is used (k) with f=0.45 and f=0.55 connected to a cavity wall (f=0.84).



### 2.2.4 Discussions

Because of the nearly equal properties of insulation materials, the attention is paid nowadays to the fire resistance of the materials; and the problems due to occurrence of moisture in the structure (interstitial condensation and Indoor Air Quality), and how to avoid these problems using materials available.

The absorption of detrimental gases does not form the main purpose of the thesis, and therefore not analyzed detailed in this study. However, the effect of wool insulation on the quality of the indoor environment depends mainly on the position of the insulation material in the construction. This property can be effective when the insulation material is directly in contact with the indoor environment; but when positioned between the inner and outer leaf of the construction, the significance of the transmission of the gases through wall is only relative to ventilation<sup>10</sup>.

As nitrogen containing material, wool releases also HCN when exposed to fire; furthermore HCN is the major toxicant for wool. Although HCN is more toxicant than CO, when toxicant gases are released during fire, the LT50 value for wool is found to be lone of the largest (23 minutes); which indicates that wool has less danger than, for example, nylon (LT50 is 7 minutes).

<sup>&</sup>lt;sup>10</sup> One should not forget that the indoor air gases are larger than water molecules, and therefore more difficult to diffuse in the wall.



# 3 Moisture Transport

This chapter is about the theory and the mechanisms of moisture flux in porous materials.

# 3.1 Moist Air

Air consists of many gases including water vapor, and each gas has its own partial pressure in total air pressure. The relation between the water vapor content in the air and the total air pressure can be approximated with the Ideal Gas Law. The partial pressure of water vapor is denoted as  $p_v$  (Pa) and the relation is given as:

$$p_{v} = \frac{m \cdot R_{v} \cdot T}{V} \Leftrightarrow p_{v} = v \cdot R_{v} \cdot T$$
(3.1)

*V* is the volume of the space (m<sup>3</sup>), the *m* (kg) is the vapor content in the volume *V*, and *v* (kg/m<sup>3</sup>) is the called as vapor content.  $R_v$  is the gas constant for water vapor (462 J/kgK) and T is the temperature in Kelvin.

Air can contain a maximum amount of water vapor (humidity by volume) due to liquid-gas water. equilibrium for This amount of water vapor is temperature dependent and increases exponentially as the temperature increases. Figure 3-1 shows the relation between the maximum possible water vapor content  $V_s$  (in the vertical axis as  $g/m^3$ ) and the temperature T (in the horizontal axis) between -20  ${}^{0}C$  and 30  ${}^{0}C$ .



Figure 3-1: Max. Moisture content of air at saturation

The ratio between the amount of water vapor in the air at a certain temperature and the maximum amount of water vapor that air can contain at the same temperature is defined as the Relative Humidity ( $\phi$ , RH, Rel. Hum.). The Relative Humidity is given usually in percentages and shown as:

$$\phi = \frac{v}{v_s} \cdot 100\% \tag{3.2}$$



### **3.2 Moisture sorption in Porous Materials**

Solid surfaces have the tendency to capture and localize the moisture on them. This phenomenon is called *adsorption*. The amount of localized water molecules depend on the relative humidity, the temperature and the surface area. The dependence on the temperature is usually neglected because the influence is relatively smaller than the other two. The moisture content w of the material in equilibrium with the environment (air-filled pore) at given relative humidity is usually written as  $w(\phi)$  in (kg/m<sup>3</sup>) or (g/m<sup>3</sup>).



The moisture ratio of a material can be written as:

$$u = \frac{w(\phi)}{\rho_{dry}} \cdot 100\% \tag{3.3}$$

Figure 3-2: example of a porous material in humid area

 $\rho_{dry}$ : Dry density of the material (dried in an environment with the temperature of 105  $^{0}$ C)

If the environment is absolutely dry, the amount of adsorbed water is zero. As the relative humidity, or moisture content, increases, the water molecules are adsorbed to the material, first in the form of a monomolecular layer and than in multi-molecular layers. In this stage, there will be equilibrium between the moisture content of the porous material and the relative humidity (in the range of 0-98% RH). This equilibrium curve is called the sorption isotherm of the material. The state from absolute dry to certain moisture content at 98% RH is called the hygroscopic range of the material.

If the material is in contact with liquid water, a large amount of water molecules can be fixed to the pore structure. Maximum amount of moisture content of a material that is in contact with water is called capillary moisture content ( $w_{cap}$  [kg/m<sup>3</sup>]). The maximum possible water content of a material called saturated moisture content ( $w_{sat}$  [kg/m<sup>3</sup>]).



Figure 3-3: Principle figure of the Storage mechanism in a porous material



### **3.2.1** Types of Adsorption

Adsorption is a surface phenomenon where the molecules of a gas (water vapor) or liquid is taken up and spread on the surface of the material including pores. Adsorption is an exothermic process; and because of its exothermic nature, the amount of adsorbed gas decreases as the temperature of the system increases. Also high vacuum reduces the pressure of the gas and reduces the adsorption. Adsorption is usually divided in to two classes; namely physical adsorption and chemisorption.

Physical adsorption is very rapid and reversible. It can occur on almost all solid surfaces except under high temperature and vacuum conditions. high Physical adsorption occurs usually as a multi layer process. The available surface area does not limit the amount of vapor that can be adsorbed on a surface but molecules can lie on each other on several layers. The first layer can be either physically or chemically adsorbed, but the subsequent layers are adsorbed physically. The kinetics of physical adsorption is assumed basically to



Figure 3-4: Multilayer adsorption on a solid surface

be dominated by vapor diffusion (occurs as rapid as the molecules arrives at the surface). The process is reversible and the equilibrium is attained quickly, as the molecule arrives to the surface.

On the contrary, chemisorption is slower than physical adsorption. Because chemisorption needs high activation energy, it is expected to be only as a monolayer. Due to the high activation energy, chemisorption may not be reversible; or is only reversible at high temperatures.

The moisture sorption properties of wool are further analyzed in Chapter 4.

### 3.2.2 Adsorption Isotherms Classification

Adsorption is usually described as a function of temperature and partial pressure (n = f(P,T)), where n is the amount of absorbate). The general relationship between the amounts of gas adsorbed by a solid at a constant temperature as a function of gas pressure is called adsorption isotherm. This relationship is temperature depended, but because it is written as a function of the relative humidity and measured at constant temperatures, the temperature dependence is neglected. Gas pressure can also be related to the relative humidity as done in this study.

Brauner, Emmet and Teller have defined five types of sorption isotherms to be able to explain the relation between the amount of adsorbed molecules and the vapor pressure of the environment, which is also called as "BET-isotherm" (See Figure 3-5). These five types of BET-isotherms are valid for the molecules which form multilayer on a solid.





Figure 3-5: Shapes of the five principal types of the BET-isotherms, Literature 29

The sorption isotherms can be divided into three regions; namely monolayer, multilayer and capillary condensation, see Figure 3-6.. The boundaries of these regions can differ from one material to another.



Figure 3-6: Typical Adsorption- Desorption isotherms showing Hysteresis, Lit: 31

Because it is experimentally difficult to maintain very low relative humidity and also relative humidity conditions above 98% RH, the sorption curve stops at 98% RH.



By any change in the relative humidity, the material exchanges water with the ambient air until equilibrium is reached. The water exchange per relative humidity step is called the specific moisture capacity of the material.

If a saturated porous material is allowed to dry, it does not follow the adsorption isotherm. Usually it contains more moisture at any given relative humidity than the adsorption isotherm. This reverse process of adsorption is called desorption and the difference between adsorption and desorption is defined as hysteresis.

### **3.3 Transport Mechanisms**

The transport of mass (water vapor) within materials can occur either by diffusion or convection.

### 3.3.1 Diffusion

Gas molecules move in a closed system to reach the equilibrium. Water (vapor) moves in porous materials under influence of water (vapor) pressure difference or concentration difference. The direction of the movement is always to the side of lower concentration or pressure. A general expression for the moisture flow can be written as the product of a transport property and the gradient of the driving force (potential).

*Flux* = *Transportproperty* × *gradient*(*drivingforce*)

This potential can be chosen in different forms  $(p, v, \varphi, u, w)$ . Moisture content of the material (w) and the partial pressure (p) of the fluid are the most common potentials in the literature.

### **3.3.2 Moisture Transport in Air**

The diffusive flux g  $(kg/m^2s)$  in air in steady-state conditions is as follows according to Fick's law:

$$g = -D\frac{dv}{dx} \tag{3.4}$$

*D*: diffusion coefficient of water vapor in air v: vapor concentration in air [kg/m<sup>3</sup>]

The diffusion coefficient *D* is temperature dependent:

$$D = (22.2 + 0.14 \cdot T) \cdot 10^{-6} [\text{m}^2/\text{s}], \text{T} [^{0}\text{C}] \qquad (3.5)$$



Figure 3-7: Diffusion of water vapor in air



### 3.3.3 Convection

Convection involves the collective movements of the molecules. It, therefore, is relevant for fluids. During convective flow the fluid moves with an average velocity.

 $Flux[molecules / m^2s] = velocity[m / s] \times concentration[molecules / m^3]$ 

The velocity is a physical quantity of fluid's motion. Velocity of a fluid simply means the total speed of the fluid molecule in a particular direction. Analyzing the mass transport by convection requires analyzing the flow characteristics of the fluids. The convective flow of water vapor, although sometimes important in building construction, is however not considered in this study.

# **3.4 Moisture Transport in Porous Materials**

The moisture flows are caused, in general terms, by diffusion and convection of water vapor; and by capillary action. At low relative humidity, if air convection is not considered, water vapor diffusion dominates the transport. At higher relative humidity conditions, the moisture transport becomes a combination of diffusion and capillary action. Capillary action dominates when the relative humidity increases close to 100%. This research focuses only on the diffusive water vapor transport.

## **3.4.1 Description of Diffusive Transport**

Fick (1855) has stated that there is a linear relation between the rate of the diffusion and the local concentration of the gases (which is later also called as Fick's First Law). Fick's first law for the diffusion of water vapor through a porous material is written as:

$$g = -\delta_{\nu} \frac{\partial \nu}{\partial x}$$
(3.6)

 $\delta_{v}$ : diffusion coefficient of the material [m<sup>2</sup>/s] *v*: vapor concentration in air [kg/m<sup>3</sup>]

The driving potential for the diffusion can be characterized with any of the variables "p, v,  $\varphi$ , u, w" because of the relations with the moisture, as mentioned in chapter 3.3.1.

### **Diffusion Coefficient Forms**

The diffusion coefficient differs also according to the chosen driving potential.

When the partial pressure is chosen as the driving force; the equation becomes as:

$$g = -\delta_p \cdot \frac{\partial p}{\partial x} \quad [\text{kg/sm}^2]$$
(3.7)

 $\delta_p$  : the water vapor permeability [kg/s.m.Pa]

p: the partial vapor pressure [Pa]

And if the driving force is chosen as the moisture content (w) of the material, the equation becomes as:

$$g_w = -D_w \cdot \frac{\partial w}{\partial x} \tag{3.8}$$

 $D_w$ : water vapor diffusion coefficient with regard to moisture content *w w*: moisture content of the material [kg/m<sup>3</sup>]

All the types of diffusion coefficients are related and can be converted to each other. For example; the relation between  $D_w$  and  $\delta_p$  for constant temperature can be derivated as:

$$g = -\delta_p \cdot \frac{\partial p}{\partial x} = -D_w \cdot \frac{\partial w}{\partial x}$$
(3.9)

$$D_{w} = \delta_{p} \frac{\partial p}{\partial w} = \delta_{p} \cdot p_{sat} \cdot \frac{\partial \phi}{\partial w}$$
(3.10)

As explained in Chapter 3.2.2, any change in the relative humidity of the environment  $(\Delta \varphi)$  causes a change in the moisture content  $(\Delta w)$ . The term  $\frac{dw}{d\phi}$  is called the differential moisture capacity of the material, corresponding to the slope of the isotherm, and is denoted here by the symbol  $\xi$ . With the introduction of the moisture capacity, we may write:

$$D_{w} = \frac{\delta_{p} \cdot p_{s}}{\frac{\partial w}{\partial \phi}} = \delta_{p} \cdot p_{s} \cdot \frac{1}{\xi}$$
(3.11)

Similarly, the relation between  $\delta_p$  and  $\delta_v$  is given by

$$-\delta_{p} \cdot \frac{\partial p}{\partial x} = -\delta_{v} \cdot \frac{\partial v}{\partial x}$$
(3.12)

$$\delta_p = \delta_v \cdot \frac{\partial v}{\partial p} = \frac{\delta_v}{R_v \cdot T}$$
(3.13)

Where;  $R_v$  is the gas constant of water.

#### Vapor Resistance Factor

To compare the rate of the diffusion through a material ( $\delta_v$ ), with the diffusion coefficient of water vapor in air (*D*), Kirscher has introduced "vapor resistance factor" ( $\mu$ -factor) for materials:

$$\mu = \frac{D}{\delta_{\nu}} \left[ - \right] \tag{3.14}$$

*μ*: Vapor Resistance Factor [-]

The  $\mu$ -factor of a material indicates "how many times less" a material transports water vapor by diffusion than a still air layer. The  $\mu$ -factor of stagnant air equals 1.





Fick's first law can now be written as:

$$g = -\frac{D}{\mu}\frac{\partial v}{\partial x}$$
(3.15)

Because  $\mu$ -factor regards the transport of moisture through the material, the pore structure in the material and the dimensions of the material influence the value of the  $\mu$ -factor. A highly porous material with a closed pore structure can have a larger  $\mu$ -factor than a less porous material with an open structure.

The  $\mu$ -factor depends also on the moisture content of the material. De Wit [44] states that higher moisture content leads to lower  $\mu$ -factor; because of the shortening of the diffusion path by water islands in the material at higher moisture contents.

### 3.4.2 Capillary Suction

The transport of water becomes a combination of the slow diffusion process and rapid capillary suction at larger relative humidity conditions (between 95% RH and 100% RH). At these points, there cannot be made any strict separation between the diffusion and capillary suction. Therefore, another term (*the critical moisture content*) is defined. The diffusion governs the transport phenomena below the critical moisture content; and capillary suction above the critical moisture content.

Capillary suction is the attraction between the water molecules and the pore walls. If the radius of the pore is very small (capillary), the water in the pore is sucked upwards and the water rises up until the balance is reached with the gravitational forces.

Capillary suction is not taken into account in this study, because this study focuses only the transport within the hygroscopic region.

Delft

### 3.5 Moisture Balance of a Material

Fick's second law is used in non-steady state diffusion. The one-dimensional moisture balance equation for (Fickian) diffusive transport is written as:

$$-\frac{\partial g}{\partial x} = \frac{\partial w}{\partial t}$$
(3.16)

The right hand side of the equation can be written using the chain rule as:

$$\frac{\partial w}{\partial t} = \frac{\partial w}{\partial \phi} \frac{\partial \phi}{\partial v} \frac{\partial v}{\partial t} \Longrightarrow$$
(3.17)

$$\frac{\partial w}{\partial t} = \frac{\xi(\phi)}{v_s(T)} \frac{\partial v}{\partial t}$$
(3.18)

The left hand side of the equation can be written as:

$$-\frac{\partial g}{\partial x} = -\frac{\partial}{\partial x} \left( -\delta_{v} \frac{\partial v}{\partial x} \right)$$
(3.19)

The moisture balance equation, for constant temperature, can be written as:

$$\frac{\partial}{\partial x} \left( \delta_{v} \frac{\partial v}{\partial x} \right) = \frac{\xi(\phi)}{v_{s}(T)} \frac{\partial v}{\partial t} \Leftrightarrow \frac{\partial}{\partial x} \left( a_{v} \frac{\partial v}{\partial x} \right) = \frac{\partial v}{\partial t}$$
(Diffusion equation) (3.20)

$$a_{\nu} = \frac{\delta_{\nu} \cdot v_s(T)}{\xi(\phi)} \, [\text{m}^2/\text{s}], \ a_{\nu}: \text{ moisture diffusivity parameter}$$
(3.21)

### 3.5.1 Moisture Balance Using other Potentials

The moisture balance of the materials can also be written by using the potentials p,  $\varphi$ , w. In this case, the right hand side of the equation become for p according to the chain rule as:

$$\frac{\partial w}{\partial t} = \frac{\partial w}{\partial \phi} \frac{\partial \phi}{\partial p} \frac{\partial p}{\partial t} \Longrightarrow \frac{\partial w}{\partial t} = \frac{\xi(\phi)}{P_s(T)} \frac{\partial p}{\partial t}$$
(3.22)

And the left-hand side of the equation;

$$-\frac{\partial g}{\partial x} = -\frac{\partial}{\partial x} \left( -\delta_p \frac{\partial p}{\partial x} \right)$$
(3.23)

Combination of the left- and the right hand side of the equation is as follows;





$$\frac{\partial}{\partial x} \left( \delta_p \frac{\partial p}{\partial x} \right) = \frac{\xi(\phi)}{p_s(T)} \frac{\partial p}{\partial t} \Leftrightarrow \frac{\partial}{\partial x} \left( \frac{\delta_v p_s(T)}{R_v T \xi(\phi)} \frac{\partial p}{\partial x} \right) = \frac{\partial p}{\partial t} \Leftrightarrow \frac{\partial}{\partial x} \left( a_v \frac{\partial p}{\partial x} \right) = \frac{\partial p}{\partial t}$$
(3.24)

Where 
$$a_v = \left(\frac{\delta_v p_s(T)}{R_v T \xi(\phi)}\right)$$
 (3.25)

When the equations (3.21) and (3.25) are compared, one can see that the diffusion coefficient  $a_v$  is independent of the choice of potential.



# 4 Hygroscopic Properties of Wool

Wool is an organic complex material that is composed of cells, surrounded by a cell membrane complex. Wool fibers exhibit hygroscopic behavior, which allows them to absorb and adsorb high amount of moisture (up to 33-34% (kg/kg)).

# 4.1 Diffusion through Wool Bulk

As in all porous materials the diffusion is mainly dependent on the porosity of the material. Padfield [42] explains the movement of water vapor through fibrous, porous materials in three forms (Figure 4-1):

- A: Diffusion through the air channels
- B: Absorption in to the fibers
- C: Movement in the material

He states that the dominant mechanism for the transport of water is the movement through the wool air channels (process A). Diffusion in to the wool fiber (Process B) is actually the absorption into the wool fiber, and occurs only in the beginning stage; until the material reaches the equilibrium with the RH. Diffusion within the fiber (Process C) has the least effect on the movement of water vapor through the material itself.



Figure 4-1: Water transport in Wool, Lit: 42



Figure 4-2: Picture of a Wool fiber, Lit: 36

There could no information be found about the contribution of process B on process C. Padfield [42] states again that the movement in the fiber (Process C) can become dominant in paper fiber insulation, wood shavings, or cellular cement; but he has given no information about this process in wool fiber. Furthermore, he states the impossibility of separating processes A and C.

There could also <u>not much information be found</u> about the transport of water (vapor) in the wool fiber. Because it can absorb much water, the process C can also be significant in sheep wool. Because of the lack of this information in the literature, the processes A and C in Figure 4-1 are assumed as one diffusion form and called as *sorption*.



## 4.2 Sorption

The sorption and desorption of moisture under the influence of relative humidity is one of the fundamental properties of wool fiber (d'Arcy, Watt, 1979). Downes and Mackay [43] describe adsorption<sup>11</sup> into the wool fiber in two stages; the first stage happens in a few minutes and the second stage many hours at room temperature. The uptake of moisture during the first stage of absorption occurs by a Fickian mechanism with a concentration-dependent diffusion coefficient; but the second stage<sup>12</sup> does not occur by a diffusion-controlled mechanism. Therefore, no generalized form of Fick's diffusion equation is sufficient to describe the whole course of the sorption in the wool fiber [50]. It is known that absorbed water causes swelling of the fiber. As swelling causes more available surface area for the adsorption, the swelling and the total sorption of the fiber is interdependent.

There is no information available in the literatures about chemisorption in wool. But because of multilayer which occur on wool fiber, the speed of sorption and, fast reversible sorption (adsorption and desorption) which occurs one after another; it can be assumed that chemisorption doesn't occur in sheep wool. Therefore, chemisorption is not studied and not taken in to account in this research

Speakman et. al. [7] is one of the first scientists who have studied water absorption properties of wool. They distinguish the moistening process of wool into three stages. The results of their studies based on at 105 <sup>0</sup>C dried wool are;

- 1. Water absorption up to 5% of the weight of dry the fiber: This water has been strongly bound.
- 2. Water absorption of 5-26%: The adsorption goes approximately proportionally with the humidity percentage.
- 3. Water absorption of 26-34%. The adsorption approaches to a constant value. The fiber saturates at 33-34%. [7]

The absorption uptake curves are also studied by Watt et. al. (1960) [49]. They distinguish moisture uptake into three categories according to the concentration of water in the wool fibers, which also corresponds to the studies of Speakman.

Water vapor adsorption on wool fits the BET isotherm [35, 36, and 38]. The adsorbed water forms a continuous and very ordered bulk liquid phase as multilayer which is explained by Miyagawa (1987) and by Sen'i Gakkaishi et. al. (1989) as the reason of high adsorption percentage.

<sup>&</sup>lt;sup>11</sup> Usually it takes more than two weeks until moisture sorption equilibrium is reached [33].

<sup>&</sup>lt;sup>12</sup> The slow increase in concentration during the second stage is thought to result from the swelling of the wool fiber.



Figure 4-3 Water ad-and de-sorption curve, Source: Doschawol

### 4.2.1 Temperature Dependency of Sorption Isotherm

The water adsorption process is an exothermic process over the whole range of adsorption process [37]. The heat of adsorption of water vapor is dependent on the moisture content of the wool.

It has also been shown that the sorption process of sheep wool is temperature dependent. Gakkaishi (et. al 1989) has measured that the higher the temperature, the less amount of adsorbed water in equilibrium at any given relative humidity [35].

Wortmann (et al. 2000) has measured the sorption properties of wool at different temperatures. The sorption decreases slightly as the temperature increases (from 34% to 29%). By analyzing Figure 4-4, it can be concluded that the temperature in the range applicable in buildings has not much effect on the sorption properties of sheep wool.



Figure 4-4: Sorption isotherms (kg/kg) at different temperatures as line plots, Lit. 37




# 5 Experiments

Three separate experiments were done to better understand how wool behaves in humid environment; namely, the measurement of the  $\mu$ -factor, the measurement of the sorption isotherm and the measurement of the diffusion process of water vapor in wool. The sorption and permeability tests are well known, but the diffusion experiment is quite unique. The test material (100% wool) which is obtained from Doschawol, in the Netherlands, is used in the experiments.

# 5.1 The Climate Chamber

All the three experiments were carried out simultaneously in a climate chamber (*Votsch Industrietechnik VC4100*). The experiment space in the climate-chamber has a cubic form with a volume of about  $1m^3$ . The relative humidity in the chamber can be set between 10% and 98% within +10  $^{0}$ C and +95  $^{0}$ C temperature range. In order to get a constant and homogeneous humidity level, the air in the chamber is well mixed by four wall-mounted ventilators. The standard working temperature and relative humidity range is given in the Picture 5-1.



Picture 5-1: Climate Chamber

According to the manufacturer, the relative humidity fluctuation is < 3%. The chamber was tested for 18 h before starting the experiments and it has been seen during the test period that the chamber can adapt the temperature and relative humidity changes easily during increasing the relative humidity of the environment (Figure 5-2).





Figure 5-2: Testing the RH and the temperature of the climate chamber before the experiment (The actual temperature (dark blue) follows the calibrated temperature (red line). The actual RH (brown line) follows the calibrated RH (green) during increasing, but deviates somewhat during decreasing the RH)

#### 5.1.1 Measurement Environment

#### **Relative Humidity Range**

The RH in the chamber during the experiments was increased stepwise to be able to measure the diffusion process of water vapor in wool between two constant RH conditions.

Before starting the experiments, the samples were conditioned for one day in the chamber at 20  $^{0}$ C and 20% RH. The next day the relative humidity of the chamber was raised to 45% (the start point of the experiments) and the samples were conditioned for one more day before placing them in the set ups in the climate chamber.

The experiments in the chamber started at 45% RH followed by consecutive increment steps of 15% until 90% RH. Decisive for the duration of each step was the diffusion experiment a next step change of the relative humidity was imposed only when the relative humidity at the bottom of the cylinder had reached at least half of its final value.

#### Ventilation

There is strong ventilation flow in the test chamber. For this reason, the open side of the glass tube (see Picture 5-2) was covered with a -damp open- nylon cover to minimize convective flow in the wool at the open side of the column. Furthermore, the wet up for the measurement of the sorption isotherm (see Picture 5-4) was protected by plates from five sides in order to minimize the influence of the turbulent air flow on the weight measurements.



Picture 5-2: nylon cover at the open side of the glass tube



Picture 5-3: Ventilator in the chamber



**Picture 5-4: plates for the balance** 

#### Temperature

Because only the behavior in humid environment is analyzed, the experiments are done at a fixed temperature, which is 20  $^{0}$ C. The temperature has fluctuated between 20  $^{0}$ C and 20,50  $^{0}$ C during the experiment; It is also assumed that the small fluctuations in temperature did not influence the experiments.

#### Data acquisition

The relative humidity sensors in the glass tube for the diffusion process and the two balances (one for measuring the sorption isotherm and one for measuring the  $\mu$ -factor) are connected via cables to a device and than to the computer. With a software program - called LabVIEW<sup>13</sup>- the date, time, RH and temperature values of the data can be read and saved every minute by the computer.

# **5.2 Experimental Set-ups**

### 5.2.1 The Sorption Isotherm

There is much information in the literature about measuring the sorption isotherms of hygroscopic materials. Determination of hygroscopic sorption of building materials is standardized by ISO (International Organization for Standardization) under the code of ISO 12571:2000(E), (**ISO 12571**<sup>14</sup>: Hygrothermal performance of building materials and products-Determination of hygroscopic sorption properties).

This standard specifies two alternative methods for determining hygroscopic sorption properties of porous building materials and products:

- Using desiccators and weighing cups (desiccator method);
- Using a climatic chamber (climatic chamber method)

The moisture content at a constant temperature is determined when equilibrium with each environment is reached. Equilibrium with the test environment is established by weighing the specimen until constant mass is reached. Constant mass is reached if the change of the mass between three weighing, each made at least 24h apart, is less than 0.1% of the total mass. After establishing the moisture content at each relative humidity, the adsorption curve can be drawn.

For the adsorption curve, the mean of the calculated moisture contents for the various specimens at each relative humidity are taken in to account. After calculation of the mean moisture content of the specimen at each relative humidity, the adsorption and curve can be drawn by connecting the data points with straight lines.

<sup>&</sup>lt;sup>13</sup> LabVIEW is a graphical software program for creating flexible and scalable design, control, and test applications.

<sup>&</sup>lt;sup>14</sup> International Standard ISO 12571 is prepared by the European Committee for Standardization (CEN) in collaboration with ISO Technical committee TC163, in accordance with the agreement on technical cooperation between ISO and CEN.

The standard doesn't specify the method for sampling. For that reason, a cylindrical prepared wool specimen (D=18cm, h=5cm) has been cut from the middle of the specimen, and than sliced -for the experiment- in to three pieces to reduce the time to reach the equilibrium (more available surface area which is in contact with the environment).

The sliced wool specimens were placed on a mass balance with a metal mesh in between. The moisture content of the specimen could be followed from the balance that is connected to the computer. (See Picture 5-5)



Picture 5-5: Wool specimen and the roster in between and the balance

### 5.2.2 Water Permeability

Traditional cup method is commonly used to determine the water permeability of the building materials (wood, flooring and board materials). In this test method, a specimen of known area and thickness separates two spaces differing in the relative humidity. Then the rate of vapor transport across the specimen at a steady state is determined gravimetrically. From these data an average value for the water permeability of the specimen can be Figure 5-3: Traditional cup method calculated.



An aluminum cup (D=30cm, height = 17cm) and Calcium Nitrate Tetrahydrate (Ca(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O) saturated salt solution were used for measuring the permeability of the wool specimen. Saturated Calcium nitrate tetrahydrate solution creates 56% relative humidity at 18 <sup>0</sup>C in its environment according to the literature. A before hand calibrated escort sensor (denoted as TU1) is used to measure the relative humidity in the aluminum cup during the experiment. The escort sensor saves the data every 15 minutes and the data can be read only after the experiments end.



Picture 5-6: Schematic Drawing of the set-up, Salt (Ca (NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O), Aluminum Cup and the Sensor



The prepared salt solution was placed in a plastic cup (to prevent chemical reaction with aluminum), and this plastic cup was placed in the middle of the aluminum cup, with the escort sensor in another cup in the salt solution. To keep the wool specimen stable in the aluminum cup, a raster was placed above these plastic cups. (See Picture 5-7)



Picture 5-7: The set up of the water permeability experiment is shown step wise

The conditioned wool specimen (thickness= 10cm) was placed in the aluminum cup and the aluminum cup was put on a balance to measure the changes of the weight during the experiment.

### 5.2.3 Diffusion Experiment

A glass tube (diameter = 18cm and length = 61 cm) was designed with 7 holes on certain points on it for insertion of relative humidity and temperature sensors. The holes were exactly on the points of 0, 30, 40, 50, 55 and 60cm from the closed side of the glass tube and the seventh hole is on the same point with the one on 50cm but with a  $90^{0}$  angle.



Figure 5-4: Schematic Drawing, Dimensions and the Glass Tube

Eight sensors were used during the diffusion experiment (seven (so called RV1, RV2, ..., RV7) in the glass tube and one (RV8) out of the glass tube (in the chamber)). This last sensor (RV8) was placed in the chamber to get more reliable data about the relative humidity of the chamber during the whole experiment period.



Picture 5-8: The Sensors which are connected to the computer



The relative humidity of the room (where the climate chamber is placed) was also set to 45% RH in order to minimize the influence of the outside environment. The glass tube was filled with 12 wool specimens (thickness 5 cm, diameter 18 cm, conditioned at 45% relative humidity) and its open side was covered with a very open nylon cover during the experiment in order to minimize the effect of the ventilation airflow in the chamber on the transport in the wool.



Picture 5-9: Preparation of the diffusion experiments in three steps



# 6 Results

The experimental data is analyzed in this chapter.

# 6.1 Experimental conditions

The first eight days of the experiment were used for testing, controlling and reconditioning of the materials and specimens. The gathering of data from the first part of the experiment has started on the 9<sup>th</sup> day of the total experimental period. Therefore, the first 8 days of the experiments are not taken into account while analyzing the experiments.

Between day 22-25 and 77-78 there was an electricity interruption on the computer. The chamber continued working during this interruption, but the computer had stopped the data acquisition. It may be assumed that these interruptions didn't affect the course of the experiment. In addition, on day 42 the experiment was disturbed for a short time. Because of this short period, this distortion has no effect on the outcome of the experiment. Furthermore, it has also been seen during the experiment (Figure 6-1) that it is very difficult to keep the chamber at constant RH at high RH conditions.

# 6.2 Sorption Isotherm

The relative humidity and sorption data of the experiment is shown in Figure 6-1. The sorption of water vapor can be analyzed in two stages; there is a fast sorption in less than one day (first stage) and a second stage until it reaches the equilibrium.



# 6.2.1 Measurement

The data for each RH step concern the equilibrium water content. Because wool reaches equilibrium in more than two weeks (the fast first stage and the slow second stage; see Figure 6-1); the average of the sorption between  $16^{th}$  and  $19^{th}$  days were chosen as reference data to measure the sorption isotherm. These reference data are given in Table 6-1:



	Regain (gr)
RH (%)	(16-19)days
43,77	0,00
60,46	0,79
76,63	1,98
95,21	4,22

Table 6-1 Regain data for wool at different RH steps

### 6.2.2 Parameterization

The equation for the sorption isotherm of hygroscopic materials as defined by Hansen (1986) was used for parameterization:

$$w = w_{sat} \left( 1 - \frac{\ln(\phi)}{A} \right)^{\left(-\frac{1}{n}\right)} A, n, w_{sat}: \text{material regression parameters}$$
(6.1)

where w is the amount of water sorption  $(kg/m^3)$  and  $\varphi$  is the relative humidity. The parameters (A, n and  $w_{sat}$ ) are regression parameters which are specific for each hygroscopic material.

A program was written in MATLAB<sup>15</sup> to determine the regression parameters. The program and the results are shown in APPENDIX II.3. The coefficients according to the formula of Hansen as results of the calculation with MATLAB are as follows:

#### <u>Sorption Parameters</u> <u>According to the formula of Hansen:</u>

$$w_{sat} = 6.3072 \text{ (kg/m}^3)$$
  
 $A = 0.2655$   
 $n = 1.3634$ 

So, the formula of the adsorption isotherm for the sheep wool reads:

$$w = 6,3072 \left( 1 - \frac{\ln(\phi)}{0,2655} \right)^{\left(-\frac{1}{1,3634}\right)}$$
(6.2)

which is depicted in Figure 6-2.



<sup>&</sup>lt;sup>15</sup> MATLAB is data-manipulation software package that allows analyzing and visualizing data using existing functions and user-designed programs.



Figure 6-2: Total Sorption diagram in the range of 0-100% (a); and in the range of 40-100% (b)

Figure 6-2 (a) and (b) can be found on a bigger format in APPENDIX II.3.



# 6.2.3 The Hygroscopic Moisture Capacity $(\xi)$

It is noted in previous chapters that the hygroscopic moisture capacity is one of the essential properties of a hygroscopic material which determines the diffusion of water vapor in a porous material. The hygroscopic moisture capacity is defined as the slope of the sorption isotherm Therefore, the derivative  $(dw/d\varphi)$  of equation (6.2) gives the hygroscopic moisture capacity ( $\xi$ ) for the adsorption phase.

The equation of hygroscopic moisture capacity for **adsorption** curve can be written as in equation(6.3):

$$\xi = \frac{\partial w}{\partial \varphi} = 17,42404 \cdot \frac{1}{\varphi} \cdot \left(1 - \frac{\ln \varphi}{0,2655}\right)^{-1.73346} [0,40 \le \phi \le 1]$$
(6.3)

The graph of the hygroscopic moisture capacity can be drawn between the ranges of 40-100% RH.



Figure 6-3: Hygroscopic Moisture Capacity

The moisture capacity ( $\xi$ ) is analyzed for each RH step and the graph is drawn in the same range to be able to calculate the  $\mu$ -factor at the diffusion experiment.



Figure 6-4: ξ for each RH step



### 6.2.4 Discussion

Because the total amount of sorption is important in the experiment, the terms adsorption and absorption are combined together in this research. The averages of "three days of regain on selected certain days (16-19days)" at each RH step are used to draw the sorption graph. The sorption occurs in two stages within the experiment range. The first stage is fast, and occurs in couple of hours; and the second stage is very slow and it takes days to reach the equilibrium. The second stage of the sorption is not visible (or not visible) at 45-60% RH range, but it can be seen more clearly in the range of 75-90% RH (See Figure 6-1 and Appendix II.2). Analyzing the sorption behavior of sheep wool in lower relative humidity's will help to understand and interpret the sorption behavior. It is very early at this point to comment about the reasons of the second stage, but this slow second stage of the adsorption can be due to swelling of wool with increasing relative humidity. In fact, Watt [51] points out the structural changes within the wool-water complex during sorption.



# 6.3 Water Vapor Permeability

The results of the experiment had to be compared with a 2D-cylindrical finite-element calculation because the salt solution did not cover the whole bottom surface of the aluminium cup.

As mentioned before, we used one sensor in the aluminum cup just under wool specimen (TU1), and one just on the wool specimen (TU2) to measure the relative humidity in- and outside of the aluminum cup during the experiment. There is also one sensor in the chamber (RV8). A balance is used to measure the weight increase of the set up.

# 6.3.1 Measurement

The RH of the environment was increased stepwise and the resistance of the wool specimen against the diffusion of the water vapor was analyzed as explained in the theory. The relative humidity during the RH steps in the chamber and the corresponding weight increase are given in Figure 6-5.

The analysis of the permeability experiment at RH steps has started about two to five days after each relative humidity step because of

two sorption stages of wool (fast and slow) and each RH step on this experiment was analyzed apart from each other.



Figure 6-5: Weight increase per RH step

The rate of the vapor diffusion from one side of the aluminum cup to the other was used in the analysis of the permeability. The rate of the diffusion ("amount of diffused water vapor (gr/day) per RH step per day" from one side of the aluminum cup to the other) was found by the determining the slopes of the diffusion rate at each RH step.

The slope of each RH step was found by applying "trend lines" in a linear function form of " $y = a \cdot x + b$ " on the diffusion data. The coefficient 'a' gives us the rate of the diffusion by time.



## 6.3.2 Diffusion Graph

The diffusion rate of water vapor through wool per RH step is shown in Table 6-2 and the graph was drawn (Figure 6-6). The graph shows that the RH in the aluminum cup because of the salt solution is about 57%, which also corresponds with the literature.

Table 6-2: RI	Measured rates per I step
RH (%)	diff. rate (gr/day)
44,15	-1,5053
60,13	0,4708
76,79	2,1218
95,16	3, <mark>6</mark> 98



Figure 6-6: Diffusion Graph for the measured rates

The  $\mu$ -factor for wool was calculated from the diffusion rate per RH difference. The rate per 1% RH difference is calculated and given in Table 6-3.

#### Table 6-3: Diffusion rate per 1% RH difference

RH(%)	diff. rate(gr/day)	rate per RH difference
44,15	-1,5053	
		0,123660826
60,13	0,4708	0.000000.0
76 70	2 1218	0,09909964
10,13	2,1210	0.08580294
95,16	3,698	-,
	1% RH difference	e at 20 'C=>23,4 Pa

The outcome of the experiment needs to be corrected because the salt solution doesn't cover the whole bottom surface area. Comsol Multiphysics<sup>16</sup> was used for modelling the experiment; because the cross-section of the set-up can easily be drawn on cylindrical coordinates and the calculation can be done more accurately.

<sup>&</sup>lt;sup>16</sup> Comsol Multiphysics is a scientific software program for the modeling and simulation of any physicsbased system. For more information:www.comsol.com



### 6.3.3 Model "Comsol Multiphysics 3.1"

Because of cylindrical coordinates, the cross section of the set up was drawn on scale and the calculations were done as a half on the Comsol model (Figure 6-7). This model is based upon 1 Pa pressure difference between the top and the bottom of the wool specimen. The relative humidity changes in the aluminium cup can be seen as isotherms within '1 Pa' difference.



Figure 6-7: Schematic drawing (a), Set up (b), and Model (c)

## 6.3.4 Results

The results of the experiment are converted to same units as in the Comsol model (also Appendix II.5) in order to be able to make a comparison. The three diffusive fluxes for 1 Pa difference at each RH step of the experiment are given in Table 6-4.

Table	6-4:	Diffusive	flux	per	RH	step
	· · ·	211140110		P**		P

Relative Humidity	flux at 1 Pa
0,4415	difference
0,6013	6,11649E-11
0,7679	4,90165E-11
0,9516	4,24397E-11

The model presents the diffusive flux according to chosen  $\mu$ -factor. The diffusive flux in the model for the  $\mu$ -factor (**Kw** value for the model), in the range of 1,2 and 2,1, is analyzed by choosing the  $\mu$ -factor on certain points (See Table 6-5) and compared with the data from the experiment.

Table 6-5: Diffusive I	Flux for the model
according to chosen	U-factor

Kw	Flux ( <b>kg/s</b> ) bij diff. of 1 Pa
1,2	6,11E-11
1,25	5,97E-11
1,3	5,84E-11
1,5	5,35E-11
1,7	4,94E-11
1,9	4,59E-11
2,1	4,29E-11



Figure 6-8: Graph of diffusive flux according to chosen  $\mu$ -factor

When the diffusive flux of the experiment (Table 6-4) and the diffusive flux for the model (Table 6-5) were compared, the  $\mu$ -factor of the setup could be found by interpolation. The interpolated  $\mu$ -factor's on different RH steps for sheep wool and the graph of  $\mu$ -factor is shown at Table 6-6 and Figure 6-9.

or for the RH steps
the Mu-factor(Kw)
1,20
1,72
2,12



Figure 6-9: Graph for the  $\mu$ -factor

# 6.3.5 Discussions

Because the escort sensor in the aluminum cup has stopped earlier than expected, it was not possible to measure the relative humidity inside the aluminum cup. Therefore the diffusion formula can not be accurately applied on the water permeability experiment. But the  $\mu$ -factor of the specimen can be calculated by the rate of the diffusion. Because the salt solution does not cover the whole bottom surface, the outcome of the experiment was compared with a 2D-cylindrical finite-element calculation. The analyzed outcome show that  $\mu$ -factor lies between 1,2 (at 60% RH) and 2,12 (at 95% RH). The  $\mu$ -factor was expected to decrease with the increasing RH, but it has been seen that it has increased. The increase of the  $\mu$ -factor is probably due to dilution of the (top layer of the) salt solution which yields a higher RH inside the aluminum cup than anticipated from the water activity of the saturated. Because of this, the measurement of the permeability at lower RH at the start of the experiment is the most reliable. Therefore, from this experiment the  $\mu$ -factor is most probably about 1,2. The uncertainty value for this result has to be calculated. Because  $\mu$ -factor can not be lower than "1", magnitude of  $\pm 0,2$  for the uncertainty can be assumed as a begin value.



# 6.4 Diffusion of Water Vapor

The diffusion of water vapor in the wool-filled glass tube was analyzed and compared separately for each RH step with a numerical model which was made in MS Excel. With the help of this comparison, the diffusion coefficient of the wool specimen was estimated according to chosen  $\mu$ -factor.



Picture 6-1: RV sensors in the glass tube



Figure 6-10: RV sensors during the Diffusion Experiment

# 6.4.1 Numerical Model in Excel

The use of numerical methods for the solution of diffusion problems has increased in the past years, especially by use of computers. Numerical models use a numerical time-stepping procedure to find the system's behavior over time.

The numerical model used a grid point at every 2,5 cm in space (over the length of the wool column) and a time step of 1 hour. The diffusion coefficient is assumed to change linearly with relative humidity. More information on the diffusion model is given at Appendix II.6.

# 6.4.2 Comparison of the Experiment and the Model

The diffusion equation (3.21) was converted to formula (6.4) in order to use in the analysis.



$$a_{v} = \frac{\delta_{a}}{\mu} \cdot p_{sat} \cdot \frac{1}{\xi(\phi)}$$
(6.4)

 $\delta_a = 1,8. \ 10^{-10} \ [s]$ : water permeability coefficient of stagnant air

Given the (measured) dependence  $\xi(\varphi)$  and  $\mu$ -factor, the diffusion coefficient  $a_v$  follows from the sorption and permeability measurements. As  $\xi(\varphi)$  increases with relative humidity, the diffusion coefficient decreases with increasing RH, as can be seen in Figure 6-10.

The experimental results and the model calculations are drawn in one graph for each RH step. Each sensor on the graph has a different color. The corresponding RH predicted by the numerical model is shown by thick lines. Since the permeability experiment didn't give very reliable data, the  $\mu$ -factor is varied somewhat in these calculations

Since every next RH step of the experiment started before the last sensor had reached its final RH, the initial values for each RH sensor at each RH step in the numerical model were obtained from a second order polynomial fit using a data analysis program called DATAFIT<sup>17</sup> (See also Appendix II.6).

<sup>&</sup>lt;sup>17</sup> DATAFIT is a science and engineering tool that simplifies the tasks of data plotting, regression analysis (Curve Fitting) and statistical analysis. (www. oakdaleengr.com)



### • Range 45-60% RH



The  $\mu$ -factor which fits the model best to the experiment was found to be smaller than 1 ( $\mu$ =0,88).

Figure 6-11: Diffusion coefficients for the range 45-60%



Figure 6-12: Diffusion according to  $\mu = 1$ 

Figure 6-13: Diffusion according to  $\mu = 1,2$ 



Figure 6-14: Diffusion according to  $\mu = 0.88$ 

While the other sensors of the model fit pretty well to the experiment, RV6 of the experiment increases faster than the model RV6\_m.



#### Range 60-75% RH



Figure 6-15: Diffusion coefficients for the range 60-75%



The best fit is found for  $\mu = 1,10$ . The fast increase of RV6 of the experiment with respect to the model (RV6\_m) continues also at this step.



Figure 6-17: Diffusion for  $\mu = 0.80$ 

Figure 6-16: Diffusion for  $\mu = 1,30$ 





Another interesting point is that the RV5 and RV6 intersect at the end of this range. The reason of this phenomenon is not known exactly. Two possibilities could be thought; either the sensors are not working accurately, or really the RV5 and RV6 are equal. (It should not be forgotten that the distance between RV5 and RV6 is 30cm.)

• Range 75-90% RH



Figure 6-19: Diffusion Coefficients for the range 75-90% RH



Figure 6-21: Diffusion according to  $\mu = 1$ 





Figure 6-20: Diffusion according to  $\mu = 1,55$ 



Figure 6-22: Diffusion according to  $\mu = 1,3$ 



# 6.4.3 Discussions

Two of the RV sensors at the diffusion graphs deviate from the model at all the RH steps; namely the sensors RV3 and RV\_6. The RV3 deviates from RV3\_m while the other sensors of the model and the corresponding sensors of the experiment fit each other (except for RV6\_m). The deviation of these sensors can be seen more clearly at higher RH steps. The deviation of RV3 is thought to be either due to the local density difference of the wool specimen or due to the restrictions and assumptions of the model.

RV6 increases more rapid than the corresponding model (RV6\_m) but moves parallel to the model at all the RH steps. Therefore, the reasons of the deviation of RV6; is thought to be either due to the model restrictions, or the late sorption property of wool.

The predicted  $\mu$ -factors at each RH step which fits best to the diffusion experiment is given in Table 6-7 and Figure 6-23. The predicted  $\mu$ -factor's which fit the best to the diffusion experiment is found to be lower than 1 at the 45-60% RH step of the experiment. But we know from the definition that  $\mu$ -factor can not be lower than 1. Another interesting result is the increasing values of the  $\mu$ -factor with increasing RH steps, instead of decreasing.

Table 6-7 µ-factor at RH steps

RH	MU_FACTOR
0,6	0,88
0,75	1,1
0,9	1,3



Figure 6-23: µ-factor per RH step

The predicted  $\mu$ -factors of the diffusion experiment were calculated according to the  $\xi$ -value which is found from the reference data of 16-19 days of the sorption isotherm. The effects of changing the reference data for the sorption isotherm (in the beginning or at the very end of the isotherm) has to be analyzed in order to see the effects of the chosen  $\xi$  value on the  $\mu$ -factor.





# 7 Case Study

Simulation program is a computer program that makes an abstract model of a particular system. The modeling of a real case on the simulation programs helps us to understand the influences of different circumstances on the behavior of the materials and to find practical solutions to problems. A practical case of a thatched roof is modeled and analyzed with the simulation program WUFI in this chapter.

# 7.1 The Case

Thatching is the craft of covering a roof with vegetation such as straw, water reed, sedge, rushes and heather. It is probably one of the oldest roofing materials and has been used in both tropical and temperate climates. It is still used in many countries in the world and in the European countries like England, Denmark and the Netherlands. The most commonly used thatching material is water reed and it



Picture 7-2 Thatched Roof

Picture 7-1 Close up of a Thatched Roof

is used for any type of buildings. There are two common types of thatching; the Open Structure (Traditional) (see also Figure 7-2 and Appendix III.3) and the Closed Structure (the Screw Roof) (see also Figure 7-1 and Appendix III.3). The closed structure is used more and more in order to meet the building regulations.



Figure 7-1 Closed Structure

Picture 7-3 Closed Structure





Figure 7-2 Open Structure

Picture 7-4 Open structure

The fact that water vapor moves under temperature gradient to the colder side of the construction and forms condensation at the inside surface leads man to find solutions to this problem. This problem comes across especially in hot and humid weather circumstances.

That hygroscopic materials including sheep wool can absorb water is thought to be an alternative solution for this problem. It is thought that insulating the construction with hygroscopic insulation material from the inside and then to place a vapor barrier can help to avoid condensation by the absorption property of wool. Therefore a traditional construction type, thatched roofs, was analyzed with a simulation program, WUFI.

# **7.2 WUFI**

The simulations were done by WUFI 3.3-Pro. WUFI is a Windows-based, menu-driven program for the hygrothermal analysis of building envelope constructions which is developed by Fraunhofer Institut Bauphysik (IBP) in Germany. It can calculate the heat and moisture transport in one-dimensional multi-layered building components.

# 7.2.1 Case Assembly

The number of the layers in a generated construction type and the thicknesses of them can be applied; and the types of the materials (Material Database button) can be chosen with the user interface as either graph or table (Figure 7-3). The most common materials on the market can be found in the Material Database of WUFI with their basic<sup>18</sup> and optional<sup>19</sup> hygrothermal properties. The materials in the database can be modified by inserting new parameters under another name. A material, which can not be found in the database, can be described saved with its hygrothermal and properties (Figure 7-4).

The basic parameters of the new material can be found from the literatures and applied on the table (Basic Values table on Figure 7-4). These parameters of a new material are required as a minimum in order to make a calculation.

The optional data parameters are free of choice to fill. If there is a need to use one of these parameters, the optional data are entered in tables (Edit Table button in Figure 7-4) and seen as a graph in the user interface.

Project     Cose: Open structure     Cose: Open, wool     Cose: Cosed, wool     Cose: Cosed, wool foil (Curren     Generation     Component     Anamby/Monitor Position     Superstand	Project/Case: SIMULAT	Project/Case: SIMULATION of THATCH/closed_Wool_foil				
	Assembly/Monitor Positions Assembly Layer Name thatch 130ko/m3	Orientation/Inclination/Height	Surface Transfer Coeffic Thickn.[m]	ients Initial Conditions		
	Extenior	0.3 0	0125 0,10 0,001	E materia cara		
Control				Rew Layer		
Case: 5 closed structure				Belete		
				Edit Assembly by: Greph C Table		
	6.					
	Assign from	Grid				
	🖨 Material Databas	e Automatic Grid:	C Coorse C Mean C Fine			

Figure 7-3 User Interface of WUFI

Material-/Layer Data		×
Layer/Material Name sheep wool		•
Material Data Info-Text		
r Basic Values		Optional Data
Bulk density [kg/m²]	25	Moisture Storage Function Liquid Transport Coefficient, Suction
Porosity [m <sup>9</sup> /m <sup>9</sup> ]	0.8	Liquid Transport Coefficient, Redistribution
Specific Heat Capacity, Dry [J/kgK]	1800	Water vapour diffusion resistance factor, moisture-depen
Thermal Conductivity, Dry [W/mK]	0.035	Graph Edit Table
Water vapour diffusion resistance factor [-]	1.1	10
Coptional Parameter		1
		ε 8 · · · · · · · · · · · · · · · · · ·
		2
		tten o
		Ğ 4
		ate
Typical Built-In Moisture [kg/m²] 2,5		≥ 2
Color Layer Thickn. [m]  U,1	J	0 0.2 0.4 0.6 0.8 1.0
		Relative Humidity [ - ]
Paste into Material Database		✓ OK ¥ Abort 7 Help
		ou i Teh

Figure 7-4 Describing a new material



<sup>&</sup>lt;sup>18</sup> Basic Parameters: Bulk Density [kg/m<sup>3</sup>], Porosity [m<sup>3</sup>/m<sup>3</sup>], Specific Heat Capacity Dry [J/kgK], Thermal Conductivity Dry [W/mK], μ-factor [-]

<sup>&</sup>lt;sup>19</sup> Optional Parameters: Moisture Storage Function, Liquid Transport Coefficients, moisture-dependent thermal conductivity, moisture-dependent  $\mu$ -factor

## 7.2.2 **Positioning the Structure**

The orientation, inclination and positioning of the construction component can also be defined by WUFI. The orientation (1 on Figure 7-5) is the compass direction towards which the construction is facing. The inclination (2 on Figure 7-5) is the angle at which the analyzed surface is tilted with respect to the horizontal. The angle differs from  $0^{\circ}$ (flat roof) to  $90^{\circ}$  (vertical wall). (1) and (2) are needed for calculating the rain and radiation loads on the surface.



**Figure 7-5 Orientation and Positioning** 

The height (3 on Figure 7-5) and the rain coefficients are used to calculate the driving rain load on the building component.

### 7.2.3 Surface Transfer Coefficients

The surface transfer coefficients point out to which extent the conditions in the surroundings affect the building component, especially the heat and moisture flows through the surfaces. WUFI uses the following surface transfer coefficients for the cases; the Heat Resistance<sup>20</sup>, Sd-value<sup>21</sup> ( $\mu$ d), Short-Wave Radiation Absorptivity<sup>22</sup>, Long-Wave Radiation Emissivity<sup>23</sup> and Rain Water Absorption Factor<sup>24</sup> for exterior surfaces and; the Heat Resistance and Sd-value (µd) for the interior surfaces. It is also possible to use user-defined values for each Figure 7-6 Surface Transfer Coefficients

Exterior Surface Heat Resistance [m²K/W]	0.052	Boof	+
	i i wind dep	pendent	
Sd-Value [m]	_	No coating	•
Short-Wave Radiation Absorptivity [-]	0.4	Wood (spruce):untreated	•
Long-Wave Radiation Emissivity [ - ]	0.9		
Rain Water Absorption Factor [-]	<u> </u>	According to inclination and construction type	Ŧ
Interior Surface			
Heat Resistance [m²K/W]	0.13	(Roof)	
Sd-Value [m]	_	No coating	•

parameter above.



<sup>&</sup>lt;sup>20</sup> Governs the convective and (long-wave) radiative heat exchange between the component and its surroundings [m2K/W]

<sup>&</sup>lt;sup>21</sup> µd value of the component [m]

<sup>&</sup>lt;sup>22</sup> The proportion of total (short-wave) solar radiation that is absorbed by the component [-]

<sup>&</sup>lt;sup>23</sup> The efficiency of long-wave emission [-]

 $<sup>^{24}</sup>$  The influence of rain is given with the rain water absorption factor [-]. This factor depends on the roughness, orientation and inclination of the structure. It is equal to "1" for horizontal surfaces and "0" for vertical surfaces. WUFI uses a value of 0,7 for ordinary walls.

## 7.2.4 The Climate

The climate of the cases can be generated by WUFI weather generator. WUFI has its own temperature and relative humidity of some countries (USA and Canada (54 cities), Germany (Kassel and Holzkirchen), Switzerland (Davos, Locarno and Zurich) and Finland (Espoo)) (See Figure 7-7). The climate of a certain place can also be derived from a measured available data (Figure 7-8). In this case; the parameters temperature, relative humidity, driving rain and radiation have to be saved in one of TRY, WET formats in \*.KLI files<sup>25</sup>.





**Interior Climate** 



#### **Figure 7-9 Interior Climate**

Figure 7-8 Generating Own Climate file

The interior temperatures and relative humidity is generated by WUFI as a sine wave with a period of one year ((1) and where the mean values. (3)).the amplitudes and the time of the maximum values can be specified by the user. The specified climate is seen as a graphical diagram (2). The predefined mean values and amplitudes can be selected by clicking on the appropriate section of the "moisture load"<sup>26</sup> diagram (4). If the option "Air Conditioning" (below in the dialog box) is selected, both the temperature and the relative humidity will be set to fixed value during the whole year.



<sup>&</sup>lt;sup>25</sup> These are special formats and files for meteorological data.

<sup>&</sup>lt;sup>26</sup> Moisture load is the difference of the water vapor concentrations in the interior and the exterior air.

# 7.2.5 View Calculations

WUFI views the calculation results of the model in the form of a film; so that the thermal and hygric processes in the structure components can be shown as an animation during the simulation period. The film can be started, paused, stopped or reset by the control buttons ((1) on the Figure 7-10).



Figure 7-10: Example film display of WUFI; Source WUFI

Because the climate data is available and coupled on the model, WUFI applies the selected climate data while running the calculation. The box (2) on the display shows the current date the film has reached.

In the left part of the screen, the current progression of the hourly profiles of temperature, relative humidity and water content through the structure is displayed as a film (3) on the two main graphs. The water content (blue) and relative humidity (green) are displayed in the same diagram and, the temperature (red) is displayed in a separate diagram. The X-axis corresponds to the thickness of the modeled structure. The "left Y-axis" corresponds to the water content, and the "right Y-axis" to the relative humidity through the structure. The current RH of inside, outside and through the structure is shown with a green triangle on the right side, on the left side and as a dark green line respectively on the graph.

The current temperature of the inside, the outside and through the structure is shown with a red triangle on the right side, on the left side and as a red line respectively on the graph. The yellow area shows the history of the temperature through the structure until that moment of the film. The vertical lines on (3) (except for the grid lines) show the component boundaries of the structure.



# 7.3 The Simulations

Both types of thatched roof (open- and closed structure) were simulated. The simulations were done for 30 cm thick thatch as mentioned in the *Literature 54*. Both construction types were analyzed first without insulation, secondly with insulation and at the last analysis with insulation plus vapor barrier. The selected materials for the insulation were the Sheep wool, Flax and glass wool insulation to see the effect of different kind of insulation materials on the moisture transport through the structure. The parameters for the sheep wool were applied to the material database according to the results of the experiments as found in the previous chapter. The properties of the materials used for the simulations are as follows:

Name	Thatch	OSB	Plywood	Sheep Wool	Flax	Glass Wool	PVC Roof Membrane
Thickness [m]	0,30	0,02	0,02	0,10	0,10	0,10	0,001
Density [kg/m <sup>3</sup> ]	130	555	500	25	38	30	1000
Porosity [m <sup>3</sup> /m <sup>3</sup> ]	0,70	0,60	0,5	0,9	0,95	0,99	0,0002
Hygroscopic	Yes	Yes	Yes	Yes	Yes	No	No
*WC (98% RH) [kg/m <sup>3</sup> ]	35	82	150	5,8	13	0	0
Wsat [kg/m <sup>3</sup> ]	42	593	350	6,1	348	0	0
Ther. Cond. [W/mK]	0,15	0,101	0,1	0,035	0,038	0,035	0,16
μ -factor [-]	3	287	700	1,2	1,5	1,3	15000
* WC· Water Content							

 Table 7-1: Properties of the Materials which are used in the Simulations

The  $\mu$ -factors for the materials are taken from the database of WUFI and give the  $\mu$ -factors for each material at 0% RH. The simulations were done in periods of two years (from **January 1<sup>st</sup> 2004** until **January 1<sup>st</sup> 2006**). Because the most critical orientation for the rain in the Netherlands is the South-West (SW) orientation, the simulations of the thatched roof, which has a slope of 45<sup>0</sup>, were done in this orientation. The rain water absorption factor is chosen as 0,9 because also the thatch is a highly porous and hygroscopic material.

It has been mentioned that the climate data of a case can be generated with the climate generator. Because there was not enough time to generate a climate data for a city in the Netherlands, and selecting a climate data of a city nearby the Netherlands makes not much difference on the outcome of the simulations; the exterior climate of Kassel (Germany) has been chosen for the simulations. The interior climate for the simulations was designed as 21 <sup>o</sup>C with amplitude of 1 <sup>o</sup>C for the temperature, and 60% with amplitude of 10% for the relative humidity.

The relative humidity between the layers is monitored since they are the critical points on the structure. The relative humidity of the critical points in the simulated structure is mentioned in the relative humidity graphs with both names of the two layers. For example; the relative humidity between the insulation and OSB is emphasized by OSB-wool (between the insulation and OSB) (Figure 7-11).



Figure 7-11 relative humidity between the layers of the structure

# 7.3.1 Open Structure

#### Without insulation

The traditional open structure was analyzed first without insulation as it is in old buildings, where thatch lays with 30 cm of thickness between outside and inside.



Figure 7-12: Application of open structure thatched roof

The traditional structure is very damp open (Figure 7-14). This is also an expected result from a very open structure.



Figure 7-13 Water content of the thatch during two years of simulation period

The water content of the thatch (Figure 7-13) is below the maximum (Table 7-1) during the two years simulation period. There is not much difference between the relative humidity of inside and the outside. Except for the surface (<3cm), the relative humidity can be assumed constant and equal to outside through the structure.



Figure 7-14: Result of the simulation for uninsulated thatched roof (yellow region is the temperature within the simulation period and green region is the relative humidity through the thatched roof.)

The temporal sudden relative humidity changes are buffered by thatch. The water content during the simulation period is also equally spread through the thatch.



# 7.3.2 Open Structure + Sheep Wool insulation

It is well known that to lay an insulation layer behind an open structure causes condensation problems in the construction because of outwards movement of the inside moisture due to the temperature gradient. But still, sheep wool insulation is laid behind the open structure to see how the structure reacts and if sheep wool insulation has any positive effect against the condensation which is expected to occur between the thatch and insulation.





Figure 7-15 Insulation layer behind the thatch

Figure 7-16 the outside RH (red) and the RH between the thatch and insulation material (blue) during simulation period of two years



**Figure 7-18 Water Content of the materials** through the simulation period



Figure 7-17 Simulation result of the insulated (sheep wool) open structure

As expected, the winter months were very problematic (Figure 7-16). The moisture condenses in the insulation and the condensed water moves outwards through the thatch except for the summer months (Figure 7-17).

Wool has reached the maximum water content  $(6.3 \text{ kg/m}^3)$  and condensed with water content of 9,17 kg/m<sup>3</sup>. Because further information could not be found, it is not possible to interpret about the condensation in the thatch. But thatch can absorb  $32 \text{ kg/m}^3$  water at 95 % RH. So, it can be possible that the moisture in the thatch has not condensed.



# 7.3.3 Open Structure + Insulation + Retarder

Because of condensation in the insulation and in the thatch, the structure was covered in this case with a vapor barrier from the inside.



Figure 7-19 Open structure + insulation + vapor barrier

The relative humidity of the exterior air is mainly buffered by the thatch. The amplitude of the humidity is hindered until the vapor reaches the insulation (Figure 7-20).



Figure 7-20 relative humidity in the construction

The relative humidity between insulation and retarder increases only in the summer months (180-240 days). The relative humidity increase between the thatch and the insulation is due to the difference of  $\mu$ -factor between two surfaces.



Figure 7-22 water content of the materials during simulation period



Figure 7-21 simulation results of open structure + insulation + vapor retarder

The materials have not reached the maximum water content during the simulation period of two years.

Table 7-2 Water Content [kg/m3]

	Literature	Simulation		
material	98% RH	Max.		
thatch	35	18,39		
sheep wool	5,8	3,55		



Another type of retarder with lower  $\mu$ -factor (PA membrane;  $\mu$ -factor = 4380 at 0%RH) has let more vapor from one side to the other; and therefore there is less relative humidity gradient through the thatch (Figure 7-24). Because the retarder is more open, the relative humidity between the thatch and wool fluctuates in the structure more than the one where the retarder has higher  $\mu$ -factor.



Figure 7-23 relative humidity through the structure



Figure 7-25 water content of the materials through the simulation period



Figure 7-24 Simulation of open structure which has lower  $\mu\text{-}factor$ 

Table 7-3 water content of thematerials [kg/m3]

	Literature	Simulation	
material	98% RH	Max.	
thatch	35	21,38	
sheep wool	5,8	3,91	

The water content of the materials in both cases dose not differ much from each other (Table 7-3) and (Table 7-4).

## 7.3.4 Closed Structure

The thatched roofs are built according to building regulations; and therefore they have to be built up as closed structure. The roof can be covered with different kinds of wooden boards under the thatch. The Organization of Thatchers in the Netherlands has not defined the sort of wooden boards; therefore OSB is chosen for plating under thatch. The only defined parameter about the wood plate is that the thickness of the plate has to be thicker than 19 mm.



Figure 7-27 Closed Structure Thatched Roof

The roof is covered with OSB (Table 7-1), with thickness of 20 mm. The wooden board under the thatch functions as a vapor barrier because of high  $\mu$ -factor (Figure 7-26).



Figure 7-28 RH of the structure during the simulation period



Figure 7-29 water content of the materials during simulation period



Figure 7-26 Simulation of Closed Structure (OSB)

The relative humidity inside the thatch does not fluctuate much during the simulation period. The sudden relative humidity changes on both sides of the construction (exterior and interior) are hindered by thatch and by OSB respectively (Figure 7-28).



#### 7.3.5 Closed Structure + Sheep Wool insulation

Sheep wool insulation is laid behind the structure although it is known that insulating structures from inside can cause condensation and it is advised to use vapor barriers from the inside. This case shows how serious the problem can be.



Figure 7-30 Insulated closed Structure

The relative humidity in the insulation increases rapidly due to wooden plate (OSB) layer. The moisture begins to condense in the insulation and on the wooden plate. The calculated water content for sheep wool reaches up to  $40 \text{kg/m}^3$ , which is not realistic. Because the thatch has high water capacity, the fluctuation of the relative humidity in the thatch is relatively small (Figure 7-31).

The relative humidity between sheep wool insulation and OSB (red, Figure 7-32) explains the high water content in Figure 7-31. The moisture moves outwards due to temperature gradient; which causes condensation on the colder surface of OSB. However, relative humidity of inside and the relative humidity between the OSB and thatch move with phase difference during summer months while the relative humidity is decreasing (Figure 7-32).



Figure 7-33 Water content of Materials



Figure 7-31 Simulation of insulated Closed Structure



Figure 7-32 Relative Humidity between the layers of the structure (between the insulation and OSB, OSB and thatch; and interior RH)


#### 7.3.6 Closed Structure + Insulation + Vapor Barrier

The insulated closed structure was analyzed with two kinds of insulation materials; first with sheep wool insulation and than Glass wool.



Figure 7-34 Insulated Closed Structure with Vapor Barrier

#### **Sheep Wool Insulation**

The relative humidity between the OSB and sheep wool has increased only in the beginning of the simulation; probably because of the begin values of the simulation (Figure 7-35). The vapor between the OSB and wool, which may contain condensation risk at extreme temperatures, is absorbed especially by OSB. The outside relative humidity is buffered by the thatch until the moisture reaches to the OSB (Figure 7-36).





Figure 7-35: simulation of insulated closed structure +vapor barrier

Figure 7-36 Relative humidity through the structure

The water (vapor) content of the materials never reaches the maximum (Table 7-4). The moisture of the construction is absorbed especially by the thatch and OSB.

Table	7-4	Water	Content	[kø/m3]
able	/	vv ater	Content	[kg/mJ]

	Literature	Simulation
Material	98% RH	Max.
thatch	35	18,27
OSB	82	32,30
Sheep Wool	5,8	3,18



Figure 7-37 Water content of the materials during simulation period



#### **Glass Wool Insulation**

The structure was insulated this time with another fibrous insulation material (glass wool), which cannot practically absorb any moisture; and the transport of the moisture is analyzed during the simulation period.



Figure 7-39 Water content during simulation period



Figure 7-38 Simulation of the structure

The simulation with glass wool has shown that behavior of total structure is similar to the one with the sheep wool in same circumstances (Figure 7-35) and (Figure 7-38). The water content of the thatch and the OSB is almost equal during the simulation period (Figure 7-39) and (Table 7-5).

Table 7-5	Water	Contents	of the	materials
[kg/m3]				

	Literature	Simulation
Material	98% RH	Max.
thatch	35	18,21
OSB	82	31,29
Glass Wool	0	0,94

The only difference is the water content of the glass wool insulation. It is known that glass wool absorb no water; therefore it is assumed that the water is in vapor form in glass wool.



#### 7.3.7 Discussions

Thatch has, as an individual material, an open structure; and because of the high absorption capacity ( $35 \text{ kg/m}^3$  at 98% RH) and thickness, it can buffer the moisture.

Because there is not much difference between the behaviors of two vapor-tight and insulated closed structures (closed structure with sheep wool and the one with glass wool), there has been another simulation done with another type of wooden plate. In this case; the OSB is replaced with Spruce (longitudinal direction, which has lower  $\mu$ -factor) and the PVC roof membrane ( $\mu$ = 15000) is replaced with PE-membrane ( $\mu$ = 2000) (Figure 7-40).





The hygroscopic behavior of the insulation material makes not much difference. But the  $\mu$ -factors of the wooden plates influence directly the transport of moisture. The wooden plate with high  $\mu$ -factor can cause condensation problems on the side to the insulation material. The hygroscopic behavior of the insulation material does not affect much the behavior of the total construction, but the  $\mu$ -factor.



#### **Colder Climates**

The last simulation is done in Espoo (Finland) to see the effects of the materials in colder climates, but it is not given in detail here.



Figure 7-41 Comparison of the construction materials in Colder Climates

For both sheep wool and glass wool insulation, the best materials are the combination of vapor retarders with high  $\mu$ -factor and wooden boards with low  $\mu$ -factor. The worst case is the combination of vapor retarders with low  $\mu$ -factor and wooden boards with high  $\mu$ -factor. Again here, the  $\mu$ -factor of the wooden board is decisive.





## 8 Conclusions and Recommendations

As sheep wool insulation is quite new in the construction market, its material properties pertaining to civil engineering applications are not well-known yet.

Moisture behavior of wool formed the main topic of this research. The hygroscopic behavior of sheep wool insulation and the effect of sheep wool insulation in building constructions on the moisture balance were analyzed. Three controlled experiments were carried out on sheep wool. First, the vapor resistance of the wool was measured using the principle of the cup-method. Second, the sorption isotherm was determined using mass measurements. In the third experiment, the time- and space-dependent moisture transfer in a column of wool was measured.

#### Vapor Permeability

In the vapor permeability experiment, the  $\mu$ -factor of the specimen was calculated from the measured rate of the diffusion. Due to the cylindrical geometry of the experimental set, the results had to be compared with a 2D -cylindrical finite-element calculation. This analysis indicated that the vapor resistance ( $\mu$ -factor) lies between 1,2 (at 60% RH) and 2,12 (at 95% RH). As the  $\mu$ -factor is expected to be constant or decrease with increasing RH, it is speculated that the increase of  $\mu$ -factor was probably due to dilution of the (top layer of the) salt solution. Consequently, the measurement of the permeability at lowest measured RH at the start of the experiment is probably the most reliable one. Therefore, it is concluded from this experiment that the  $\mu$ -factor is 1,2 ±0,2.

#### Sorption isotherm

In these measurements it was seen that the sorption occurred in two stages after a step in RH. This agrees with findings of Watt [49, 50 and 51]. The first stage was fast, and occurred within a couple of hours, while the second step was very slow and took days to reach thermodynamic equilibrium. The second sorption stage was hardly present in the 45-60% RH range, but it was clearly observed in the range of 75-90% RH. This slow second stage of the adsorption is thought to be due to swelling of wool with increasing relative humidity. Watt [51] refers to structural changes within the wool-water complex during sorption.

A third experiment was carried out which focused on the time and space-dependent vapor transfer in a column of wool. The data from this experiment was used to determine whether the vapor transfer inside the wool specimen could be described using a one effective differential equation for the vapor transfer (Fickian diffusion model) or that non-Fickian effects do occur. The model showed reliable results for the first 30 cm except for the last sensor in the bottom of the glass tube (60 cm). The last sensor at the bottom of the glass tube increased more rapid than the corresponding model at all RH steps. This deviation was more visible at higher RH steps.



The  $\mu$ -factor for the model in the diffusion experiment which fitted the best to the experiment was found to be " $\mu$ -factor = 0.88" at the first step of the experiment (45-60% RH), -with increasing values-  $\mu$ -factor = 1.1 for 60-75% RH step and  $\mu$ -factor = 1.3 for 75-90% RH step. These findings contradict expectations of a constant of decreasing vapor resistance with relative humidity. A likely cause of this unexpected result, and the deviation between experiment and model mentioned above, is thought to relate to the slow second stage of absorption. This (time- and/or concentration dependent) sorption behavior leads to non-Fickian diffusion and is not taken into account in the tested diffusion model.

The measured properties of sheep wool were applied in a simulation program to evaluate practical consequences of using sheep wool in building constructions. The condensation at the colder side of the construction which occurs under a temperature gradient was analyzed for European climates (Kassel, Germany) for open- and closed- structure thatched roofs. The thatched roof was analyzed first without an insulation layer and vapor retarder; to see how thatch reacts in different weather circumstances. It was seen that as thatch is very open to vapor flow and; because of its relatively high sorption capacity and thickness (usually 30cm thick), the daily outside relative humidity changes was buffered within first 5 cm of the material.

When an insulation layer was applied to the (open structure) thatched roof, it was seen during simulations that -as expected- the winter months were very problematic (**Figure 7-16**), regardless of the type of the insulation material. The moisture condensed in the insulation and the condensed water moved outwards through the thatch except for the summer months. When a vapor retarder was applied to the insulated thatched roof (open structure), the condensation which occurs due to the inner vapor concentration was prevented; again regardless of the type of the insulation material.

Because wooden plates have higher resistance than insulation materials, applying an insulation layer to the closed structure thatched roof without a vapor retarder was the worst case of all; regardless of the type of the insulation material. The interstitial condensation was prevented when a vapor retarder was applied to the insulated closed structure. The best construction type for the closed structure thatched roofs was the combination of vapor retarders with high resistance and wooden boards with low resistance against water vapor transport.

Simulations showed that the hygroscopic behavior of the insulation materials have not much effect on closed structure thatched roofs, in European countries, on avoiding interstitial condensation which occurs due to the temperature gradient. But the resistance of the wooden plates against water vapor can cause interstitial condensation which occurs due to inside relative humidity. Therefore; instead of hygroscopic insulation materials, vapor retarders seems to be the essential construction materials for the thatched roofs.

#### Recommendations

Much study still has to be done on sheep wool to be able to fully understand the behavior and the properties of the material. It is important to know the fractions of <u>ad</u>sorbed water



and <u>ab</u>sorbed water as a function of relative humidity, the rate at which water molecules penetrate a wool fiber and the accompanying swelling as that increased the surface area for adsorption. If the kinetics of sorption is known, there would be a better basis for a mathematical description of water vapor diffusion in the bulk wool material. As wool bulk shows non-Fickian behavior, more specific study on the diffusion of water vapor in wool column for both sorption and desorption will help to understand and formulate the behavior better.

All the experiments in the literatures were done with the wool dried at 105  $^{0}$ C; but it is known that wool consists of proteins; and proteins begin to change their structure at about 70  $^{0}$ C (was also seen at fire resistance tests as 100  $^{0}$ C). Therefore; the effects of drying wool at 105  $^{0}$ C on the hygric behavior also have to be analyzed. Furthermore, the outer side of the insulation materials reaches, especially in the summer moths, up to70-80  $^{0}$ C. Therefore the durability of the sheep wool insulation has to be analyzed also for practical cases.

The diffusion processes (consequently the  $\mu$ -factor) of the wool materials at different densities have to be analyzed for next studies. If a correlation can be found between the diffusion properties and the density; the resistance of each wool product against vapor diffusion can be determined.

Mixing wool with artificial fibers (nylon, polyester) while producing insulation materials would be, from the viewpoint of vapor diffusion behavior, also another interesting topic to be studied.



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## I APPENDIX WOOL

#### I.1 Fiber Characteristics of Wool

Wool is produced in the fiber follicle in the skin of the sheep. Wool consists of a group of proteins which is called keratins. Keratins are classified as hard of soft according to their tactical properties. The difference of hard keratins (such as wool, hair, horn, and feathers) from soft keratins (such as in skin) are the higher concentration of sulphur (3%). The sulphur is present in the form of residues of the amino acid cystine. Keratins are classified as alpha or beta-types according to their X-ray diffraction patterns. [6]

Each wool fiber has an outer layer of flat, scale-like cells which overlap like shingles and which are covered with a thin membrane. This is known as the epidermis. This membrane keeps away rain, but water vapor can penetrate it. The protein cells in the center of the fiber absorb the moisture, which may penetrate the membrane. This is known as the cortex.



Figure 1: Morphological Components of a Wool fiber, source: Literature6

Clean wool contains only 82% of the keratinous proteins, which are characterized by a high concentration of cystine. Approximately 17% of wool is composed of nonkeratinous proteins (nonkeratinous because of their low cystine content). The wool contains 1% by mass of waxy lipids; this is believed to be concentrated in the intercellular regions of the fiber. These nonproteinaceous proteins are concentrated in specific regions of the structure. Nonkeratinous proteins more labile and less resistant to chemical attack than keratinous proteins because of lower concentration of disulphide cross links.

#### I.2 Morphological Structure of Wool

Fine wool contains two types of cell: **cuticle** and **cortex** held together by a cell membrane complex. These two together form the major part of the clean wool. Both the cortex and the cuticle influence the fiber properties of the wool.





Figure 2: simplified diagram of wool, source Literature 6



Figure 3: photo of a wool fiber, Source Literature6

#### Cortex

The cortex (constitutes almost 90% of keratin fibres) has a bilateral structure and is **responsible for the mechanical behavior**. Cortex is the carrier of the characteristics of wool properties such as elasticity, ductility and swelling force. Wool fibers contain two main types of cortical cell, orthocortical and paracortical. The cells of cortex contain around 13% of nonkeratinous proteins. The orthocortical and paracortical cells are defined by the distribution of nonkeratinous proteins in the cell. Another difference is the amount of crosslinked matrix between the microfibrils in each cell. These differences make the orthocortex more accessible to reagents and more chemically reactive than the paracortex. Chemicals reach the cortical cells by diffusion along the network of the cell membrane complex that extends through the whole fiber.



Figure 4: structure of cortex, Source: literature 6

#### The Cuticle

The cuticle cells constitute the outer surface of the wool fiber and **responsible for wet ability, tactile properties and felting behavior**.(*Felting occurs when individual fibers move preferentially in one direction. Such movement occurs readily when the fiber assembly is agitated in water.*)



Approximately 10% of the wool fiber consists of cuticle cells. The cell overlaps on each other like tiles on a roof. The function of cuticle cells in keratin fibers is to anchor the fiber in the follicle on the skin of the animal. The amount of each cuticle cell visible on the wool surface varies with the fiber diameter.

The cuticle has higher cystine content than whole wool. Cuticle cells are also rich in cysteic acid, serine, praline, gylicine and valine. They are poorer than whole wool in aspartic acid, threonine, glutamic acid, methionine, isoleucine, leucine, tyrosine, phenylalanine and arginine. The cuticle is less extensible than the cortex, maybe because of the higher level of cystine. Cystine plays an important role in the stabilization of the fiber structure of wool through the cross linking action their disulphide bonds. The disulphide bonds are responsible for the relatively good strength of wool, and particularly for its low material swelling.

The cuticle cells have two distinct major layers, namely exocuticle and endocuticle. The third layer, the epicuticle, is defined as the part of cuticle cell resistant membrane that is located on the fiber surface covering the surface. It is known that it is the resistant membrane system that surrounds all cuticle and cortical cells and is strongly hydrophobic in character.



Figure 5: Structure of Cuticle, Source Literature6

#### Exocuticle

This is a layer just below the epicuticle and in merino wool it represents the 60% of the total cuticle cell. The major part of the cystine content is believed to be in the exocuticle cell.

#### Endocuticle

The endocuticle is a layer lying under the exocuticle. It is bounded by cell membrane complex which separates it from the other cuticle cells or from the cells of the cortex. The endocuticle is about the 40% of the whole cuticle.



#### I.3 Chemical Bonding

#### **Covalent bond**

Covalent bonding is a form of chemical bonding characterized by the sharing of one or more pairs of electrons, by two atoms, in order to produce a mutual attraction; atoms tend to share electrons, so as to fill their outer electron shells. Such bonds are always stronger than the intermolecular hydrogen bond and similar in strength or stronger than the ionic bond. Commonly, covalent bond implies the sharing of just a single pair of electrons.

#### Peptide Bonds (reaction of two amino acids)

A peptide bond is a chemical bond formed between two molecules when the carboxyl group of one molecule reacts with the amino group of the other molecule, releasing a molecule of water (H2O). This is a dehydration synthesis reaction, and occurs between amino acids.

Polypeptides are chains of amino acids. Proteins are made up of one or more polypeptide molecules. Individual peptide chains in wool are held together by various types of covalent crosslinks and noncovalent interactions.

#### **Disulfide Bonds**

A disulfide bond (SS-bond), also called a disulfide bridge, is a strong covalent bond between two sulfhydryl groups. This bond is very important to the folding, structure, and function of proteins. Disulfide bonds are formed by oxidation of the sulphydryl groups on cysteine. The disulfide bonds of cystine form crosslinks, either between different protein chains or between different parts of the same protein chain. Different protein chains or loops within a single chain are held together by the strong covalent disulfide bonds. The amino acid cysteine undergoes oxidation and reduction reactions involving the -SH (sulfhydryl group). The oxidation of two sulfhydryl groups results in the formation of a disulfide bond by the removal of two hydrogens. The reduction of a disulfide bond is the opposite reaction which again leads to two separate cysteine molecules.

#### Noncovalent Bonds (between side chains)

These are secondary bonds and can occur within a single protein chain or between different chains. The noncovalent bonds in keratin fall in to three main groups. These are hydrogen bonds, ionic bonds and hydrophobic bonds. These names refer to the way the side groups interact with the environment

#### Hydrogen Bonds

A hydrogen bond is a type of attractive intermolecular force that exists between two partial electric charges of opposite polarity (between carbonyl groups and hydroxyl or amino groups). Although stronger than most other intermolecular forces, the hydrogen bond is much weaker than both the ionic bond and the covalent bond. Within proteins, it can exist between two parts of the same molecule and figures as an important constraint on such molecules' overall shape. The –CO and –NH groups in the peptide chains can interact through hydrogen bonds.



#### Ionic Bond

Ionic bonding occurs between charged particles (between acidic and basic side chains). These may be atoms or groups of atoms. In either case, the particles must have lost or gained electrons. Electrons have a negative charge, so a particle that gains electrons gains a negative charge. Equally, a particle that loses electrons must be left with a positive charge (assuming it started with no charge). Since opposite charges attract, the particles in an ionic compound are held together by this attraction. The concentration of ionic bonds depends on pH of the environment.

#### **Hydrophobic Bonds**

Hydrophobic interactions occur between non-polar molecules in the presence of a polar solvent such as water. In the context of protein structures, several amino acid side chains are, to varying degrees, hydrophobic. This type of bonding is believed to contribute to the mechanical strength of keratin, particularly at high water contents. The strength of hydrophobic interactions is not affected by changes in pH or in salt concentration.



# I.4 Influencing Factors on the Strength of the Fiber

The greater the moisture content of the fiber, the further it can be stretched. The water works as a plasticizer on the fiber [8]. In the Figure 3-3, we see the load-extension diagram of a single wool fiber at 25  $^{0}$ C, at relative humidity's 0%, 50% and 100%. The dry fiber resists elongation because of hydrogen bonds. In the presence of water some hydrogen bonded groups exchange their partners for movable water molecules.

Temperature has the same effect as the moisture content. As the temperature rises, the fiber becomes weaker and becomes easy to elongate. The result is expected for the action of water on a hydrogen-bonded structure.



Figure 8: Load-extension curve showing the effect of pH, Source: Lit. 8; (1Mg/cm<sup>2</sup> =100 N/mm<sup>2</sup>)



Figure 6: Load-Extension Curves of Wool Fiber under different RH conditions, Source Lit. 8 (1Mg/cm<sup>2</sup> =100 N/mm<sup>2</sup>)



Figure 7: Load-extension Curves for wool at various temperatures, Source: Lit. 8; (1Mg/cm<sup>2</sup> =100 N/mm<sup>2</sup>)

Both the effects of acid and alkali cause a decrease in the strength of the wool fiber [8].

The strength of wet wool depends to a large extent, on the covalent cross-links on the disulphide bonds. [8]



#### I.5 Chemical Properties of Wool

To explain the chemical properties of wool, first we have to have a knowledge about the proteins which constitutes the wool.

Considerations General concerning Protein: It has been estimated that wool contains about 170 types of protein molecule. Protein is a chemical complex that has been built from amino acids. The basic structural units of proteins are alphaamino acids. These basics of protein molecules are organic connections which chemically characterized are by simultaneous presence of basic and acid group in the molecule. Protein has therefore both basic and acid properties. Completely pure Protein reacts little acid, in other words, the isoelectric point lies on the acid side. (pH=4.8)

Amino acids are called biochemical building blocks. They form short polymer chains called polypeptides or peptides which in turn form structures called proteins. A **polymer** is a long, repeating chain of atoms, formed through the linkage of many molecules. We can say that proteins are polymers of amino acids. 18 different amino



Figure 9: Chemical Structure of wool, Literature: Doschawol

acids are used to synthesize keratin. The shape and other properties of each protein are dictated by the precise sequence of amino acids in it.

Keratin molecules are helical and fibrous, twisting around each other to form strands called intermediate filaments. These proteins contain a high percentage of sulfur-containing amino acids, largely cysteine, which form disulfide bridges between the individual molecules resulting in a fairly rigid structure.

The general formula of an amino acid is given below (COOH-CHNH2-R):

Each amino acid consists of an alpha carbon atom (in the middle) to which is attached

- a hydrogen atom
- an amino group (hence "amino" acid)





- A carboxyl group (-COOH). This gives up a proton and is thus an acid (hence amino "acid")
- One of 20 different "R" groups. The "R" represents a *side chain* specific to each amino acid. It is the structure of the R group that determines which one of the 18 amino acid it is and its special properties.

Amino acids are usually classified by properties of the side chain into four groups: acidic, basic, hydrophilic (polar), and hydrophobic (nonpolar). Complete hydrolysis of wool yields a mixture containing eighteen amino acids. There is a considerable variation in the values for amino acid content. Although some of the differences may be due to experimental error, there are several factors. These differences are influenced by genetic origin, physiological state and nutrition. The method of cleaning the sample before testing may also affect the result. In the table below we can see the structure and amount of the eighteen amino acids which form the wool.

Amino acid	Structure	[33,93]	[198]	Nature of side-chain
Lysine	H <sub>2</sub> N(CH <sub>2</sub> ) <sub>4</sub> CHCOOH   NH <sub>2</sub>	3.1	2.8	Basic
Methionine	H <sub>3</sub> CS(CH <sub>2</sub> ) <sub>2</sub> CHCOOH   NH <sub>2</sub>	0.5	0.4	Sulphur- containing
Cystine <sup>(d)</sup>	HOOCCH <mark>CH<sub>2</sub>SSCH<sub>2</sub>CHCOOH   H<sub>2</sub>N NH<sub>2</sub></mark>	10.5 <sup>(e)</sup>	10.0 <sup>(e)</sup>	Sulphur- containing
Tryptophan	H CH2CHCOOH	See te	×t	Heterocyclic
Proline	Соон	5.9	7.2	Heterocyclic

Figure 10: Structure and amount of Amino acids in wool, Source: Literature6

		Mol %		
Amino acid	Structure <sup>(a)</sup>	[33,93]	[198]	Nature of side-chain
Glycine	HCHCOOH   NH <sub>2</sub>	8.6	8.2	Hydrocarbon
Alanine	СН <sub>3</sub> СНСООН   NH <sub>2</sub>	5.3	5.4	Hydrocarbon
Phenylalanine	CH2CHCOOH	2.9	2.8	Hydrocarbon
Valine	H <sub>3</sub> CCHCHCOOH II H <sub>3</sub> C NH <sub>2</sub>	5.5	5.7	Hydrocarbon
Leucine	H <sub>3</sub> CCHCH <sub>2</sub> CHCOOH     H <sub>3</sub> C NH <sub>2</sub>	7.7	7.7	Hydrocarbon
Isoleucine	H <sub>3</sub> CCH <sub>2</sub> CHCHCOOH II H <sub>3</sub> CNH <sub>2</sub>	3.1	3.1	Hydrocarbon
Serine	HOCH <sub>2</sub> CHCOOH   NH <sub>2</sub>	10.3	10.5	Polar
Threonine	H <sub>3</sub> CCHCHCOOH     HO NH <sub>2</sub>	6.5	6.3	Polar
Tyrosine	но-С	4.0	3.7	Polar
Aspartic acid <sup>(b)</sup>	HOOCCH <sub>2</sub> CHCOOH	6.4	6.6	Acidic
Glutamic acid <sup>(c)</sup>	HOOCCH <sub>2</sub> CH <sub>2</sub> CHCOOH   NH <sub>2</sub>	11.9	11.9	Acidic
Histidine	CH2CHCOOH	0.9	0.8	Basic
Arginine	H 112 H <sub>2</sub> NCNH(CH <sub>2</sub> ) <sub>3</sub> CHCOOH	6.8	6.9	Basic

#### Figure 12: Figure 12 continued



The complete decrease of formaldehyde goes over two reaction phases:

An example for the reaction of formaldehyde with the amino acid side chains of the keratin is given below.

1. The bonding of reactive a hydrogen atom on the double bonding of formaldehyde. With this reaction, an amino-methyl derivative is formed.



#### I.6 Chemical Reactivity of Wool

Wool contains both dibasic and diacidic amino acids, which appear within the structure as basic and acidic side-chains. It's therefore amphoteric in character. There is always a balance between the acidic and basic groups in wool. The electrostatic interactions between the charged groups in the fiber play a role in the mechanical properties of wool.

The chemical nature of wool allows it to filter out and inactivate a wide range of chemicals, acid, neutral and alkaline materials. Furthermore, this chemical reactivity permits easy chemical modification to enable it to absorb an even every range of contaminants. For example, wool has been used to remove poisonous metals such as mercury and lead from air and water. Studies by the USA Environmental Protection Agency (USA-EPA) showed that wool can absorb up to 30% of its weight of mercury from polluted water under a wide range of environmental conditions.



Research in USA[12] has shown the chemical modification of wool on increasing absorption capacity; and treated wool can now be used to remove cadmium, iron, zinc, lead, mercury, cobalt, nickel, copper, uranium and many other metals from contaminated waters. This chemical reactivity of wool makes it useful for removing toxic chemicals such as heavy metals from the environment.

The first bonding and reaction mechanism was already described in the 40's by Fraenkel-Conrat and this has been confirmed by Mason and Griffith, 1964.

One gram wool bonds approximately 500-600  $\mu$ mol. Under ideal circumstances this is even 880 $\mu$ mol. That means that from 3.2 until 3.8% (by weight) of SO<sub>2</sub> can be bonded by one gram of wool. At 880  $\mu$ mol is that 5.6%.

For



Figure 14: Absorption of SO<sub>2</sub> by wool fiber (Temp. 21 <sup>0</sup>C, RH: 45%) Source: Doschawol



Figure 13: desorption of  $SO_2$  from the wool fiber (Temp: 50  $^{0}C$ )

NO<sub>2</sub>, it is the same principle such as for SO<sub>2</sub>.











#### Irreversible Binding with wool fiber (transformation and neutralization)

- ✓ Formaldehyde (the best known and the most reported)
- $\checkmark$  NO<sub>x</sub>, SO<sub>x</sub>
- ✓ Acetaldehyde, propionaldehyd, butyraldehyd, pentanal (from building and timber materials)
- ✓ Hexanal, heptanal, octanal, nonanal, decanal(responsible for smell),
- ✓ Benzaldehyd( Lacquers and surface treatments),
- ✓ Furfural (from cork),
- ✓ Acrolein, crotonaldehyd(beside formaldehyde, substantial components of the cigarette smoke),
- ✓ Glyoxal, glutardialdehyd, isocyanates (from different CH building products), nitrosamine(from motor-car tire), styrene.

#### **Reversible binding (decrease possibility)**

- ✓ toluol, xylene, cresol are very strongly adsorbed,
- ✓ Phenol,
- ✓ Pentachlorophenol (PCP)
- ✓ terpene, ester and amines in each case adsorbed, takes partly conversion,
- ✓ General VOC,

#### I.7 Insects

Wool can be attacked by the larvae of certain moths (Lepidoptera) and beetles (Coloeoptera). The most important wool-digesting insect pests are:

- ✓ Tineola bissiella (common cloth moths): this is worldwide distributed and therefore widely used for laboratory tests.
- Tinea (case bearing cloth moth): this exists in several forms in subtropical and temperate regions.
- ✓ Hofmannophila pseudosprettella (brown house moth): it is common in the moist temperate climates of New Zealand and western European coastal areas.
- ✓ Anthrnus (carpet beetle): this species is prevalent in subtropical and temperate regions.



Figure 17: Tineola bisselliella, Larvae, 20-24 days old olympus SZH10 stereomicroscope), Source: Literature 16



#### I.8 Wool treatments and Health Aspects

#### Treatments

As all the biotical insulation materials, sheep wool insulation also contains insect resisting agents in order to prevent attacks. Various chemicals have been applied to wool larval attack. The most used chemical in the sheep wool insulation industry is Mitin FF (sulcofuron). Mitin FF bonds chemically to the wool fiber so that there is not a problem of solvability.

#### IR Agents

Commercially available insect resist agents

Product	Trivial name of active	Active ingredient	Year of
	ingredient	in product (%)	introduction
1. Mitin FF high conc.	Sulcofenuron	80	1939
2. Eulan U33	chlorphenylid	33	1958
3. Eulan WA new	chlorphenylid	20	1961
4.Mitin LP	Chlorphenylid/flucofenuro	8	1972
5.Molantin P	chlorphenylid	32	
6. Eulan BLS	Tricholorobenzene- N-chloromethyl- sulphoamide	15	
7. Mitin4108	Chlorophenylid/ sulcofenuron	16.5 14.5	
8.Perigen	Permethrin	10	1980
9. SMA-V	Permethrin	20	1980
10. Antitarma NTC	Permethrin	7	1982
11. Mitin BC	Permethrin	10	1982
12. Mitin AL	Permethrin/ hexahydropyrimidine derivative	5 5	1983
13. Eulan SP	Cyfluthrin	3	1982
14. Cirrasol MPW	Cyhalotrin	5.6	1985
15. Eulan SPN	Permethrin	10	1987

#### Table 6: Source: Literature 6

The first pesticide emulsion to be applied commercially contained dieldrin. Because of its high mammalian and fish toxicity, dieldrin is not allowed anymore against insects.

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Sulcofuron = Mitin FF Status: ISO 1750 IUPAC: 5-chloro-2-{4-chloro-2-[3-(3,4dichlorophenyl) ureido]phenoxy}benzenesulfonic acid CAS: 5-chloro-2-[4-chloro-2-[[[(3,4-dichlorophenyl) amino] carbonyl]amino]phenoxy]benzenesulfonic acid Reg. No.: 24019-05-4 Formula: C19H12Cl4N2O5S

Figure 18: Chemical Structure of Mitin FF

#### Non-toxic Insect Resist Agents (THM Model)

Regarding sheep wool insulation, an efficient moth prevention system is required but clearly non-toxic alternatives are favored. A group of scientists are working now in Austria [16] about an alternative insecticide using toxic compounds of the plants which is called THM Model. The THM model is very safe, when compared with the pesticide model.

THM practices based on multiple-compound plant mixtures are effective and discourage development of insect resistance. Neem leaves (Azadirachta indica A. Juss) are used for this model, because they have a long history of use for many pest management purposes. The Neem products are quite promising, because of their low price, easy preparation techniques, and proven action over more than 400 species of insects. Reports indicate that Neem products are harmless to both, men and beneficial insect predators (Vogt et al. 1996, Erbach and Holst 1997). Neem products are approved worldwide to be used in organic farming. But because of its temperature sensitivity Neem alone is inappropriate to protect sheep wool insulation permanently against the attacks of insects. [16]

#### Health aspects during and after application

Because the agents are biologically active, they attract the attention of authorities and organizations that are responsible for the worker and consumer health and environmental protection. These biologically active additives are chemically bonded with the keratin and have a long life time.

During wool processing stage insecticides are breathed in during or after spraying and can also be absorbed through the skin as a result of direct contact. While using these insecticides special attention must be paid in the processing stage and therefore the prevention methods should be applied carefully.

It will not cause any problems during usage stage because these additives are then chemically bonded to wool, as sheep wool can chemically bond with many chemicals. And eventual fibers, which are breathed during application or processing, are solved in four days by human body. (Doscha, 2003)



#### I.9 Health Aspects regarding Wool

#### Allergies

Allergy has increased spectacularly since the beginning of this century, from 1% to 15-20%. The reasons for the increase of allergic rhinitis and asthma are found both in genetic and surroundings factors. On the question, if people can be allergic for wool, has been insured that it is not the case. People are not allergic for the wool fiber himself but for the additives to that, like wool dyes. The red coloring of the skin, generally on the thin skin parts, is explained as a mechanical irritation of the wool fiber on the skin.

In an experiment[19] which is done by two scientists, by means of several human sweat proofs, they have inhibited protein and lipid from wool, which was introduced after, in advance in an aqueous solution on the marked skin of a group test persons (skin prick test). Neither at hypersensitive, nor at test persons without predisposition, skin responses could be found as a result of wool extract. Both scientists have reached to this result in association with doctors of the Clinic for Dermatology of the University of Erlangen, which skin tests was carried out on 300 test persons.

#### Indoor Air Quality (IAQ) and Pollutants

There is no standardized definition for Indoor Air Quality, but in general it can be defined as the physical, chemical and biological properties that indoor air must have in order to secure high level of comfort and health. Low levels of indoor pollutants (VOC's, Radon, CO, NO<sub>x</sub>, particulate matters etc.) are important for IAQ and the health of occupants. The complaints due to the poor indoor IAQ are: lower respiratory, mucous membrane and central nervous system, eye irritation, headaches, fatigue, difficult sleeping, chest pain.

It has been shown in an experiment [27] that mass transfer between indoor air and permeable and hygroscopic building envelope can affect the indoor concentration of gases and water vapor. But to achieve this, the entire depth of the building envelope must be permeable and the permeability of building materials to these gases must be known. Only recently measurements have been done on the diffusion of gases through building materials (Kirchner et al., 1999). He mentions that the diffusion of gases through the building envelope significantly increases the effective ventilation rate for poorly ventilated rooms but moderately increases the effective ventilation rate of well-ventilated rooms [27]. The diffusion depends also to the size of the gases. The larger (bigger) the gas is the less is the diffusion.

Indoor air has been shown (by NIOSH (The National Institute for Occupational Safety and Health)) to be more polluted in most cases than outdoor air, especially in tight buildings. Sheep wool has ability to react with many chemicals and gases. The tests of NIOSH have shown that wool can continue absorbing for many years but at reduced rates in the later years. Accelerated simulated long term exposures (equivalent to 27 years) showed that wool continues to absorb with a rate of 60%. Wool is found to have irreversible sorption with these gases and is also not degraded by these gases. As insulation material sheep wool can also have a positive effect on IAQ on permeable and hygroscopic buildings. But 'to what extend is this positive effect' is not studied yet.

#### I.10 Factors affecting fungal growth

#### Water

Fungal growth requires the availability of water. The RH or  $a_w$  influence each of the main phases in growth cycle. The ultimate lower  $a_w$  limit for fungal germination is studies by many workers and is accepted as 0.64.

#### Temperature

The other important factor for fungal growth is the temperature. The majority of fungi in indoor environments grow at temperatures between 10 and 35  $^{0}$ C, however some of the fungi grow between 2 and 5  $^{0}$ C and others between 55 and 60  $^{0}$ C. The optimum temperature for the growth of fungi is between 22 and 35  $^{0}$ C. Indoor environments offer a wide variety of substrates for growth. Condensation is the principal source of moisture on internal surfaces of dwellings. Porous building materials such as concrete, brick and plaster can provide a moisture reservoir allowing fungal growth. Interior dampness problems are related to construction faults, such as cold bridging in combination with inadequate ventilation.

#### Nutrients

Due to the absence of chlorophyll in fungi, they can not photosynthesize, so they have to live as parasites. Fungi show an ability to utilize a wide range of carbon atoms. Fungi secure carbon from carbohydrates, especially glucose, and some of them can utilize alcohols and organic acids. Carbon can also be utilized from proteins and some fats.

Nitrogen is the second element that is required. Most fungi are able to utilize amino acids, inorganic nitrogen in the form of nitrates or ammonium. All elements which are in the cell as minor proportions (phosphorus, sulphur) can be utilized in forms of inorganic salts.

#### Alkalinity

The pH limits for common fungi differ greatly in within the range of pH2.2 to 9.6 (Carpenter, 1972). Generally most fungi prefer grow most rapidly between pH5 and pH8 (Carpenter, 1972; Silliker and Elliot, 1980).

#### **Other Factors**

Radiation, air movements and oxygen may also be important. In general growth of fungi is slightly affected by these factors. A fuller explanation can be found in the work of Scott (1957), Gottlieb (1978) and Weersink (1987).

#### Radiation

Radiation may as well a stimulating effect as inhibiting effect on growth of fungi. In particular the ultraviolet part of the spectrum may be harmful (Panasenko, 1967; Gottlieb, 1978). Many fungi are able to reproduce in the dark, and daylight may stimulate reproduction.

#### Air Movements

Moving air, even when it has no drying effect, may slow down the growth of fungi.



## **II APPENDIX for the EXPERIMENTS**

#### **II.1** Temperature during the Experiments

The temperature of the chamber is tried to be kept constant at 20  $^{0}$ C to avoid the effect of the temperature. The temperature fluctuation at the experiment (45-90% RH) is about 0.5  $^{0}$ C (between 20-20.50  $^{0}$ C). Because the fluctuation of the temperature is not extreme and this temperature (+/- 1  $^{0}$ C) change has no great effect on the sorption properties of wool specimen, the temperature during the experiment is assumed to be constant at 20  $^{0}$ C.



Figure 19: Temperature inside the chamber during the experiments





#### **II.2** Appendix for Sorption Isotherm





(c)



#### **II.3** Program for Sorption in MATLAB

```
function w = wc adsorption(x,phi)
w = x(1).*(1-\log(phi)./x(2)).^{(-1./x(3))};
-----
clear
w_offset_at_44 = 2.25;
phi = [0.4377 0.605 0.766 0.952];
PSI = w offset at 44 + [0 0.785 1.976 4.219]./1000./(pi*0.09^2*0.05);
x data = phi;
ydata = PSI;
wsat_ini = 9;
A ini = 0.1;
n_ini = 3;
x0 = [wsat_ini A_ini n_ini];0
lb = [];
ub = [];
wi = wc_adsorption(x0,phi);
options = optimset('MaxFunEval',10000,'LargeScale','off','LevenbergMarquardt','on');
% x_opt = fminsearch(@obj,x0,options,phi,Wdata);
x_opt = lsqcurvefit(@wc_adsorption,x0,xdata,ydata,lb,ub,options);
plot(phi,ydata,'+');
hold on
phi = 0.01:0.01:0.99;
plot(phi,wc_adsorption(x_opt,phi));
hold on
x_opt
```

- 1

Optimization terminated: directional derivative along search direction less than TolFun and infinity-norm of gradient less than 10\*(TolFun+TolX).

x\_opt =

\_\_\_\_\_

#### 6.3072 0.2655 1.3634

By applying the coefficients on the formula, the function becomes as:

$$w = w_{sat} \left( 1 - \frac{\ln(\phi)}{A} \right)^{\left(-\frac{1}{n}\right)} \qquad w = 6,3072 \left( 1 - \frac{\ln(\phi)}{0,2655} \right)^{\left(-\frac{1}{1,3634}\right)}$$







Figure 21 (a, b): Sorption isotherm of sheep wool, in the range 0-100%RH (a), and 40-100% RH (b)



#### **II.4** Hygroscopic Moisture Capacity

For both of the functions; the hygroscopic moisture capacity is found by applying the chain rule in differential equations.

$$y = a \cdot f^{n}(x) \Rightarrow \frac{dy}{dx} = a \cdot n \cdot f^{n-1}(x)$$
  

$$y = \ln(f(x)) \Rightarrow \frac{dy}{dx} = f'(x) \cdot f(x)^{-1}$$
 (Chain Rule)

For the adsorption curve;

$$w = 6,3072 \left( 1 - \frac{\ln(\varphi)}{0,2655} \right)^{\left(-\frac{1}{1,3634}\right)} \Rightarrow$$
  
$$\xi = \frac{dw}{d\varphi} = 6,3072 \cdot \left(-\frac{1}{1,3634}\right) \cdot \left(1 - \frac{\ln\varphi}{0,2655}\right)^{\left(-\frac{1}{1,3634}-1\right)} \cdot \left(\frac{-1}{0,2655} \cdot \frac{1}{\varphi}\right)$$

$$\xi = \frac{\partial w}{\partial \varphi} = 17,42404 \cdot \frac{1}{\varphi} \cdot \left(1 - \frac{\ln \varphi}{0,2655}\right)^{-1.73346}$$

#### **II.5** The **µ**-factor for the Wool

 $\frac{\delta}{\mu_{total}} \cdot A \cdot \frac{\Delta P}{d} = \frac{1.8 \cdot 10^{-10}}{\mu} \cdot (\pi \cdot 0.15^2) \cdot \frac{\Delta P}{0.10m} = \frac{rateper\Delta RH(gr/day/RH)}{86400 \cdot 1000}$ 

 $\delta = 1.8 \cdot 10^{-10} [kg / msPa] or[s]$ : vapor permeability of stagnant air



Figure 22 : Schematic drawing of the aluminum cup



Figure 23 : Half Cross-Section of the Set up in COMSOL



### II.6 Numerical Model in MS Excel



$$g_w^t = -D^t(C_p) \left[ \frac{C_p^t - C_w^t}{X_p - X_w} \right]$$
$$g_e^t = -D^t(C_p) \left[ \frac{C_e^t - C_p^t}{X_e - X_p} \right]$$

The volume of node at "*p*" is:

$$\frac{1}{2} \left( X_p - X_w \right) + \frac{1}{2} \left( X_e - X_p \right)$$

The conservation of mass equation can be written as:

$$\begin{aligned} &\frac{1}{2} \Big( X_p - X_w \Big) + \frac{1}{2} \Big( X_e - X_p \Big) \cdot \Big( C_p^{t + \Delta t} - C_p^t \Big) = \Big( g_w^t - g_e^t \Big) \cdot \Delta t \\ &C_p^{t + \Delta t} = C_p^t + \left( \frac{\Big( g_w^t - g_e^t \Big) \cdot \Delta t}{\frac{1}{2} \Big( X_p - X_w \Big) + \frac{1}{2} \Big( X_e - X_p \Big)} \right) \end{aligned}$$




## Data Fitting by DATAFIT at the Beginning of each RH Step

Sheep Wool and Hygrothermal Properties of Sheep wool insulation



#### • Range 60-75 %RH

Sensors	Distance	Rel. Hum.		
RV1	0.00	0.740		
RV2	0.05	0.635		
RV3	0.10	0.591		
RV4	0.20	0.572		
RV5	0.30	0.559		
RV6	0.60	0.553		



		Equation:			
		3: a+b*x^.5+c*exp(-:	x)		▼ ± ▲ ▼
t Information	Data Table   Model Plot	Residual Scatter Residual	Probability Evaluate		
	_				 
DataFit ver	son 8.1.69				 
Results fro	m project "e:\modele~1	\datafit\60-75df.dft"			 
Equation ID	D: a+b*x^.5+c*exp(-x)				 
Model Defi	nition:				 
Y = a+b*x*	`,5+c*exp(-x)				
Number of	observations = 6				
Number of	missing observations =	0			
Solver type	e: Nonlinear				 
Nonlinear if	teration limit = 250				 
Diverging nonlinear iteration limit =10					
Number of nonlinear iterations performed = 11					
Residual tolerance = 0,0000000001					
Sum of Residuals = -3,21964677141295E-15					
Average Residual = -5,36607795235492E-16					
Residual S	um of Squares (Absolu	te) = 1,418688444304878	E-04		
Residual S	um of Squares (Relativ	e) = 1,41868844430487E	-04		
Standard E	Error of the Estimate = 6	6,87674449213888E-03			
Coefficient	of Multiple Determination	on (R^2) = 0,9943620806			
Proportion	of Variance Explained :	= 99,43620806%			 
Adjusted c	oefficient of multiple de	termination (Ra^2) = 0,99	06034677		 
Durbin-Wat	tson statistic = 3,06056	393043161			
Dograecia	n Variable Deculto				
Variable	Value	Standard Error	t ratio	Proh(t)	
a	1 /9093606317019	0.079475931974413	18 75959207	0.00033	
h	-0.682937744519435	0.046116539718425	-14 80895463	0,00055	
о С	-0,002037744913433	7.50651421097423E-02	-9 978284775	0,0000/	
L .	-0,140021004001001	r,50051421037423E-02	-5,570204775	0,00214	

# Figure 24 : Fitting the data for each RH sensor in the beginning 60-75% RH step, and the regression analysis



• Kange 75-90 %KH
-------------------

0					
Sensors	Distance	Rel. Hum.			
RV1	0.00	0.892			
RV2	0.05	0.796			
RV3	0.10	0.741			
RV4	0.20	0.723			
RV5	0.30	0.707			
RV6	0.60	0.705			



		Equation:						
		3: a+prx .o+crexp(-	xj					
-it Information	it Information   Data Table   Model Plot   Residual Scatter   Residual Probability   Evaluate							
DataEituara	Jan 0 1 C0		1					
DataFit vers	guri o. 1.69 n project "et)medeleut	1) distofit) 75, 00 df d#"						
Equation ID	n projectille, vnoueler-i slovetska 540*over(jiv)	r tuatant v 5-500i. uit						
Model Defin	ition:							
$V = a + h^* v^A$	5+c*evn(-v)							
1 - atb x ;	,510 exp(*x)							
Number of c	hservations = 6							
Number of r	missing observations =	:0						
Solver type:	Nonlinear							
Nonlinear it	eration limit = 250							
Diverging no	onlinear iteration limit =	=10						
Number of nonlinear iterations performed = 1								
Residual to	Residual tolerance = 0,000000001							
Sum of Residuals = 5,55111512312578E-16								
Average Re	Average Residual = 9,25185853854297E-17							
Residual Sum of Squares (Absolute) = 1,80080400915951E-04								
Residual Sum of Squares (Relative) = 1,80080400915951E-04								
Standard E	rror of the Estimate = 3	7,74769645154719E-03						
Coefficient of Multiple Determination (R^2) = 0,9929815106								
Proportion of	of Variance Explained	= 99,29815106%						
Adjusted co	Adjusted coefficient of multiple determination (Ra^2) = 0,9883025177							
Durbin-Wat	son statistic = 3,12043	3480308804						
<b>n</b> .	V . LL D K							
variable	value 1.ccochono7c4100	Standard Effor	t-ratio	PTOD(1)				
a	0 6061000 45106 451	0,93417003596308E-02	10,07020222	0,00034				
0	0,000100040200401	0,001007201704000 9.45700760900739E-00	-13,35525042 9.007027164	0,0009				
L.	-0,708030628470451	0,457 227 020307 30E-02	-9,097027104	0,00201				

## **III APPENDIX for Case Study**

#### III.1 Water Reed (www.riet.nl)

Water reed (In Latin: Phramites australis) is a plant from the family of grass. It is a plant growing in wet places throughout the temperate parts of the world. Water reed is also growing everywhere in Holland as a result of being a "wet" country. It has been used for many centuries to cover the roofs of the houses and windmills.



Reed field

Apart from thatching reed is used to keep in place drift sand along the coast, covering sewerage and to produce garden material like fencing for example.

## **III.2 Durability of a Thatched Roof**

The durability of a thatched roof depends on many things. In general: the dryer the roof, the longer the lifespan will be. The following factors are important for the durability: the slope of the roof and the thatcher's craftsmanship.

The lifespan of the thatched roof depends so much on the slope that the expected lifespan can be given as function of the slope:



25 degrees up to 15 years30 degrees 10-20 years45 degrees 25-45 years50 degrees 45 years and longer

In general every thatched roof should have a slope of at least 45 degrees.



## **III.3** The Construction

There are two basic construction methods in use in Holland: The "traditional open construction", and the closed construction ("the screw roof"),

Both constructions can be made with extra insulation and any finish as wished from the inside. The closed construction is more and more favored because of better insulating (no draft) and fire-retardant properties.

#### The traditional open construction

The traditional open construction is the thatched roof as it has been used for ages. Here the reed is fastened onto rafters. This construction always leaves air layer underneath the thatch. The thatch contributes partly to the insulation because of drafts and there is no definite barrier between outside and inside. To meet Dutch regulations on insulation, extra insulation has to be applied.

When a fire starts in the thatch it only takes minutes before the fire will have found its way to the inside of the house. Oxygen will be sucked through the thatch by the fire from underneath. Once started it is very hard for the fire department to extinguish, because it is very hard for them to reach the fire under the thatch.



Figure 25 : Cross-section of a thatched roof (open structure)





The open character of this roof can be seen from the inside as well. Because the traditional open construction is less fire retardant and has more draft and dust, builders tend to make more closed constructions.

If better insulation is needed, the traditional roof can have extra insulation as part of the construction of the roof by means of a **sandwich panel** mounted underneath the thatch or by additional insulation applied from the inside (afterwards). If insulation is fitted



afterwards, a vapor barrier has to be mounted on the inside of the insulation to prevent condensation of vapor inside the thatch.

#### The closed construction "the screw roof"

The roof is covered first with wooden plating at this type of construction and the reed is fastened by screws on "closed", wooden plating at onto the roof.

The wooden sheet material (19 mm thick because of the length of the screws) can be a triplex or plywood. There exists no air layer underneath the thatch. The whole thatch helps to make a 30 cm thick layer of insulating material. The roof doesn't allow any draft and there is a definite division between outside and inside at this type of construction.

Fire in the thatch cannot draw oxygen from underneath; therefore it is relatively difficult to burn. Even when it burns, it burns not as fiercely and it will burn on the outside.



Figure 26 : Cross-section of a thatched roof (closed structure)

The thatch is not visible from the inside. Sandwich panels can be used inside for better insulation.



#### **Properties of Thatch**

The properties of the reed are taken from the internet site of the Dutch Federation of Thatchers (www.riet.nl).

- Thatch weighs 130-150 kg per cubic meter.
- Average density of a 30 cm thick thatched roof is therefore about 40-45 kg/m<sup>3</sup> (the under construction not included)
- The thermal conductivity of reed: 0,11 0,20 (W/mK)
- The  $\mu$ -factor of reed: 3.



Water reed is a hygroscopic material. Nilsson et. al. (2005) has studied the adsorption equilibrium moisture contents of four natural fibers including water reed. The experiments were done for  $5^{0}$ C,  $15^{0}$ C, and  $25^{0}$ C [56].

Figure 27 : sorption isotherm for water reed

