DISTRIBUTION OF WATER IN FILLING FIBRES VISUALISED BY NEUTRON RADIOGRAPHY

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Neutron imaging (neutron radiography) provides a method to visualise the moisture distribution in textiles, due to sensitivity of neutrons for hydrogen. The method was used to investigate the moisture distribution in filling fibres under practically relevant conditions in duvet. Real time neutron radiography made it possible to follow the dynamics of the moisture transport without disturbing the ensemble during measurement. The distribution of water was rather even over the lyocell fleece, with somewhat higher moisture content on a bottom side (simulating skin face).

In polyester filling, water was localised in two distinct zones, near the “skin face” of the duvet model, and near the upper surface exposed to the environment. Comparing the mass increase of fleeces with known moisture sorption data, it was concluded that in lyocell fillings, most of the water was absorbed by the fibres, whereas in polyester fillings, condensation on fibre surface took place.

Keywords: lyocell, sorption, neutron radiography, imaging, non–destructive testing, visualisation

Introduction

Comfort during sleep is influenced by fibre breathability, that is, the transport of heat and moisture through duvets and bedding textiles (Bartels and Umbach, 2003; Libert et al., 1988). Breathability can be measured by the sweating guarded hot plate instrument (Umbach 1987; ISO 11092). In addition, the ability of textile materials to absorb moisture plays an important role, as this influences heat buffering capacity of textiles (Schuster et al., 2006). Thermoregulation is connected to heat capacity and heat of sorption of moist textiles (Varga et al., 2007).

Measurements with test persons of the climate in bed by Helbig (2006) showed favourable conditions when cellulosic filling fibres were used, as compared to synthetic fibres. An assessment of the effects of bedding materials on sleep comfort by a combined physiological and psychological approach showed improved sleep quality with cellulosic fibres (Moser et al., 2007). Based on Helbig’s (2006) temperature and moisture data, the gradient of heat and moisture in duvet was calculated (Suchomel et al., 2008), and the occurrence of a dew point within a duvet was identified in the polyester fillings. An important point in the context of moisture transport is the water distribution. On the microscopic level, the question arises whether water is present in free water films or droplets on the fibre surface, or absorbed in the fibre structure. Cellulosic fibres absorb water into their structure, whereas on synthetic fibres, water films or droplets are formed on the fibre surfaces. This can be visualised on the level of individual fibres (Schuster et
environmental scanning electron microscopy. The distribution of water on a larger scale, e.g. over the cross-section of a duvet, is more difficult to assess by non-destructive methods.

Neutron radiography (NR) provides an efficient tool for investigation into non-destructive testing as well as for many applications in fundamental research. Neutrons, due to their specific properties, offer a unique probe for materials research and characterisation (Zawisky et al., 2008). When a neutron beam passes through a material, the neutrons will interact with the nuclei of the material (Pel et al., 1993). A neutron beam penetrating the specimen is attenuated by the sample according to the basic law of radiation attenuation. Neutrons are attenuated by some light materials, e.g. hydrogen, boron and lithium, but also penetrate many heavy materials. The attenuation of the neutron beam is determined by the cross-section for scattering and absorption of the nuclei present in the sample. Because of the relatively large scattering cross-section of hydrogen, this method is very sensitive for water. The moisture content can be determined by measuring the transmission through a sample of a given thickness $d$ (Pel et al., 1993).

**Figure 1.** NR assembly at Atomic Institute in Vienna.

At the Atomic Institute Vienna, with a 250 kW TRIGA MARK II reactor, neutron imaging has a long tradition (Zawisky et al., 2008). This low–power reactor possesses a collimated thermal beam with neutron flux of $1.3 \times 10^9$ n cm$^{-2}$s$^{-1}$. The basic experimental layout of NR consists of a neutron source, a collimator functioning as a beam formatting assembly, a detector and the sample, which is placed between the exit of the collimator and the detector (Figure 1).

For the experiments in this work, two types of detector were used: **scintillator** (with camera) and **neutron imaging plate systems (NIP)**. Scintillator / camera system – this type of detector has a neutron-sensitive scintillator screen (which converts the neutrons to photons) and a camera (which can take a picture of the light emitted by the scintillator). Typical scintillator materials are ZnS(Ag)$^6$LiF or ZnS(Cu)$^6$LiF. $^6$Li is a good choice as neutron absorber because it offers the best gamma discrimination ($\gamma$-rays represent a noise in the NR).

In the LiF:ZnS scintillator screen the neutrons are converted to green light with an absorption maximum at 520 nm:

$$^6\text{Li} + n \rightarrow ^3\text{H} + ^2\text{He} + 4.78 \text{MeV}$$

The light emitted by the scintillator is reflected to the CCD–camera by the mirror. The performance of camera can be enhanced by cooling it with LN$_2$ to $-120$ °C (Atomic Institute, Internet Page). Using this detector, 200 µm resolution can be achieved.

The **neutron imaging plate** (NIP) works on a different principle: information is stored in an imaging plate (IP), read with a scanner and afterwards erased by light (Zawisky et al., 2008). Neutrons are absorbed in Gd$_2$O$_3$ (plate) which is uniformly dispersed in the photoactive phosphorus (BaFBr:Eu) layer and an organic binder:

$$n+^{157}\text{Gd} \rightarrow ^{158}\text{Gd} + \gamma + e^- \quad (30–200 \text{ keV conversion})$$

$$n+^{155}\text{Gd} \rightarrow ^{156}\text{Gd} + \gamma + e^- \quad (30–200 \text{ keV conversion})$$

The secondary particles excite the BaFBr:Eu to a metastable state, where electrons are trapped. The information is
stored in locally trapped electron–hole pairs in the phosphor as a latent image. This information is registered by optical simulation with a focused He–Ne laser. During the readout process in the scanner, the trapped electrons are further excited by the red light from a He–Ne laser which causes luminescence of blue light which is finally detected by a photomultiplier tube. Layer thickness (135 µm) and composition have been optimised in spatial resolution and photo-stimulated luminescence. After readout the NIP is erased with bright white light and can be reused many times as long as no mechanical damage occurs. The resolution of IP is 50 µm. Every detector has certain advantages and disadvantages, and hence has specific application (Zawisky et al., 2008).

In fibre science, neutron radiography was applied for the first time by Weder et al. (2004). A selection of clothing systems composed of layers with differing water transport properties was studied to demonstrate the feasibility of this technique. The results are compared to the weights of the individual layers obtained during separate sets of measurements in the same horizontal configuration, as well as in vertical configuration on a heated cylinder. The results of the three measurement approaches agree with respect to layerwise moisture distribution in the different textile combinations, and the radiographic data provide a lateral visualisation of the distribution. A decisive role in fluid water transport is revealed for the innermost two layers (Weder et al., 2004).

Reifler et al. (2006) used neutron radiography to determine the exact water content in aramid-based soft body armour panels. While investigating the ballistic resistance of aramid–based body armour panels under wet conditions, it is important to precisely determine their water content and its chronological development. Using the neutron radiography method, the influence of water amount and location on impact testing as well as its time dependence was shown. In the ballistic panels used, spreading of water strongly depended on the kind of quilting. Very fast water migration could be observed when the panels were held vertically. Some first results regarding the water distribution in wet panels immediately after the impact are presented. On the basis of the presented results, requirements for a standard for testing the performance of ballistic panels in the wet state were deduced (Reifler et al., 2006).

The aim of our work was to visualise the moisture in lyocell filling fibres (fleece) by radiographic observations. Static and dynamic experiments were done on lyocell and polyester filling fibres. In the static experiments, fibres with defined moisture content were enclosed in the bags. The dynamic experiments were performed by using a humidifier which enabled continuous moisture evaporation with increasing temperature. The transmission data of both, lyocell and polyester fleeces were calculated from the grey–values obtained from NR images. Additional gravimetric measurements of the total mass increase were performed to support the NR data.

Materials and Methods

Materials

For examination with NR, lyocell, and polyester fleeces were prepared on the carding machine having a fleece weight of about 200 gm⁻². Polyester fleeces were thicker (6 cm) than lyocell (4 cm) due to the higher stiffness of polyester fibres. The Table 1 shows the data of the samples used for neutron radiographic examinations.

Static and dynamic experiments

Two kinds of experiment were performed: static and dynamic. In the static experiment, fibres were enclosed in sealed plastic bags containing 10 and 30 wt% water (Figure 2). The images were taken using the IP detector with exposure time 50 min to obtain high–quality images.
Table 1. Fleeces used for neutron radiography.

<table>
<thead>
<tr>
<th>Type</th>
<th>Characteristics</th>
<th>Supplier</th>
<th>Additional information</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lyocell fill</td>
<td>Fleece, 200; thickness 4 cm</td>
<td>LENZING AG, AT</td>
<td>Fibres 6.7 dtex; 60 mm, unfinished</td>
</tr>
<tr>
<td>Polyester fill</td>
<td>Fleece, 200; thickness 6 cm</td>
<td>ADVANSA, TR</td>
<td>Fibres 6.7 dtex, 60 mm</td>
</tr>
</tbody>
</table>

Dynamic neutron radiography or real time neutron radiography means that images can be made continuously after a fixed time interval depending on the exposure time chosen. The process (water sorption) can be followed in real time without any disturbance to the experimental arrangement. Using the scintillator detector, a time between two images can be decreased to 53 seconds while the use of the imaging plate consumes time for reading and erasing information.

For the dynamic experiments with textile fibres, a humidifier was placed between the neutron source and the detector. This was a steel tray with a layer of wetted viscose fleece. The experimental setup was adjusted a lot until the optimal experimental conditions were found. First, the aluminium box was on the humidifier between fleeces, then the fleeces were packed into pillows. To avoid liquid water being transported from the humidifier to the test fleece by direct contact of the fleece with the humidifier, aluminium wires were put below the pillows as a support. Finally, the best results came when a polypropylene fly–screen was put below the fleeces.

**Dynamic Experiments on Fleeces in an Aluminium Box with Humidifier**

The first experiments were done by putting the lyocell and polyester fleeces in the aluminium box, and taking the images with the scintillator. Unfortunately, this experiment was not successful due to the thick aluminium plate. In general, aluminium is transparent for neutrons if the plate is thin enough. In our case, the plate was too thick to be transparent for neutrons (Figure 3).

**Dynamic Experiments on Pillows with Humidifier**

Fleeces were enclosed in stitched pillow–cases to simulate real conditions in a duvet. Pillow-cases in a size 20 by 20 cm and a thickness 4 cm for lyocell filling and
6 cm for polyester filling were made with a cover from a satin weave of lyocell micro fibres (0.9 dtex). Aluminium wires were placed on the humidifier walls at a height about 1.5 cm in order to see the bottom of the pillows. The purpose was to raise the samples so that the lower part of the pillow wasn’t hidden behind the walls. The fibres were then placed on the top of the wires. A series of 90 images was started at room temp using the scintillator detector interval 53 s (40 s exposure time and 13 s grabbing time). The temperature increased to 35 °C and afterwards to 50 °C. Additional images with the imaging plate detector were taken at temperature 50 °C to compare the quality of the images taken with scintillator / imaging plate detector with an exposure time 10 min (Figure 4).

**Analysis of NR images**

The analysis was done by measuring the grey–values. The lower the value, the higher is the quantity of absorbed water per sample. In the case of small absorption and low scattering, the neutron attenuation can be approximated by the exponential law:

\[ I = I_0 e^{–\Sigma t} \quad (Eq. 1) \]

where \( I \) is the intensity of the transmitted neutrons after penetrating a distance \( t \) into the target. In our case, \( t \) is the thickness of the sample. \( I_0 \) is the intensity of the beam incident on the sample. \( \Sigma \) is the macroscopic cross–section.

![Figure 4. Fleeces enclosed in pillows.](image)

**Final Dynamic Experiments with Humidifier and Imaging Plate Detector**

The experiments were started with the humidifier at ambient temperature and then increasing its temperature to 35, 50 and 60 °C. The stable temperature of 35 °C was achieved after 40 min, 50 °C after 60 min and 60 °C after 90 min. Only the imaging plate detector was used. Exposure time was 10 min (Figure 5). The grey–value calculations were made from these images.

![Figure 5. Lyocell and polyester fleeces taken with IP detector at the beginning and end of the experiment.](image)
and $\sigma$ the microscopic cross-section in cm².

This investigation was mainly concerned with the water content, therefore the intensities behind the wet sample to the dry sample were normalised in order to determine

$$T_{\text{water}} = \frac{I_{\text{wet}}}{I_{\text{dry}}} = \exp(-\sum_{\text{water}}t) \quad (\text{Eq. 4})$$

From $\sum_{\text{water}}$ the absolute density $\rho_{\text{water}}$ could be evaluated.

**Results and Discussion**

**Static Experiments**
Carded lyocell and polyester fibres were sprayed with water before enclosing into plastic bags. The fibres contained 10 and 30 wt % water. Then neutron imaging was done under stable conditions, where the exposure time was increased to 50 min – this giving maximum hydrogen sensitivity and 16 bit image gradation.

![Neutron radiographies made with an imaging plate with an exposure time of 50 minutes: left—with 10 wt% water content; right—with 30 wt% water content. The water is very homogenously distributed within the lyocell fibre. The lyocell samples with 30 wt% water look greyer in comparison with samples containing 10 wt% water. On polyester fibres the condensation in the centre of the fleece occurs. In the polyester sample with 30 wt% water, water is condensed in the bottom part of the sample. With the static experiments, the distribution of water in equilibrium conditions can be visualised enabling high-resolution images due to the high exposure time and imaging plate detector.](image)

**Dynamic Experiments on Fleeces in Aluminium Box with Humidifier**
For the dynamic experiments, the humidifier filled with a few wet viscose non–woven layers was placed in the NR–assembly between the neutron source and the detector. In the images, this is hidden behind the wall of the humidifier. An additional lyocell film (8 µm thickness) was put between wet non–woven in the humidifier and the examined fleeces to ensure slow and homogenous moisture evaporation. The fleeces (lyocell and polyester) were put on the humidifier, which simulates the humid skin of a sleeping person. To simulate the temperature differences between the skin and a cool sleeping room or outdoor environment, it was not possible in the reactor room to cool the environment. So, the humidifier temperature was raised to achieve temperature differences. The temperature was increased from 24 (room temperature) to 35, 50 °C, and 60 °C. The trials were therefore operating at a higher level of temperatures and absolute humidity; however, the principle conditions are realistic as a model. Figure 7 (A) shows the experimental arrangement showing the aluminium box (Al–box) placed close to the scintillator, (B) NR of the fleeces inside the Al–box and (C) NR of the fleeces without the Al–box. On the lower side the dark region shows the walls of the humidifier. In the middle of the Al–box, there was a double wall of 6 mm thickness. This Al–layer is visible to neutrons and the area covered in the image is quite large. So the experiment was then performed without Al–box.

**Dynamic Experiments on Pillows with Humidifier**
In order to set the experiment in conditions close to practical use of substrates, the lyocell and polyester fleeces were enclosed into pillows (made from lyocell fibres 0.9 dtex in sateen weave) and placed on the humidifier (Figure 4).
The wires were placed on the humidifier (below the pillows) in order to be able to see the bottom of the pillow on the NR images.

The experiment started at room temperature 24 °C. A few images were made in these conditions. Then the thermostat was switched to a temperature of 35 °C. A series of 90 images was started at room temperature. The temperature became stable at 35 °C in about 15 minutes and was maintained for half an hour. After that the temperature was increased to 50 °C (which took about 15 min). Again this temperature was maintained for half an hour. During all this time, images were taken at an interval of 53 seconds (the exposure time being 40 s with a grabbing time of about 13 s). Figure 8 shows images of fleeces enclosed in pillows: A–taken at room temperature after 53 s (beginning of the experiment); first image of the time series, B–at 50 °C after 80 min; image number 90 of the time series and C–at 50 °C taken with imaging plate detector after 270 min exposed for 10 min.

It can be seen from the images that no considerable difference in the water content in the lyocell fibres at the beginning and at the experiment and 90 min later could be observed. The moisture is absorbed in lyocell pillow–cases. The condensation of water is visible on the bottom of polyester filled pillows. It is unclear if the condensation occurs within the pillow or on the wires placed on the humidifier below the pillow.
It is assumed that water is absorbed by the lyocell covers therefore most probably moisture condenses on the Al–wires. It was then decided to perform further experiments without pillows. Images obtained using the imaging plate detector have a much better quality. After exposure, time is required for scanning and erasing the plate before it can be reused. So the repetition interval between two such images would be about one hour.

**Figure 9.** NR images taken with imaging plate detector: Humidifier temperature: starting at room temperature (A), at 35 °C (B), 50 °C (C) and 60 °C (D).

**Figure 10.** Neutron transmission of lyocell fleece at various humidifier temperatures.

**Figure 11.** Neutron transmission of polyester fleece at various humidifier temperatures.

**Final Dynamic Experiments with Humidifier and Imaging Plate Detector**

The experiment setup was the same as for preliminary experiments as shown in Figure 5. Lyocell and polyester fleeces were placed next to each other on the humidifier and the experiment started at room conditions (25 °C, 40 % RH). The temperature of the humidifier was raised to 35, 50, and 60 °C, respectively. The imaging plate detector was used with exposure time 10 min.
From Figure 9 it can be seen that the temperature has a detectable influence on the water content. The higher the temperature of humidifier, the higher is the water content and the lower is the overall neutron transmission. Time of the sample at a certain temperature is not a considerable influencing factor because the grey–values do not differ a lot when exposing the sample at 35 °C for a longer time period. In Figure 10, a line profile was taken vertically along the height of the sample starting from the bottom to about 2 cm above the lyocell sample. The height of the lyocell sample was 4 cm and that of the polyester sample was 6 cm. The transmission was calculated and plotted relative to the height. Lower transmission corresponds to higher value of moisture content. So we see that there is higher moisture content on the lower part of the sample which is close to the humidifier with both fibre types. As the temperature increases, the transmission decreases and hence the moisture content increases. There is a non-linear relationship between the transmission and the water content. With lyocell fibres, the decrease in transmission is quite even over the cross-section of the fleece, with somewhat higher decrease in the bottom part. With polyester, the decrease in transmission is concentrated on the region near the skin surface, and the height of approximately 5 cm, near the surface exposed to the environment. The lyocell sample changed its shape a little bit during the experiment. In this Lyocell fibres become soft after moisture has been absorbed and therefore fleece will change shape (collapse in the centre).

The neutron images support the study of swelling/condensation on hygroscopic/hydrophobic fibres by environmental scanning electron microscopy (ESEM). In those experiments, the water vapour pressure was increased in the ESEM chamber from 3 to 7 Torr at 5 °C, which corresponds to 40 to 100 % RH. From the starting point, pressure raised in 0.2 Torr steps. Strong swelling of lyocell fibres started at 5 Torr (75 % RH) and the condensation on polyester at saturated atmosphere 7 Torr (100 % RH). (K. Varga, Doctoral Thesis, 2009).

**Gravimetric Analysis**

In order to compare the transmission calculations from neutron images, the gravimetric analysis on the fleeces was performed in Lenzing AG lab by the same procedure like at Atomic Institute. Lyocell and polyester fleeces were placed on the humidifier filled with wet viscose fleece. To ensure slow and continued moisture evaporation, Lyocell foil (8 µm thickness) and fly-screen (polypropylene) were putted on the wet fleece.

After placing the CLY/PES fibres on humidifier, the thermostat was switch on to 35, 50, and 60 °C, respectively. For every experiment new CLY/PES fibres were used. The time needed to achieve stable temperature exceeded 40 min for 35 °C, 60 min for 50 °C and 90 min for achieving 60 °C. After wetting, fleeces were dried at 80 °C for 60 min in order to calculate the water content in the materials. The water content in the fleeces at various temperatures is shown in Figure 12.

The gravimetric data obtained by the same procedure like neutron images support the radiographic results. As the temperature increases, fleece samples take up more water. Therefore, the transmission of neutrons through the sample decreases with increasing water content.

Sorption isotherms (Figure 13) and water retention (Table 2) show that polyester fibres do not take up a substantial amount of water vapour or liquid water, whereas lyocell can absorb water vapour and liquid water. Comparing these numbers with the measured uptake in Figure 12, it can be concluded that in case of polyester fillings, the water in the fleece must be present mostly as free water in films of droplets on the fibre surface, whereas in the Lyocell
fillings the water is mostly absorbed into the fibre structure.

![Figure 12. Total water content in fleeces after dynamic experiments.](image)

![Figure 13. Water vapour sorption isotherms of lyocell fill and polyester fill fibres at 25 °C.](image)

**Figure 12.** Total water content in fleeces after dynamic experiments.

**Figure 13.** Water vapour sorption isotherms of lyocell fill and polyester fill fibres at 25 °C.

**Table 2.** Moisture regain and liquid water retention of the fibres used (in percent based on dry matter, indicative values).

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Water retention value</th>
<th>Moisture regain at 90 % RH</th>
<th>Moisture regain at 100 % RH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lyocell fill</td>
<td>70</td>
<td>22.8</td>
<td>33</td>
</tr>
<tr>
<td>Polyester fill</td>
<td>3</td>
<td>1.3</td>
<td>2</td>
</tr>
</tbody>
</table>

**Conclusions**

In this study, the water distribution in lyocell and polyester filling fibres under simulated practical conditions was visualised by the non-destructive method of neutron imaging (neutron radiography). Raising the temperature difference between a humidifier simulating the human skin, and the cooler environment, it was shown that an increasing amount of water was taken up by the lyocell fibres, visualised by lower neutron transmission and confirmed by the weight of the fleece. From the comparison of moisture uptake measured by fleece weight and the known data of moisture and liquid water absorption in fibres, it can be concluded that in polyester fleece condensation on surfaces occurred under applied conditions.

The distribution of water is rather even over the cross-section of lyocell feeling, with somewhat higher moisture content near the “skin face” of the duvet model. In polyester filling, water is localised mostly in two distinct zones, near the “skin face” of the duvet model, and near the upper surface. The neutron radiography method presented in this study supported the known images from environmental scanning electron microscopy where swelling of hygroscopic and water condensation on hydrophobic fibres was visualised on the fibre cross-sections.

To get more insight into water content in the fleeces during the neutron imaging, additional calculations are necessary based on the grey-values of neutron images.

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**References**


