SURFACE ROUGHENING BY WATER: GLOSS RELAXATION PROCESSES

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ABSTRACT

A new instrument is used to measure simultaneously the gloss and the moisture content of a sample of paper as it is subjected to changes in relative humidity. Measurements made with five different papers confirm the overall inverse relationship between gloss and moisture content reported in the literature, although there are reversals (gloss increasing with moisture content) which are unexplained. A significant result of this work is that gloss lags behind moisture content after a step change in RH, apparently reflecting the continuing structural rearrangement of the fibers in the bulk of the sheet as moisture diffuses into fiber walls. This explains why, in order to stabilize sheet gloss to moisture effects, it is not enough to stabilize the surface only, as is done in surface treatment processes involving a thermal gradient.

INTRODUCTION

Surface roughening is a prevailing issue for coaters and printers. It inevitably occurs when paper is contacted with water in processes such as coating and offset printing or simply when it is exposed to high relative humidity. Surface roughening can be detrimental to print quality. Aspler’s review[1] introduces publications about sheet roughening in heatset offset printing of higher quality mechanical paper grades, water-based gravure and water-based flexography.

Gloss is one of the most sensitive properties indicative of surface roughening. Air-leak methods are rather less sensitive because, during the test, the surface is subjected to a load and is compressed and deformed. This compression and deformation increase with the moisture content and roughness measurements may become misleading. A contactless gloss measurement on the other hand, is quite suitable and is frequently used to characterize surface structural changes.

This paper describes a special glossmeter, referred to as "GlossMachine", which allows the concurrent measurement of gloss and moisture content of a paper sample subjected to varying relative humidity. Since moisture sorption-desorption processes are the cause of the changes in surface structure, the concurrent measurement of these two parameters provides useful information on the roughening processes in addition to that on the roughened surface.

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EXPERIMENTAL

Design of the GlossMachine

Figure 1 shows a sketch of the sample chamber of the GlossMachine. A paper sample is attached to a metal stage with two-sided adhesive tape. The whole sample stage is directly supported on a top-loading balance so that sample weight can be converted into moisture content. The chamber ceiling is equipped with a light source (LED of visible red light) and detector for gloss measurement set at equidistant angles (75°). 75° was selected in accordance with TAPPI Test Method T480. The illuminated area on the sample used for gloss reading is about 10 mm diameter. Humid air is supplied by small openings made on both the sides of the chamber. This air inlet is connected to an air generator providing air at a controlled relative humidity (RH). Figure 2 shows a sketch of the air generator. Bubbles coming from the sparger are saturated. The air-flow rate is 1.0 L/min. At this rate it takes only three seconds to supply a volume of air equivalent to the sample chamber capacity. After the sparger cell, the saturated air goes through a reheater cell to raise its temperature to the sample chamber temperature. All air ducts are well insulated to avoid condensation. But, for more exact RH, the temperature of the sparger cell is adjusted to that in the sample chamber.

The ratio of water vapor pressure at temperature $T_1$ in the sample chamber to that at $T_0$ in the sparger cell determines the RH. The RH was calculated based on the vapor pressure table of the atmospheric pressure. Practically, a target RH is attained by lowering only $T_0$. $T_1$ was basically room temperature, within 2.5°C. For 0% RH, air was run through a column of Silica gel.

Data acquisition of the GlossMachine gloss (GM-gloss), weight and RH are all made methodically by a single software. Resistance temperature detectors and Peltier junction modules are used to trace the temperature changes quickly and to raise or lower the temperatures immediately through feedback loops.

A metal plate is used as a reference to compensate for possible electrical drift in the voltage (GM-gloss) output. This reference system also permits to eliminate the change in the detector output resulting from the absorption of light in the near infrared band (The LED emits near infrared light in part.) by moisture. The reference plate is moved back and forth across the light path by a step motor. The ratio of paper gloss to that of the reference plate provides a relative gloss value that was stable over a long period of time.

Samples

To check the reproducibility of the GlossMachine, a sample of supercalendered, uncoated and filled wood-containing paper (A) was used. Additionally, softcalendered wood-containing base paper for LWC (B), softcalendered wood-free base(C), softcalendered wood-containing coated paper (D) and supercalendered wood-containing coated paper (E) were used. Table I lists TAPPI gloss, Parker Print-Surf roughness and basis weight of the papers.

The measurements were made on the felt side, unless mentioned. The crossmachine direction was aligned parallel to the plane of incidence and reflection.
**Measurement Procedure**

At first, the sample stage is tared with an adhesive tape, without paper. Then, a paper sample is attached, folded around the edge of the sample stage to cover most of the backside. This is to provide as large a sample surface as possible to gain sensitivity in weighing. The sample chamber and the balance are enclosed in a plastic bag until the end of the measurement to prevent cooling drafts. For about 1 min, at the beginning of the run, the sample was weighed with no air supplied. Then, air set at the desired RH is sent at a rate of 1.0 L/min. This causes a deflection of 0.04 g due to air flow pressure for which a correction was made to the weight measurements. Gloss and weight were measured continually every 6 s for the first 10 min after every RH switch, and then every minute. The RH schedule most often used was 4 h at 0 % RH, 3 h at 90 %, and 3 h at 0 % sequentially. To look at RH cycling effects, the last two steps were repeated. Moisture content of paper was calculated on the dry basis as follows:

\[
MC(\%) = \frac{W - W_0 - W_a}{W_0} \times 100
\]

where, \( MC \): moisture content (dry basis)
\( W \): sample weight under measurement
\( W_0 \): sample weight after running dry air (through a silica gel column) for 4 h
\( W_a \): increase in weight due to pressure from air flow

TAPPI gloss (75 °) was also measured. Measurements were made while the sample was still held on the stage as it was in the GlossMachine.

**RESULTS AND DISCUSSION**

**Reproducibility of GlossMachine**

*Figure 3* shows two typical successive runs of sample A, where gloss and moisture content are monitored versus time. Gloss is expressed as a relative value (see experimental), so the values are not comparable to TAPPI gloss. The RH history was: 0 % for initial 4 h, subsequently, 90 % for 3 h and then 0 % for 3 h. The results of the two runs agree fairly well in terms of both gloss and moisture content and the small difference in the gloss curve is attributed to a small difference in the initial gloss values (36.9 versus 38.0). Also, the relative vertical positions of the sample surface, the reference surface, the light source and the detector may not have been always exactly the same. This small variation from measurement to measurement may cause additional error of up to about 4 %. Therefore, it would not be accurate to compare details of the gloss change between samples unless the gloss could be connected to the absolute gloss as TAPPI gloss. However, the gloss change with time of a sample, once mounted, is considered to be well scaled because the relative locations of the components remain constant during the measurement.

**Correspondence between GlossMachine gloss and TAPPI Gloss**

*Figure 4* shows the relationship between gloss as measured by GM- and TAPPI gloss. TAPPI gloss was measured on every sample used both before and after GlossMachine runs. The two values were very consistent, even if there were marginal deviations because of varying relative positions of the
GlossMachine components are assumed to occur at random. Namely, for example, the position of the light source and detector varies marginally relative to that of the sample every time the sample chamber lid is closed. The relationship suggests that approximate conversion to TAPPI gloss would be easy.

**Gloss: surface or bulk property**

**Figure 5** shows the gloss relaxation and the moisture content change as a function of time for the supercalendered, uncoated and filled wood-containing paper (A). The sample was dried at 0% RH for initial 4 h then air at 90% RH was sent for 12 h. After switching to 90% RH, the moisture content quickly increased and continued to increase even after 12 h. Gloss decreased also but its initial rate of decrease was slower than the rate of moisture pickup, as **Figure 6** shows. Gloss is predominantly determined by the refractive index and the surface shape[2]. Considering this, one might expect that changes in the surface structure of this uncoated paper would take place immediately as water molecules condense on the surface fibers. In other words, one might have expected surface changes to take place ahead of the bulk moisture pickup. The fact that gloss lags behind moisture pickup and is still decreasing well after the paper surface can be considered to be at equilibrium with the new RH (a few seconds considering that humidity measured by a sensor using a thin cellulose acetate film stabilizes within 10 seconds) suggests that the surface continues to reflect the reconfiguration of fibers across the bulk even after a long time. This roughening has been ascribed to fiber swelling, stress relaxation and shape recovery of the fibers in the bulk as water diffuses into fiber walls[3]. In this sense, gloss relaxation can be considered, paradoxically, as a "bulk" process. This finding is novel and helps explain the rather poor efficiency of calendering processes using a thermal gradient(TG), such as TG calendering or softcalendering, in stabilizing the gloss to moisture effects[4].

**Effect of RH cycling**

**Figure 7** shows the gloss relaxation and moisture content change as RH was varied cyclically (thick lines). The sample used for these data was also sample A. In the first drying step, the gloss decreased as the moisture content decreased. (Indeed, the initial drop in gloss, during the very first drying step, may seem abnormal since gloss normally decreases with increasing humidity. It should be noted, however, that with the other 4 papers tested, gloss always increased with drying and decreased with moistening.) In the subsequent exposure to 90 % RH, the gloss dropped substantially. In the drying step from 90 % to 0 % RH, it increased and leveled off quickly, but to a value (86.9) that was well below the initial gloss (111.9). After the second cycle, the gloss level was even lower (80.4) than after the first one, though the loss in gloss was less than in the first cycle. Such irreversibility is often observed in cellulosic-water phenomena even though it is still incompletely understood[5]. Thus, gloss reduction was accompanied by irreversible changes as if mechanical relaxation of internal stresses had occurred.

The thin lines in Fig. 7 show the results of an experiment where the exposure to 90 % RH was of the same total length (6 h), but in only one cycle. The drying periods were the same, 4 h for the initial drying, 3 h for the redrying. In this case, the gloss recovered to 81.3, a value lower than that after the first cycle but higher than after the second cycle of the other run, even though the initial gloss was lower (109.1). This indicates that 2 cycles of shrinkage and expansion of the fibers caused greater irreversible roughening (28 % gloss reduction), than one cycle (25 % gloss reduction), for the same total exposure time.
Gloss versus moisture content

Figure 8 shows the relationship between gloss and moisture content using the same experimental data as in Fig. 7. It is not a simple relationship of gloss decreasing when moisture content increases. As pointed out earlier, in the first drying step, the gloss unexpectedly decreases first as moisture content decreases. It then increases slightly as moisture content increases up to about 5% (the first 4 or 5 min). Then it decreases linearly with increasing moisture content. The same phenomenon is observed in the succeeding two cycles, but the slope is lower in each successive cycle. Note that during the drying from 90% RH to 0% RH, the gloss first decreases with decreasing RH (instead of increasing) and after the moisture content decreased to about 8%, it increases continuously until the sample is dry, in a sigma-shaped curve. We do not understand these apparent reversals of the expected gloss-moisture content relationship but suspect that they depend on the conditioning history. Since gloss accompanies dimensional changes, it is interesting to compare our results with those of Uesaka [6] who observed a similar hysteresis in hygroexpansion strain with moisture content.

Other types of paper.

Figures 9 and 10 show results obtained with the other four papers (samples B to E) examined. The same behavior is observed - the gloss decreasing with increasing moisture, with the gloss lagging behind moisture pickup - except for the distinct anomalous reversals. It was difficult to discuss the influence of the coating and fiber sources.

CONCLUSION

A new instrument is used to measure simultaneously the gloss and the moisture content of a sample of paper as it is subjected to changes in relative humidity. Measurements made with five different papers confirm the overall inverse relationship between gloss and moisture content reported in the literature, although there are reversals (gloss increasing with moisture content) which are unexplained.

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REFERENCES

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<table>
<thead>
<tr>
<th>Kind of paper</th>
<th>TAPPI Glossa)</th>
<th>Roughnessb) (µm)</th>
<th>Basis weight (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Supercalendered, uncoated and filled wood-containing paper (A)</td>
<td>35.3 (CD)</td>
<td>1.6</td>
<td>51.1</td>
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<tr>
<td></td>
<td>38.9 (MD)</td>
<td></td>
<td></td>
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<tr>
<td>Softcalendered wood-free base paper (B)</td>
<td>28.9</td>
<td>2.8</td>
<td>77.9</td>
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<td>Supercalendered wood-containing coated paper (D)</td>
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<td>1.6</td>
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<td>Softcalendered wood-containing coated paper (E)</td>
<td>45.7</td>
<td>1.4</td>
<td>46.7</td>
</tr>
</tbody>
</table>

a) 75 ° gloss on the feltside in the crossmachine direction.
b) By Parker Print-Surf at a load of 0.1 MPa with soft backing.
Figure 1  Sample chamber of Glossmachine.

Figure 2  Air generator to provide controlled relative humidity.
Fig. 3 Two Glossmachine runs exhibiting sufficient reproducibility. Same line pair was obtained at one run. RH was 0 % for the initial 4 hours; subsequently 90% 3 hours; 0 % 3 hours.

Fig. 4 Relationship between Glossmachine gloss and Tappi gloss both at 75 °C.

\[ G_{GM} = 2.54 \times G_{TAPPI} + 9.65 \]

\[ R^2 = 0.983 \]
Fig. 5  Gloss relaxation and moisture content change with time. RH was switched from 0 to 90%.

Fig. 6  Gloss relaxation and moisture content change on exposure to high humidity (blow-up of the previous figure around the humidity change).
Fig. 7  Influence of humidity cycling on gloss relaxation vs. continuous humidifying.

Fig. 8  Gloss change with moisture content for supercalendered, uncoated and filled wood-containing paper (A).
Fig. 9  Gloss relaxation and moisture content change for wood-free(WF) and wood-containing(WC) basepapers.

Fig. 10  Gloss relaxation and moisture content change for soft(SOFT)- and super(SUP)calendered coated wood-containing papers.