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Non-destructive high resolution measurements of spatial filler content distribution in paper

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KEYWORDS: Paper properties, Variations, Filler content, Spatial, Grammage, Image registration, High resolution, Point-wise

SUMMARY: In this work, a non-destructive method is presented that enables the measurement of filler content in paper with high spatial resolution. The method uses an X-ray fluorescence setup that enables high resolution measurements of calcium content in the paper, together with a beta radiography measurement method, to assess the local filler content in the paper. An image registration method is used to combine the two measurement maps, and a calibration polynomial is applied to the point-wise values in order to calculate the local filler content.

The measurement methods show good accuracy. The grammage and the calcium content can be measured at a spatial resolution of 0.1 mm x 0.1 mm but the resolution for the filler content map was in this work chosen to 1 mm x 1 mm in order to minimize image registration errors.

The method is illustrated using two paper samples, a laboratory paper and a commercial 80 g/m² copy paper. From the methods used in this work, a difference is shown between the two paper samples in how the filler content distribution is related to the paper formation.

With the help of image registration, point-wise measurements of filler content from both sides of the samples can be compared.

The method can be used together with other high resolution measurements in order to analyze the simultaneous interrelation between different paper properties. The high resolution measurements of filler content will be particularly valuable for the analysis of the underlying causes to optical variations in paper and print.

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The quality impression of a paper is strongly affected by the small-scale variations in the paper structure, which result in disturbing effects such as gloss variations, opacity variations, and print mottle. One approach to identify the causes to such problems is to use image registration to combine 2D measurement maps of different paper properties and of optical response. The standard method to assess the filler content in paper from ash residue (ISO 1761 2001) gives no information of the lateral distribution of filler particles in the paper. The

lateral variations of filler content are expected to give a large impact on the optical variations of paper. With the possibility to acquire 2D maps of local filler content in paper, the filler content distribution at 1 mm² measurement scale can be compared to the grammage distribution at the same scale, which in turn may be used in the analysis of the possible causes of optical variations in paper.

In order to determine local filler content, i.e. the quotient between filler weight and total material weight at 1 mm² scale in paper, both the filler amount and grammage need to be measured at high resolution.

Rindby et. al. (1989) presented a method to obtain micro beams of X-rays with a diameter of a few micrometers and with intensity sufficient for trace element analysis. One application of this is to detect calcium atoms in paper at high resolution. The most common filler in paper is calcium carbonate, so by measuring the calcium content in the paper at high resolution, the point-wise filler amount can be assessed.

A traditional way to measure the formation of paper is to use β -radiography, where a uniform ¹⁴C source is used with an X-ray film for detection. This method has often been used as a reference method in paper formation analysis, but has the disadvantage of long exposure times and a complicated film developing process. A storage phosphor screen (SPS) method has been introduced for the beta radiography measurement of paper that requires shorter exposure time and eliminates the need for a film developing process (Keller, Pawlak, 2001). The storage phosphor screens are scanned with a specially designed scanner, and can be reused indefinitely. Avikainen and Erkkilä (2003) made a comparison between traditional beta radiography and SPS measurement techniques. They found that the SPS method provides a viable and reliable alternative to traditional beta radiography in analyzing the formation of paper if the stochastic background noise has been compensated for properly.

Point-wise combination of calcium measurements and beta radiography images is made with image registration, also called matching, where 2D images from the two different measurements are aligned in order to combine the two datasets for each location on the sample. Examples of works where image registration has been used to combine measurements from different devices in paper applications are the work by (Kajanto, 1989; Mettänen et al. 2007; Chinga, Syverud, 2007).

A work that includes a description of the image registration methods in detail has been presented by (Hirn et al., 2008). They proposed a method that enables the combination of paper property maps measured with different resolution. They found that the registration error was minimized when the registration was made at high resolution and the rescaling to target resolution was made subsequently.

Aim of the present study

We here present a measurement method that enables non-destructive high resolution measurements of the spatial filler distribution in paper using X-ray fluorescence and beta formation. To illustrate the method, a laboratory paper and a standard commercial 80 g/m² copy paper have been measured and compared.

Materials and Methods

Equipment and procedure

The procedure to assess the filler content at 1 mm² spatial resolution in paper is shown schematically in Fig 1. A high resolution X-ray fluorescence method, described in detail below, was used to detect the calcium atoms of the filler particles, and a beta radiography method was used to assess the grammage distribution in the sample. The point-wise filler content was calculated from the two measurements with the help of a calibration polynomial that was derived from standard measurements on a designed set of paper samples with three levels of grammage and filler content.

Paper samples

The method presented in this study was applied on two paper samples: a laboratory paper produced on a small experimental paper machine (XPM), and a commercial 80 g/m² copy paper. The XPM is a scale model of a paper machine with all process steps such as closed loop white water system, wire- press-, and dryer sections represented. Apart from the size, the main difference compared to a full scale paper machine is the production speed. The XPM runs with less than 2 m/min, while a full scale paper machine has a production speed of over 1000 m/min. A pulp mixture of two chemical pulps was used when producing paper samples in the XPM. It consisted of 70% hardwood and 30% softwood, refined to 26°SR. In order to make the XPM paper comparable to the commercial paper, the target grammage of the XPM paper was set to 80 g/m². Calcium carbonate was used as filler in both of the paper samples used. The filler content of the commercial copy paper was 23.4% and the filler content of the XPM paper was 22.4%. In contrary to the commercial paper the XPM paper did not contain any fluorescent whitening agent or dye.

High resolution calcium measurements – X-ray fluorescence

The X-ray fluorescence (XRF) measurement setup for measuring the local calcium amount in the sample is based on a chromium X-ray source that bombards the sample with X-rays. A thin glass cone is mounted on the emitter window on the X-ray tube so that the X-rays are focused to a spot with a diameter of 0.1 mm (Rindby et. al., 1989).

When an X-ray hits an atom of the material, an electron of the atom can be excited.

When a higher energy electron fills the hole, a fluorescent X-ray is emitted with an energy that is characteristic of the atom. This can be used to identify the atoms in the material.

Fig 1. Overview of the process of calculation of local filler content from measurements of grammage and number of calcium detections.

Fig 2. Measurement geometry for the XRF setup.

The fluorescent X-ray photons are collected with a nitrogen-cooled energy-dispersive sensor and the resulting signals are counted in a pulse processor. By counting the pulses of the photons with characteristic energies corresponding to the calcium atoms in the material, the number of detected filler atoms can be counted at the measured spot.

The X-ray source is mounted so that the X-rays hit the sample in the normal direction and the detector registers fluorescent X-rays from an angle of 45 degrees to the sample surface. The setup has a high sensitivity to the distance between sample and detector. Therefore, the paper is pressed between a measurement frame and a plastic plate in the sample holder in order to keep it flat (Fig 2). The sample holder is placed on a stepper table, enabling the acquisition of a 2D measurement map of calcium content in the sample with a spatial resolution of 0.1 mm.

In order to translate the number of calcium detections to filler content, a set of nine Formette samples were produced with three levels of filler content between 21% and 30%, and three levels of grammage between 71 g/m² and 99 g/m². The samples were produced in a Formette Dynamique to ensure good formation. In order to derive a calibration polynomial, the mean values of the calcium measurements of each of the nine samples were related to grammage and filler content of the samples, assessed according to standard methods (ISO 536 1995; ISO 1762 2001). The polynomial was derived using a partial least-squares method. The resulting calibration polynomial was the following:

$$f = 4.4 - 1.8 \cdot 10^{-1} \times w + 3.8 \cdot 10^{-3} \times N_{Ca}, \quad [1]$$

where f is filler content in percent, w is grammage in g/m², and N_{Ca} is number of calcium detections. This polynomial was then used on measurements of the XPM paper and the copy paper to estimate the local filler content from grammage and number of calcium detections.

High resolution grammage measurements – beta formation

For measurements of grammage variations, a beta-radiation based formation measurement method with fluorescent imaging plates was used (NSP 5 2009). An illustration of the method is shown in Fig 3. The sample is pressed between a ^{14}C β -radiating plate and an imaging plate. The ^{14}C β -radiating emits an even radiation over the sample surface. The local amount of beta radiation transmitted through the sample, and thus the number of electrons that hit the imaging plate, depends on the local grammage of the paper. After the exposure of the imaging plate it is placed in a special scanner for reading fluorescent imaging plates, FUJIFILM BAS-1800, which scans the plate with a spatial resolution of at least 0.1 mm. In this work a resolution of 0.2 mm was used. The intensity values of the scanned image are calculated into grammage values with the help of reference pads that are included in the same measured image.

It is necessary make the beta formation measurements in a room with controlled humidity, because the moisture content in the paper will be included in the measured grammage. The beta formation measurement equipment used here was placed in a room with a controlled humidity of 50% RH and a temperature of 23°C.

Image registration

For a point-wise combination of the measurements of calcium and grammage, an image registration method was used. The measurement area was 40 mm x 40 mm in size. As registration marks, five holes, each with a diameter of 3.0 mm, were placed at the edges of the measurement area. The center points of the landmark holes were found with image analysis by making a suitable threshold of the measured images and calculating the mean values of x- and y-coordinates for each of the holes. The image registration was made by using the calculated center points of the holes in each 2D measurement image. The distance between the holes was divided into 30 equal intervals by horizontal and vertical lines, creating a matrix of 30 x 30 elements, where each element corresponded to the mean value of approximately 1 mm² of the paper (Fig 4a). By using a grid in this manner, errors from possible dimensional differences and rotation differences between measurements were minimized. From the resulting matrix, the innermost 24x24 elements, corresponding to 24 mm x 24 mm were used for the analysis, in order to exclude image registration landmarks from the analysis (Fig 4b).

Measurement accuracy

The accuracy of the calculated values of local filler content is dependent on the accuracy of the measurement methods and the image registration method. The measurement accuracy of the different methods was estimated from the point-wise difference between repeated measurements on the same sample.

Fig 3. Illustration of the principle of the beta-radiation based formation measurement method. The sample is pressed between a ^{14}C radioactive plate and an imaging plate. The intensity of the transmitted beta radiation from the ^{14}C radioactive source on the imaging plate depends on the local grammage of the sample.

Fig 4 a). Matching grid, drawn on 2D beta formation image. b) Resulting 30 x 30 pixels grammage map. The innermost 24 x 24 pixels that were used for the analysis are marked with a white frame.

XRF measurements

The measurement time at each measurement point on the sample was set to 1 s. By making repeated measurements of the same spot, the accuracy of the X-ray fluorescence detector could be determined. Twenty repeated measurements on the same spot gave a standard deviation of 1,7% of the mean number of calcium detections at the spot. When applying the image registration method, the measurements, which were made with steps of 0,2 mm, were averaged over a surface of 1 mm². The standard

Fig 5. Point-wise comparison of two calcium measurements of XPM paper (a) and copy paper (b), averaged to 1 mm². For both samples, the mean number of detections were higher on the wire side.

deviation of the difference between two 1mm² surfaces was 0,6% of the mean number of calcium detections.

Due to the dampening of X-rays in the paper material, the z-distribution of the filler particles will affect the measurement result. By using the image registration method, measurements from both sides of the samples can be compared. In Fig 5, the pointwise number of calcium detections is compared between the top side and the wire side of the two paper samples. The correlation coefficient between the two sides was 0.75 for the XPM paper and 0.93 for the copy paper. For both paper samples, the mean number of detected filler particles was higher on the wire side than on the top side.

Beta formation

When comparing the point-wise values of two beta formation images of the same sample, the image registration method had to be used, because the sample cannot be replaced on exactly the same position in the sample holder between the exposures. Therefore, the difference between two beta formation images is also dependent on the accuracy of the image registration method used. The standard deviation of the point-wise difference between two measurements was 2% of the

Fig 6. Point-wise comparison of two grammage measurements averaged to 1 mm² of the same sample (a), and point-wise comparison of two grammage maps each calculated as a mean of two measurements (b).

variation interval when the measurement images were downsampled to a resolution of 1 mm.

The noise level in beta formation is usually estimated by using the variance measured in the reference pads. The exposure time affects the accuracy of the method. However, at a certain limit a longer exposure time does not significantly reduce the noise level of the measurement, because of the limited accuracy of the scanner. The noise level can then be reduced further by averaging over several measurements of the same sample, using an image registration method to combine the measurements. In Fig 6a, the grammage values of two measurements of the same sample are compared. Fig 6b shows the effect of adding more measurements by comparing two mean values. The variance of the point-wise difference between two measurements is lower when mean value of repeated measurements is used. The noise level can thus be partly compensated for by using repeated measurements of the same sample.

Image registration

When comparing distances between the image registration landmarks for the different measurement images, the maximal difference in distance between two

(a. Filler content vs. grammage in the XPM paper)

(b. Filler content vs. grammage in the copy paper)

Fig 7. Filler content vs. grammage at 1 mm scale a) for the XPM sample, and b) for a commercial copy paper. The light red points represent measurements of the top side of the paper, while the dark blue points represent measurements of the wire side of the paper.

Table 1. Mean grammage (*Mean w*), standard deviation of grammage (σw), mean number of calcium detections (*Mean N_{Ca}*), and standard deviation of number of calcium detections (σN_{Ca}) for the XPM paper and the copy paper.

Sample	Mean w	σw	Mean N _{Ca}	σN_{Ca}
XPM	75.6 g/m ²	4.2 g/m ²	2220	41
Copy	77.3 g/m ²	3.8 g/m ²	2190	93

images gives a measure of the accuracy of the image registration method. In order to have a reliable image registration method, the maximal difference between the maps cannot be too large. The largest hole-to-hole difference between two measured 2D-images was 0.2 mm, which is the same as the resolution of the XRF and beta formation measurements.

Results

High resolution measurements

The mean value and standard deviation of grammage and number of calcium detections are presented in *Table 1*. The standard deviation of grammage were similar for the two papers, but the standard deviation of number of calcium detections showed to be twice as large for the copy paper than for the XPM paper.

Lateral distribution of filler content

In *Fig 7*, the point-wise filler content of the XPM paper sample and the 80 g/m² commercial copy paper estimated at 1 mm² scale by the method presented in this work is plotted against grammage measured with beta formation. The light red points in the graphs represent measurements of the top side of the sample, while the dark blue points in the graphs represent measurements of the wire side. The correlation between grammage and filler content is approximately the same for the two sides, but the mean value of measured filler content is significantly higher for the wire side.

Table 2. Filler content assessed by ash residue, together with mean value and standard deviations of the local filler content assessed at 1 mm² resolution for both sides of the two paper samples.

	Ash residue	Mean Top side	σ Top side	Mean Back side	σ back side
XPM	22.4%	22.6%	0.7%	23.8%	0.7%
Copy	23.4%	21.6%	0.9%	22.9%	0.9%

The correlation between filler content and grammage is positive for the copy paper but negative for the XPM paper. The quotient between local filler weight and local grammage is lower in the flocs than between the flocs in the XPM paper.

The mean values of filler content estimated from measurements of the two sides of the two samples are shown in *Table 2*. The difference between the two sides showed to be approximately the same for the two samples.

Discussion

The correlation between filler content and grammage, shown for the two papers in *Figs 7a* and *7b*, was positive for the copy paper but negative for the XPM paper. A negative correlation between small-scale filler content and corresponding grammage at the same point implies that there are a higher proportion of filler particles between flocs than in the flocs. This could be due to the much lower speed of the XPM compared to a full-scale paper machine, which gives a higher retention.

By using image registration, a measurement of front side and wire side of the paper has been compared point-wise. However, the setup used here does not enable calculation of x-ray absorption properties of the material. With a modification of the sample holder that enables measurements of transmitted x-ray photons, this could be possible. From estimated absorption coefficients of the paper components, the local z-distribution of filler particles in the paper may be estimated more accurately.

The value assessed by ash residue (ISO 1762 2001) differs from the mean value of filler content for the copy paper (*Table 2*). This can be due to the calibration polynomial used for the calculation of local filler content from number calcium detections, which is derived from paper samples more similar to the XPM paper than the copy paper. The mean measured filler content in the copy paper should be closer to the value assessed by ash residue if an additional calibration polynomial was derived that better suited the copy paper. However, the main purpose with this measurement method is to assess the small-scale variations of filler content in the paper, and the offset of the mean value is then of minor importance.

The measurement method presented in this work can easily be applied to other uncoated paper grades, with other pulp compositions or production processes, or with different degrees of calendaring. The method can also be applied to coated paper grades. It will then be important to know the composition of the coating mixture.

Conclusion

This work presents a method for non-destructive high resolution measurements of filler content in paper. The local filler amount is dependent on the grammage distribution; therefore a matching method is proposed where the measurements of calcium content are combined with measured 2D grammage maps in order to assess local filler content in the paper.

The method has been applied on an uncoated laboratory paper and a standard commercial 80 g/m² copy paper. The correlation between point-wise grammage and filler content showed to be different for the two samples: the copy paper showed to have higher filler content in the flocs than between the flocs while the relation was the opposite for the laboratory paper.

With the help of image registration, the point-wise filler content measured from both sides of the samples can be compared. The results show a higher number of detected filler particles at the wire side of the two samples.

The method proposed in this work can be used together with other high resolution measurement maps and image registration methods in order to increase the understanding of the interrelations between lateral variations in paper properties.

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