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Non-destructive method to resolve the core and the coating on paperboard by spectroscopic x-ray imaging

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KEYWORDS: Spectroscopic X-ray imaging, Thickness measurement of layers in paperboard, Paperboard quality.

SUMMARY: Quality control is an important issue in the paperboard industry. A typical sheet of paperboard contains a core of cellulose fibers [$C_6H_{10}O_5$], coated on one or both sides with layers of calcium carbonate [$CaCO_3$] or Kaolin [$Al_2Si_2O_5(OH)_4$]. One of the major properties of a good quality paperboard is the consistency of the expected ratio between the thickness of the core and the coating layers. A measurement system to obtain this ratio could assist the paperboard industry to monitor the quality of their products in an automatic manner.

In this work, the thicknesses of the core and the coating layers on a paperboard with coating layer on only one side were measured using an X-ray imaging technique. However, the limited spectral and spatial resolution offered by the measurement system being used led to the measured thicknesses of the layers being lower than their actual thicknesses in the paperboard sample. Suggestions have been made in relation to overcoming these limitations and to enhance the performance of the method. A Monte Carlo N-particle code simulation has been used in order to verify the suggested method.

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At the present time, in the paper industry, mechanical and optical methods, such as Bendtsen and light triangulation methods are often used for surface profiling in order to observe the roughness and cockling of paper and paperboard surfaces. However, with these methods, it is not possible to measure the ratio between the thickness of the core and the coating as well as the homogeneity of the layers. Spectroscopic X-ray imaging could be a tool to obtain this ratio and, incorporating the imaging in an on-line measurement situation, the information could be sent back to the control station in the production line for quality adjustment. This article presents a method to separate the cellulose and the $CaCO_3$ layers (Norlin et al. 2009) on paperboards (*Fig 1a*) in terms of thickness using spectroscopic X-ray imaging.

The proposed method can be used to replace the Ammonium Chloride (NH_4Cl) burnout test (Dobson 1975) method, which has been widely used with regards to paper/paperboard quality inspection. In the burnout test method, a paperboard sample is treated with an NH_4Cl test solution (25 g/liter NH_4Cl in a 50/50 volume mixture of 2-Propanol/water), after which it is placed under a heat gun for two minutes to burn it out. The burnt sample (*Fig 1b*) is then examined for color change/development.

Fig 1. (a) SEM image of paperboard coating topology and (b) chemically developed paperboard.

This method involves carbonization of the paperboard contents, which is a destructive process. Random samples are examined, thus, it does not provide the required certainty in the quality of the whole production. Although it is the case that this type of inspection is conducted by skilled personnel, the possibility of human errors is not negligible. In addition, although this method provides some ideas about the uniformity in the paperboard layer structures, it does not provide the desired absolute thickness or uniformity of thickness. The quality of the paperboard depends on these two properties (Dahlström et al. 2008).

β -radiation is often used to monitor thickness of paper. Based on the fact that the absorption of β -radiation is proportional to the thickness of the paper, it is used to monitor the overall thickness of paper, not the individual thicknesses of its layers. Moreover, its short penetration depth limits its usage in measurements for paperboards.

In the X-ray spectroscopic method, the thicknesses of the cellulose and the coating layers can be measured using the difference in their X-ray absorption properties at different energies. This method is a non destructive method. In an on-line measurement system, it is possible for the entire production to be under observation.

Method

The aim of this method is to use a wide spectrum X-ray source and a well calibrated spectroscopic detector system with discrete energy binning so as to observe the X-ray absorption behavior through a paperboard sample at two energies in order to calculate the thicknesses of the

Fig 2. Aimed set-up.

Fig 3. Measurement set-up.

two layers on the paperboard. Fig 2 shows the intended set up. Due to the insufficient spectral resolution offered by the wide spectrum X-ray source, fluorescence sources were used in the actual measurements (Fig 3).

This method uses the linear attenuation coefficient of cellulose and calcium carbonate, which determines the fraction of an X-ray beam that will be absorbed in a material while passing through it. The linear attenuation coefficient for X-rays in a material depends on the photon energy and the material (Onishi et al. 2007). However, the variations in the coefficients for the majority of materials are similar, unless there is an absorption edge in the energy range of interest.

If t_1 and t_2 represent the thicknesses of the two layers in paperboard, and α_{1e1} and α_{2e1} represent the linear attenuation coefficients of the materials at a certain energy $e1$, then the X-ray response/intensity Φ , from the primary interactions can be written as:

$$\begin{aligned}\phi_{e1} &= \phi_{0e1} e^{-(\alpha_{1e1}t_1 + \alpha_{2e1}t_2)} \\ \Rightarrow \alpha_{1e1}t_1 + \alpha_{2e1}t_2 &= -\ln\left(\frac{\phi_{e1}}{\phi_{0e1}}\right)\end{aligned}\quad [2.1]$$

This equation shows that it is not possible to calculate t_1 and t_2 with only one energy. If another energy $e2$ is used, then

$$\begin{aligned}\phi_{e2} &= \phi_{0e2} e^{-(\alpha_{1e2}t_1 + \alpha_{2e2}t_2)} \\ \Rightarrow \alpha_{1e2}t_1 + \alpha_{2e2}t_2 &= -\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right)\end{aligned}\quad [2.2]$$

where, α_{1e2} and α_{2e2} represent the linear attenuation coefficients of the materials at energy $e2$.

Eq 2.2 can be written as:

$$t_2 = -\frac{\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right) + \alpha_{1e2}t_1}{\alpha_{2e2}}$$

By inserting this value of t_2 into Eq 2.1,

$$\begin{aligned}\ln\left(\frac{\phi_{e1}}{\phi_{0e1}}\right) &= \alpha_{1e1}t_1 + \frac{\alpha_{2e1}}{\alpha_{2e2}}\left(\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right) + \alpha_{1e2}t_1\right) \\ \Rightarrow \ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right) &= t_1\left(-\alpha_{1e1} + \frac{\alpha_{2e1}\alpha_{1e2}}{\alpha_{2e2}} + \frac{\alpha_{2e1}}{\alpha_{2e2}}\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right)\right) \\ \Rightarrow t_1 &= \frac{\ln\left(\frac{\phi_{e1}}{\phi_{0e1}}\right) - \frac{\alpha_{2e1}}{\alpha_{2e2}}\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right)}{-\alpha_{1e1}\left(1 - \frac{\alpha_{2e1}\alpha_{1e2}}{\alpha_{2e2}\alpha_{1e1}}\right)}\end{aligned}$$

Finally,

$$t_1 = \frac{\frac{\alpha_{2e1}}{\alpha_{2e2}\alpha_{1e1}}\ln\left(\frac{\phi_{e2}}{\phi_{0e2}}\right) - \frac{\ln\left(\frac{\phi_{e1}}{\phi_{0e1}}\right)}{\alpha_{1e1}}}{1 - \frac{\alpha_{2e1}\alpha_{1e2}}{\alpha_{2e2}\alpha_{1e1}}}\quad [2.3]$$

Now, t_2 can also be calculated by inserting the value of t_1 into Eq 2.2.

The denominator in Eq 2.3 will be close to zero if the ratio between the absorption coefficients for the two materials at the energy of interests is similar, as it is for most materials (or energies). In that case, the thickness calculation will experience a large error.

Eq 2.3 requires the calculation of the X-ray transmissions through the paperboard sample at two different energies. This is conducted by normalizing the spectral responses from the sample at these two energies with the respective corresponding source spectrum.

To separate calcium carbonate from other materials by means of spectral imaging, it is important to use energies which are just above and below the k-edge of Ca at 4 keV (Ohno et al. 2005). The achieved spectral resolutions using a wide spectrum X-ray source at the energy range of interest proved not to be sufficiently efficient for the measurements. Thus, CaO and Ti were used as 3.7 keV and 4.5 keV fluorescence energy sources. A microfocus X-ray tube with a Tungsten target was used as the primary X-ray source. Fig 3 shows the measurement set-up.

Fluorescence sources yield a large focal spot, resulting in a low spatial resolution. The measured spatial resolution of the TIMEPIX (Llopart et al. 2007) detector using the fluorescence energy was 0.45 lp/mm, which is much lower than that (12 lp/mm) of the same detector using a micro-focus X-ray source. The TIMEPIX detector was used in the photon counting mode, because its noise floor at 2.34 keV is required in order to detect the low energy photons in the measurements.

A simulation was performed using the Monte Carlo method in MCNP (Monte Carlo N-Particle Transport Code) in order to calculate the thickness of the paperboard layers using the above principles for both monochrome X-ray source and fluorescence source (Fig 4). The MCNP is a software platform to simulate the interaction of radiation with matters. The Monte Carlo method is a set of algorithms used in random sampling based computation.

Results

In the simulation, the same thicknesses as the sample paperboard (490 μm), the coating layer (60 μm) and the cellulose layer (430 μm) were used. The actual thicknesses were measured using the Millitast 1083 instrument, with 1 μm resolution. Eqs 2.3 and 2.2 were applied to the spectral responses of the samples consisting of only coating, coating with cellulose and only cellulose in order to calculate the thicknesses of the layers.

Fig 4. Calculated thicknesses from the simulation using MCNP.

Fig 5. (a) Optical image and (b) X-ray image of P. Paperboard, Q. Only CaCO_3 and R. Only cellulose layer.

Fig 6. Thickness profiles of the separated (a) Cellulose and (b) Coating layer.

The density of the paperboard was measured to be 0.72 g/cm^3 . The sample was prepared in such a way that all the areas of interest could be captured in a single image frame (256×256 pixel matrix, $1.4 \text{ cm} \times 1.4 \text{ cm}$). Fig 5a shows the measurement sample. On one side of the paperboard sample, the cellulose layer was removed (region Q) thus leaving only the coating layer. The coating layer was removed from the other side (region R) thus retaining only the cellulose layer.

The thickness of the cellulose layer for each pixel has been calculated from the spectral images (Fig 5b) (Turecek et al. 2011) at the two energies using Eq 2.3. The coating thickness has then been derived using Eq 2.2. The results are presented in Fig 6.

The linear attenuation coefficients of the two layers at the two fluorescence energies used in both the simulations and the measurements are provided in Table 1.

Discussions

This method considers only the primary interactions of the X-ray photons with the paperboard content materials. So, the influence from secondary interactions must be considered when interpreting the results.

The calculated thicknesses, both in the simulation and the measurement using fluorescence energy sources, are lower than the thicknesses measured from the simulation using monochrome sources. This is because of the high energy Compton contributions from the fluorescence sources.

Table 1. Linear attenuation coefficients of the layers on paperboard.

Materials	3.7 keV, (cm^{-1})	4.5 keV, (cm^{-1})
Cellulose	56.7995	31.5272
CaCO_3	327.91	942.5380

Fig 7. MCNP simulated spectra of the Ti and the CaO fluorescence sources.

Fig 8. Relative error in measurements of the test samples.

Fig 7 shows the Compton peaks for both of the fluorescence sources at the 15-50 keV energy range.

The proposed method assumes that the coating layer and the cellulose layer on paperboard contain only CaCO_3 and cellulose, respectively. In reality, a paper-board contains other materials beside these, for example, filler and pigment. It may also contain air gaps and even ink pigment if it is produced with recycled fiber. In a practical application, the measurement system has to be calibrated depending on the amount of these contents that present on paperboards.

Fig 8 shows the estimated error (simulated) in measurements for different energy bin sizes. It has been estimated that the measurement error exceeds 90% if the energy bin size is larger than 0.8 keV. The presented results in this article are affected by the measurement errors, as it was not possible to achieve a reasonably small bin size based on the detector system which was used.

A better spatial resolution is required in order to resolve the structures that define the paperboard quality. It might be possible to achieve the necessary spatial and spectral resolution by using X-ray sources with a small focal spot and an imaging system with a better spectral resolution i.e. MEDIPIX3 (Ballabriga et al. 2007). The architecture of MEDIPIX3 is capable of minimizing spectral distortion by using a charge summing method and can be used in the set up in Fig 2.

Conclusions

This article demonstrates that it is possible to separate the cellulose and the CaCO_3 layer on a paperboard using spectroscopic X-ray imaging. The influencing factors which affect the accuracy of the measured thicknesses are discussed and suggestions have been made to improve the performance of the proposed method.

Further investigation is going on how to apply this method to measure thicknesses of the layers on paperboard with coating on both sides, by combining it with the information obtained with the grating interferometer-based Phase Contrast X-ray Imaging technique (Weitkamp et al. 2005).

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