Experimental improvements for micro-tomography of paper and board

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Abstract
Since early in this century, micro-tomography using X-ray synchrotron source has aroused scientific interest to characterized structure of paper. This paper shows improvements that have been made since first micro-tomographic experiment. This concerns not only the X-ray beam preparation but also the paper sample preparation. To end this paper direction in the new developments are given.
Keywords: Micro-tomography, fibrous media, synchrotron, in-situ experiment.

1 Introduction

Fibrous media are ubiquitous in our lives and have a tremendous importance. The variety of their applications is tremendous as for example in medicine, in electronic or in automotive. The need for new materials development or improvement of existing ones still grows. When studying fibrous media whatever the purpose is, the fibrous structure has to be described. Micro-tomography using X-ray synchrotron radiation arouses scientific interest to achieve this goal. To illustrate this point the peculiar case of papermaking may be hold. When characterising paper with classical method [1], the resulting information are often macroscopic ones or 2D microscopic ones. Nevertheless, in the latter case, a 3D analysis may be done using statistical model [2] [3]. A thorough 3D description of paper may help to improve the end-use properties or the papermaking process itself. This paper is devoted to describe recent progresses that have been done in the micro-tomography of paper. Obviously

these improvements may be applied to other fibrous media. In a first time, the characterisation technique is described, in a second part scientific background relevant to paper characterisation using this technique is shown. The third part is dedicated to improvements of the experiment itself. Conclusions and perspective will end this paper.

2 Generalities

2.1 Theory

Synchrotron radiation is the electromagnetic radiation produced by ultra-relativistic electrons (energies of several GeV) stored in a ring when they are deviated by a magnetic field. The X-ray beam is emitted in a narrow cone tangent to the particle curved trajectory in the storage ring. Synchrotron radiation source may provide beams with very high intensity, i.e. high photons flux, and a continuous spectrum, spanning the whole range from infrared to hard X-rays. The outstanding features of third generation synchrotron radiation facilities, and more particularly the ESRF, European Synchrotron Radiation Facility, in connection with μtomography are:

- the very high intensity of the X-ray beam,
- the availability of photons over a very large energy range (energy E= 1 to 300 keV, wavelength λ=0.004 to 1.2 nm),
- the possibility of tailoring the beam to the requirements of a given experiment by choosing the most appropriate insertion device (wiggler or undulator) with a broad range of variations.

These features make it possible to perform micro-tomographic experiments that are improved. The broad energy range available allows tuning the photon energy to a given investigation, generally in terms of the deposited dose or the signal to noise ratio. Imaging on both sides of an absorption edge of a chemical element present in the sample makes it possible to map the presence of this element in the volume. The very high beam intensity reaching the sample allows to improve the spatial resolution down to the micron level, and also to perform relatively 'fast' tomography on dynamic systems. The coherence properties of the beam make it possible to obtain phase images by simply adjusting the sample-detector distance *D* ("propagation" technique). Two other aspects are not directly related to the source, but are nevertheless crucial for this type of experiment:

- The availability of a suitable high resolution CCD-based detector. It consists [4] of a scintillator screen converting the X-rays into visible light, magnifying optics and a Peltier cooled CCD. The latter needs to have, simultaneously, a large dynamic range (13 bits in our case), low noise and a short read-out time. We use the Fast Readout Low Noise (FReLoN) camera, developed at the ESRF [5].The best spatial resolution achieved being 0.5 μm.
- Appropriate image processing software and computing power are required to complete the analysis; they often remain a bottleneck.

All the presented results in this paper were performed with absorption contrast technique. This method relies on the Lambert-Beer law (Eq. 1).

$$\int_{path} \mu(x,y,z)dz = -\ln\left(\frac{I(x,y)}{I_o(x,y)}\right) \quad \textbf{1}$$

The provided beam is supposed to be monochromatic (fig. 1) and parallel, the relationship between the intensity I(x,y) transmitted after a path along *z* in the sample, the incident intensity $I_o(x,y)$ and the projection of the linear attenuation coefficient μ, is straightforward and one-to-one. Assuming that this projection is known for a large number of angular orientations of the sample, tomographic reconstruction makes it possible to quantitatively map the distribution of $\mu(x,y,z)$. The linear attenuation coefficient μ depends, for given X-ray energy, only on the composition and the density of the material.

When the beam is polychromatic the linear relationship between attenuation and materiel thickness is no longer hold. Lower energies are more absorbed than higher ones. The energy distribution is thus unevenly modified when passing through the sample and will contain a higher proportion of high energies leading to a hardening of the beam. The use of a monochromatic beam allows avoiding beam hardening artefact.

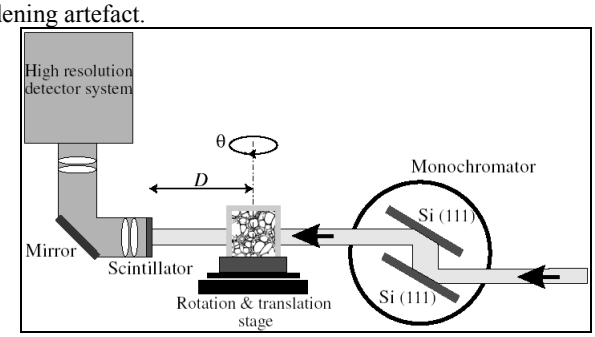

Figure 1: Principle of parallel and monochromatic beam Micro-tomography at beamline ID19 of the ESRF. The synchrotron beam is monochromatised either by a double silicon crystal monochromator or by an Al_2O_3/Ru multilayer. Radiographs for a large number of angular settings θ over a 180° range are recorded with a CCD based high resolution detector system. The sample detector distance D can be varied from ≈ 0 to 1 m.

2.2 ID19 presentation:

The ID19 beamline main features derive from the requirements of having a spectrally and spatially homogeneous, highly coherent, beam at the sample position, with beam maximum dimensions of 45 x 15 mm^2, a high photon flux and tuneable photon energy in the range of 6 to 60 keV. These requirements lead to the choices of a long beamline (145m). Three different high magnetic field devices may be used as source: a wiggler and two undulators. The long source-to-sample distance, added to the small dimensions of the source (≈ 0.1 mm) are necessary conditions to obtain a highly coherent beam (fig. 2). The

monochromatic beam is delivered, as a function of the required experimental conditions (depending essentially on the wished ΔE/E or the flux that required the experiment), by one among two possible monochromators both located close to the beginning of the experimental hutch:

- a 'single' crystal or multilayer ($\Delta E/E = 10^{-2}$),
- a double crystal, fixed exit, monochromator diffracting in the vertical plane is located in the monochromator hutch. The crystals are 30 cm long (111) Si, in symmetrical Bragg position; the first crystal is water cooled ($\Delta E/E = 10^{-4}$).

In many cases however, conventional absorption imaging fails. The spatial coherence of modern synchrotron beams makes possible a trivial form of phase imaging based on propagation [6].

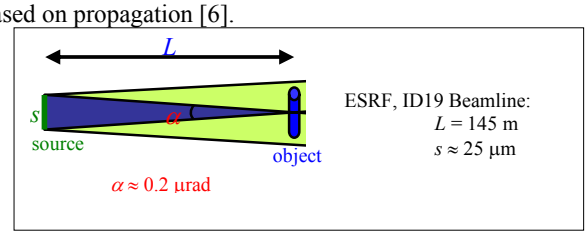

Figure 2: The small size of the electron beam cross section, the small horizontal and vertical (the green zone) divergency of the source (< 100 µrad) and large distance between the source and the sample (L). This leads to a very small angular extension (α) of the source as see.

2.3 Scientific background

First experiment on paper using Micro-tomography took place at the ESRF in 2000 on the ID22 beamline. This was collaboration with the Norwegian University of Science and technology (NTU), the Norwegian pulp and paper institute of Trondheim (PFT) to demonstrate the feasibility of the imaging paper sample. Experimental details may be found in the PhD thesis of Weitkamp T. [7]. The preliminary results [8] show the feasibility of imaging but raise the issue of the data post processing due to the amount of artefacts (artificial features added to the real structure). Later experiments on paper were all carried out on ID19 by the paper process-engineering laboratory of Grenoble. Simultaneously to these, experiments were carried out in the synchrotron by the department of Wood & Paper science part of the University of the Minnesota and presented in [9]. Recently a long term project was accepted on ID19 dedicated to paper fundamental research. This one year long term project gathers NTU, PFI, LGP2, Finish team (University of Jyvaskyla) and the University of Minnesota (Department of Wood and Paper Science).

The early experimental setup on ID22 was using a horizontal rotation axis. The X-ray beam energy was 20 keV. The pixel size was about 0.34 µm. When using paper strip of about 1 mm large mounted horizontally, the strain due to the earth gravity could not be avoided. So most of the samples were strained during their rotation leading to unusable results.

The ID19 beamline has already been presented. On this beamline the rotation axis is vertical. The paper sample is placed orthogonally to the rotation axis. This can help the paper sample to move due to the gravity. Furthermore the section that the X-ray beam crosses shows slightly the same attenuation in all the direction leading to fewer artefacts.

3 Improving micro-tomography

The end-use of the reconstructed volumes motivates directly and strongly the points to improve. In all studies using 3D geometrical description of paper structure, the data has to be segmented. The segmentation step is the pass way of attenuation map to object map. In order to improve this step, the approach consists in sorting out the factors that are adventitious in the segmentation step. Then knowing these factors, solutions to decrease them are found. Obviously, the segmentation step is very sensitive to contrast. The further the attenuation values of the different phases are, the easier the segmentation step is. The noise (the fast variation of the value attenuation in respect of the space) is another adventitious factor that brings issues when segmenting. When segmenting, the presence of artefacts raises up complex problems. Artefacts are features that appear in the reconstructed volumes that are not present in the real structure. This may be lines, segment, circles or features with much more complicated shapes. Our approach is to experimentally sort out these issues by modifying the experimental condition of the micro-tomography. Only few parameters can be tuned: beam energy, beam flux, beam size, exposure time, number of projection and sample preparation.

3.1 Contrast

Contrast may be defined as the relative difference of attenuation values between neighbour phases. The relation between the energy and the chemical nature of the component drives the attenuation coefficient value. When energy is outside the absorption edges of chemical components of studied matter, the linear absorption coefficient may be expressed as follow:

$\mu = C\frac{Z^4}{E^3}$ where C is a fixed coefficient, Z is the atomic mass and E is the beam energy.

Hence the only way to improve contrast is to change the chemical nature of components (with isotope, doping agent). In this work this solution was too complicated. Notwithstanding another way to improve artificially the contrast is to use the edge enhancement technique. Edge enhancement consists in using the sample to detector distance to outline the contours of the sample structure. Increasing this distance, all interfaces are enhanced by black-white contrast of increasing amplitude [10].

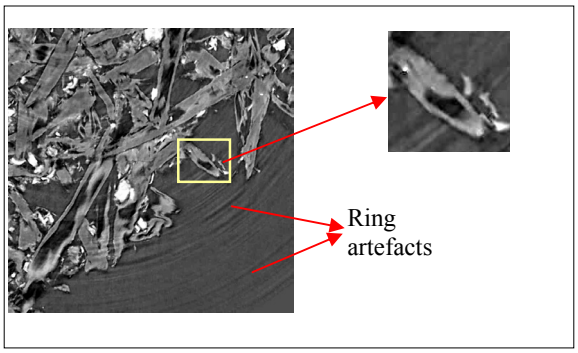

Figure 3: Reconstructed slice of paper. Fibres and fillers (white stains) are designable. In the details, the edge enhancement is shown (dark zone around the fibre).

3.2 Noise

There are different sources of noise: experimental ones and numerical ones. The experimental noise obviously comes from the detector itself but also from the beamline features. The numerical noise is due to the back-projection algorithm used for the reconstruction. Thus both origins of noise may be studied separately. In this paragraph only the experimental part is sorted out. The level of noise (standard deviation of the attenuation coefficient of the fibres) in the reconstructed volume is measured in respect of energy, exposure time and number of projection. Other experimental and numerical parameters remain constant. Nevertheless the samples of paper change for each test because this study was done on users allocated beam time. The comparison of the results with three different energies (table hereafter) tends to show that the standard deviation of the attenuation value in the fibres dramatically decrease when increasing energy of the X-ray beam. Increasing number of projection decreases noise level. Increasing exposure time leads to a better use of the dynamic range of the camera and consequently leads to less noise. But increasing exposure time has limit: the saturation of the ccd. Generally exposure time is set in the way that all pixels in the radiograph are just under the saturation level. Both latter solutions may provide collection of data that are too long in respect either of the allocated time for the users or that are not compatible with the media itself (see Open interface paragraph). Another way to increase the signal to noise ratio when using energy lower than 25 keV is to decrease the path of beam in the air. Hence fewer photons are absorbed by the air. Consequently more are available for the tomographic experiment. An easy way to decrease this path is to put as close as possible the micro-tomograph to the exit pipe bringing the X-ray beam. A more complicated way is to use pipe filled with gaseous helium to carry the beam to the micro-tomograph. Another source of noise is raised up when doing local tomography. This means that the object is not always in the field of view of the camera during the whole collection of radiographs.

Table 1: Parameters for the micro-tomographic experiments. Distance to sample and paper size are similar for each experiments.

Pixel size	Number of projection	Energy	Exposure time (s)	Ref Max (adu)	Fibre	
					μ	σ
0.7	1200	12	0.8	13375	7.68	6.27
0.7	1200	18	1	8913	3.50	2.00
0.7	1500	18	1.2	11147	2.93	1.44
0.7	1500	20.5	0.6	11277	2.01	1.68

3.3 Artefacts

Only ring artefacts are detailed in this paper. Ring artefacts are circular shaped artefacts that appear after reconstruction in cross section perpendicular to the tomographic rotation axis. They may be thick or thin, complete or partial (arc). Their origin is various. Some thin ones may be explain by abnormal pixels in the ccd camera system. These abnormal pixels have a non linear response instead of a linear one. Nevertheless the transmitted information is correlated with their neighbourhood. These abnormal pixels are always the same. A map of their places is update using the feedback of users. The resulting value of the incriminated pixel in each radiograph is replaced by the median value of its close neighbourhood. The majority of thick ring artefacts are correlated with the presence of defects in the homogeneity of the beam that go through the sample. The origin of these inhomogeneities is multiple. This may be some dusts settled either on windows (beryllium, carbon), on the monochromator or this may be material defect due to making. When the X-ray beam meets a dust, coherence is locally lost. This incoherent zone is propagating downstream the beam and growing. And then locally in the radiograph the assumption of linearity between the transmission and the attenuation coefficient value is no more verified. Another origin of ring artefact is that the beam is moving vertically in respect of the sample. Generally this is due to drifts of the monochromator due to thermal strains created by the white X-ray beam itself. When matter absorbs energy most part of it is dissipated by heat. Heat may be so important that it induces strains in the crossed structure. The multilayer used in all presented experiments is positioned in a helium atmosphere and is placed on a mixed of gallium and indium cooled by chilled water. Slits that tailored the beam play an important role. Shaping the beam at the exact size of the sample allow to maintain the coherence of the beam and to decrease the number of ring artefacts after the reconstruction. When an object such as a slit cuts the X-ray beam, some photons are diffused i.e. a small area after the slit the beam is no longer spatially coherent. Therefore three sets of slits are used. One close to the X-ray source, the second one before the monochromators, the third one close to the sample.

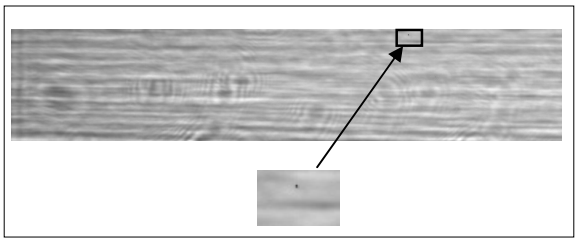

Figure 4: Image of the X-ray beam. The fringes are created by the defects on the windows crossed by the beam and on the monochromator. An example of abnormal pixel is shown in the detail.

3.4 Open interface, superposed features

After reconstruction, a first glance at few reconstructed cross-sections could show open interface or superposed features (fig 7). This happens generally when the sample moves. Papers and boards are very sensitive to air moisture. A small variation in air humidity may induce modifications in the fibrous structure. These modifications must not happen during the experiment. Hence the more stable the air environment of the studied sample is and the better the micro-tomography quality is. The key to achieve this is in experiment preparation and in sample preparation. Increasing the energy of the beam will decrease beam interaction (less absorption) with the fibre leading to less heat dissipation, and therefore less local humidity variation. Furthermore sometimes fillers added to paper are highly absorbent in particular in the case of TiO_2. The aggregates of such chemical adjuvant may be so absorbent for low energy that they introduce line artefacts (dark zone) between aggregates in the same reconstructed slices. When using a higher energy the aggregates are less absorbent and artefacts decrease or even may disappear.

3.4.1 Sample preparation

Basic and classical cautions are taken when preparing the samples. For example, plastic gloves are worn in order to prevent the sample from being soiled. The first step in preparation is to cut the sample in the way that its shape fits the camera field of view and is as close as possible as a cylinder. Notwithstanding, meet these both conditions was not possible in our case. Two methods to cut the sample were tested, one using punches and the other using scissors. The main asset of punches is that all samples are cylinders with identical diameter and fit perfectly the field of view of the camera. Nevertheless this method has a tremendous drawback. The paper structure is deeply damaged when cutting the paper either by punching or using a press drill. This happens especially with light sorts (e.g. bible paper). The other technique consists in cutting samples manually with scissors. This presents the major asset to prevent the paper structure from major deformation, notwithstanding the sample shape is no more cylindrical but more or less parallelepipedical. The drawback of using parallelepiped shape is that the studied volume is not optimum and specific artefact may appear at the

corner of the reconstructed paper volume. The second step in paper sample preparation is to fix the sample on the top of a capillary. The issue that arises in this step is to find a fixing agent that maintains on the top of the capillary the sample all the experiment long, that not penetrates paper structure and finally that will not carry out difficulties for structural analysis. Different methods to fix the sample are tested using glue for paper, specific polymer for X-ray imaging, double-sided adhesive, polymer plus post-it. In all cases, in the work of the structural analysis, the automatic separation of fibre from the fixing agent is an issue. Paper sample are prepared at least 24 hours before the experiment. The prepared samples are placed in the experimental hutch where the Micro-tomography experiments are carried out. By doing this, a stable moisture content of paper is expected to be reached.

A sample changer is used to load and unload paper sample from the micro-tomograph. Sample alignment is not automated yet and remains done by the users. The automate use allows to reduce the number of experimental hutch openings. Hence sample environment is more stable. Less hygrometry and temperature fluctuations are hoped. Classically temperature in the experimental hutch is about 23 degrees Celsius and 60% of relative humidity. Room temperature is controlled by air cooling system humidity is not. Notwithstanding investigations of technological solution for this issue are going to be found. When the X-ray beam is monochromatised, a part of the energy is dissipated by heat. Nevertheless after 5 to 10 minutes, mechanical and thermal stabilities of monochromator are expected. One of the automate assets is that there is no need to stop the beam during the sample changing. Hence the monochromator is always lighted by X-rays. Finally, incident monochromatic beam on the sample is less subject to variation. Therefore tomographic experiment is performed in better conditions.

4 Conclusion and perspectives

Micro-tomography is a powerful tool to investigate the structure of material. Nevertheless, sorting out results from micro-tomographic experiments may be dramatically difficult because of the low quality of data. Some advice has been given to help users to improve quality of such experiments. These recommendations may be applied not only for fibrous media but also for a broad range of micro-tomographic experiments. Concerning the specific study of paper, the next step is to control more accurately the humidity and the strain of the tested sample. Compression cell is presently being designed in this way. Another effort is done on the beamline automation itself in order to reduce as much as possible dead time for beamline users. Alignment and tuning of the X-ray beam are main goal of this project that is a global to the whole ESRF

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