Proceedings of the International Congress on Ultrasonics, Vienna, April 9-13, 2007, Paper ID 1625, Session R20: Resonators and waveguides
doi:10.3728/ICUltrasonics.2007.Vienna.1625_salmi

Microelasticity in wood using X-ray diffraction and ultrasound

Ari Salmi[1], Antti Meriläinen[1], Mika Torkkeli[2], Marko Peura[2], Jonne Haapalainen[1], Edward Hæggström[1,3] and Ritva Serimaa[2]

[1]Electronics Research Unit, Department of Physical Sciences, P.O.B. 64, FIN-00014 University of Helsinki, ari.salmi@helsinki.fi

[2]Division of X-ray Physics, Department of Physical Sciences, P.O.B. 64, FIN-00014 University of Helsinki

[3]Helsinki Institute of Physics, P.O.B. 64, FIN-00014 University of Helsinki

Abstract: We want to understand where wood strength comes from in order to artificially engineer the mechanical properties of wood. This study aims to measure the effect of macroscopic stress on micro scale modulus of elasticity of Scots pine (*Pinus Sylvestris L.)* wood cell walls. Our approach allows estimation of local elasticity as a function of stress as well as the stress-strain curve without inducing creep. We propose a measurement setup combining x-rays, and ultrasonics. A piezoceramic transducer launches continuously longitudinal waves in the longitudinal wood direction at a suitable frequency generating a λ/2 resonance into a 20x10x1.5 mm^3 cubic sample. The resonance is maintained by changing the sonic frequency with an Atmega 5815 microprocessor driven feedback loop into which an error signal obtained with a piezoceramic transducer is inserted. The diffraction pattern of the samples was measured after the resonance was achieved using CuK_α radiation (1.54 Å) with a beam size of 0.3x1.2mm^2. In this paper, results from both wood and clay-polymer (PP) composites are presented.

Keywords: Ultrasound, X-ray diffraction, microscale mechanical properties

A. Introduction

Wood has been an important part of Finnish economy since the early 20th century. Even though we have a strong understanding of the macroscopic properties such as stiffness and elasticity[1], and the applications of wood[2], the mechanisms in heterogeneous wood types are yet fairly unknown[3]. In general, macroscopic properties arise from the microscopic structure of materials, and thus it is vital to gain access to the micro-scale phenomena. This is especially important if one wants to understand *where does strength come from* or how to *artificially engineer the mechanical properties of wood*. There have been several attempts to cross the macro-micro –boundary, using optical methods[4], nano-indentation[5, 6] and x-rays[7]. Perré showed that macroscopic properties are sums of averages in unit cells of microscopic properties and applied this to X-ray images[3]. In addition, atomic force modulation microscopy has been applied to determine cell level mechanical properties[8].

Much of the durability problems with macro scale structures arise from stress distributions and their effect on structural degradation[9]. Since the elasticity of wood has a strong connection with its microfibril angle (MFA)[10], and thus the diffraction pattern, it is logical to do an *in situ* study of effect of stress on micro scale. There is some existing research in this field using tensile testing and x-rays[11], but however, creep is a problem during static loading tests[12] which could otherwise easily be used to apply stress fields into wood structures.

Clay-polymer composites are new materials that have risen attention recently. These materials may be used for applications that require e.g. improved thermal stability or optical clarity[13]. For further information on the nanocomposites, please read[14].

Basic acoustic theory states that standing waves may be created in geometrically suitable structures depending on the wavelength and sound velocity of the material. These waves are stress distributions resulting from interference of propagating waves, in which the displacement of the single cells is zero at the nodes and maximum half-way between two nodes. The pressure maxima are found at the nodes, while half-way between the nodes the pressure is zero.

We propose a novel measurement setup combining medium and short wavelength wave motion. To our understanding, there has been only one study before combining ultrasound resonance and x-ray diffraction[15] in a way resembling the proposed one, thus making the effort worthwhile and still serving as an indication that the proposed project is feasible.

B. Methods

Both Scots pine (*Pinus Sylvestris L.*) and clay-polymer composite (PP) samples were used. The wood samples were cut longitudinally creating 3 cm x 7 cm x 1.5 mm sheets, out of which the actual samples were cut (Fig. 1). The final samples were 1.5 mm thick, and their length was cut to match the λ/2 resonant frequency at 102 kHz.

An ultrasonic piezoceramic 100 kHz (96-106 kHz -6dB) transducer was used to launch continuous longitudinal waves (40 V_{PP}, amplified with a semi-conductor based power amplifier) in longitudinal wood direction at a suitable frequency generating a λ/2 resonance into a 20x10x1.5 mm^3 cubic sample. The sound velocity in a wood sample fluctuates as a function of relative humidity and temperature[16, 17], which is compensated by changing the frequency with a Atmel Atmega 8515 (8-bit, 16 MHz) microprocessor driven feedback loop. A second transducer was used as a receiver. The DC level of the signal was extracted using a positive peak detector, and this signal was then digitized

doi:10.3728/ICUltrasonics.2007.Vienna.1625_salmi

with a AD7623 A/D converter and directed into a microprocessor.

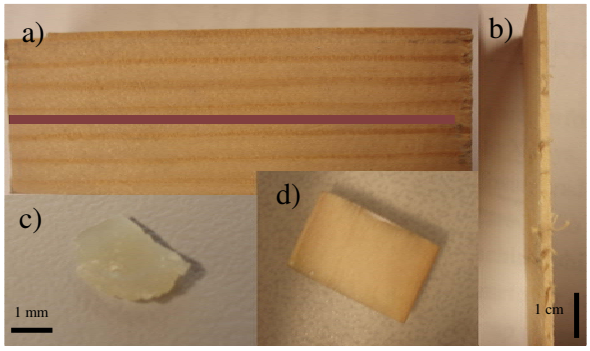

Fig. 1. Samples used in the study. (a) rectangular wood block, out of which sheets (b) were cut (marked region). Finalized samples: wood (c) and clay-polymer composite (d).

When resonance is acquired, according to the theory, no movement occurs in the ends of the sample. The microprocessor thus lowers the frequency generated by a Direct Digital Synthesis (AD9830) based numerical controlled oscillator, and then re-detects the DC level of the signal. It then scans over a 6 kHz band centred on the last resonant frequency and sets the frequency to the maximum amplitude normalized to the transducer bandwidth. The signal-to-noise ratio (SNR) affects the small fluctuation around the resonance frequency. A long (1 h) stability test was committed to ensure reliability of the setup. In addition, environmental parameters (R_H) were changed to test the resonance upholding capability.

Simulations confirming the creation of a resonant pattern were done with ABAQUS/Standard 6.6-1 finite element modelling software. The applied material model for wood was isotropic and elastic (E=2.1GPa, ν=0.3, ρ=535kg/m³), since the wave propagation was modelled only in one direction. A direct-solution steady-state dynamic analysis was made and the steady-state dynamic linearized response of a system was calculated.

A custom-made sample holder was manufactured to hold the sample in front of the x-ray beam without applying static tension to the sample. (Fig. 2.).

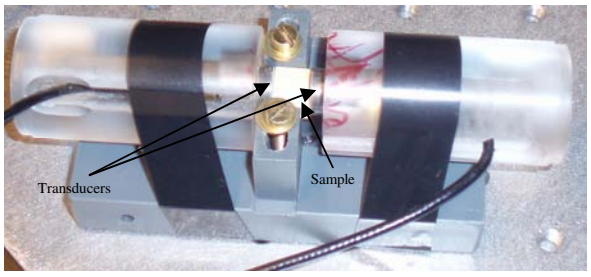

Fig. 2. Sample holder holding a wood sample.

The X-ray (Wide-angle X-ray Scattering, WAXS) measurements were carried out at room temperature using a rotating anode setup powered with Rigaku UltraX18S generator (18 kW, 50 kV, 60 mA beam current). The X-rays are focused and monochromatized with bend Cu/glass mirror and bend asymmetric Ge(111) crystal providing spectrally pure Cu Kα (λ=1.5405 Å) beam (0.3 mm × 1.2 mm on the detector). The data were collected using a Mar345 image plate detector. (Fig. 3.) The setup can detect changes in the crystalline structure of the samples.

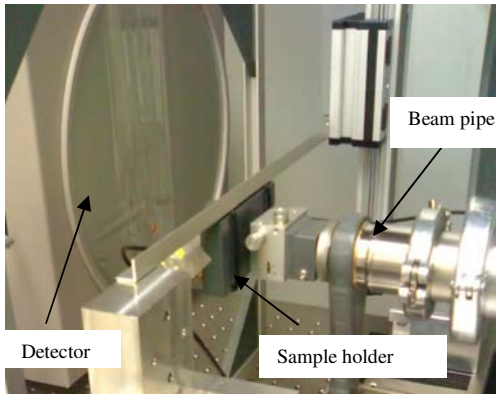

Fig.3. The WAXS setup with the sample in place.

C. Results

The FEM simulation indicated that we are able to obtain a λ/2 standing wave resonance (Fig. 4.)

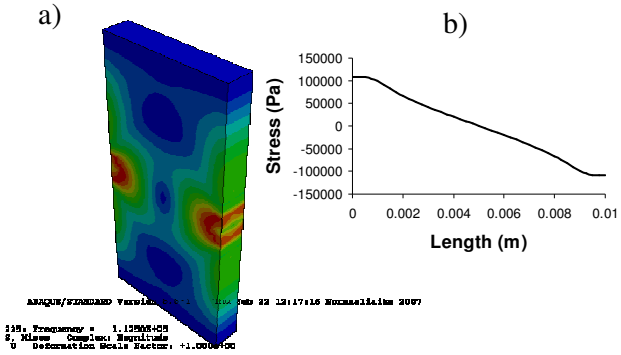

Fig. 4. Simulated stress distribution in a wooden sample. Absolute values of stress are shown in the 3D-image (a). The stress distribution along the middle axis is shown in (b).

The microprocessor controlled feedback loop showed stable behaviour, and was able to follow the changes to the resonant frequency caused by changing environmental variables (Fig. 5.)

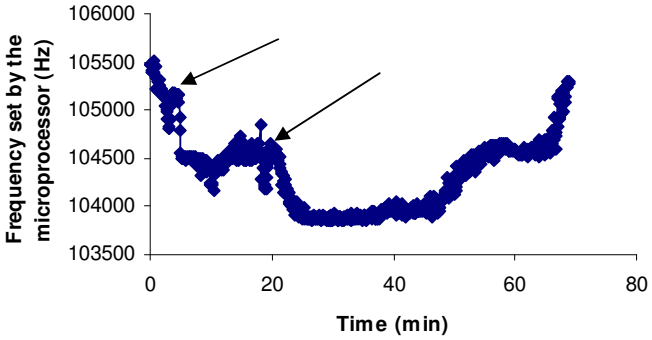

Fig. 5. Frequency set by the microcontroller as a function of time in a wooden sample. The arrows mark spots where humidity (a drop of water) is applied to the sample.

In the wood sample measurements, a clean diffraction pattern was obtained. The edge of the transducer is visible

doi:10.3728/ICUltrasonics.2007.Vienna.1625_salmi

in the figure. (Fig. 6.).

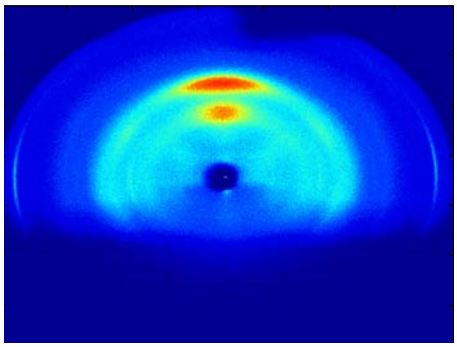

Fig. 6. Diffraction pattern of the wood sample. 004, 200, 110 and $1\bar{1}0$ peaks are present, as expected[18]. The figure is asymmetric because of the transducer edge visible in the lower region.

The intensity profile of the 004 reflection was obtained by integrating the 2D pattern over a polar angle. No clear change in the diffraction peak FWHM was detected (Fig. 7.).

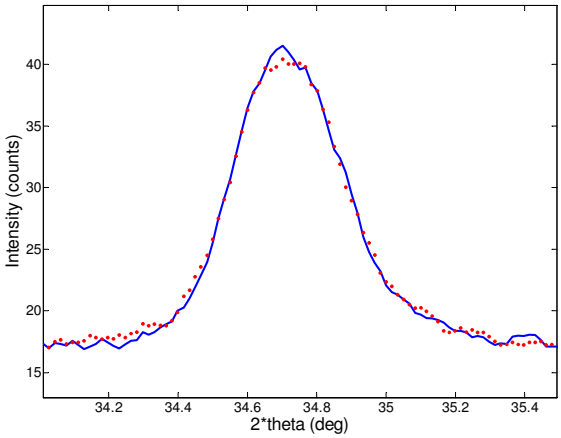

Fig. 7 The intensity profiles of the 004 reflection for the reference sample (blue dashed line) and a sample with applied ultrasonic field (red dots). No clear change in peak width is detected.

In the clay-polymer composite samples the diffraction pattern was clear, and the X-ray beam hit near the edge of the transducer in the region of pressure maxima (Fig. 8.).

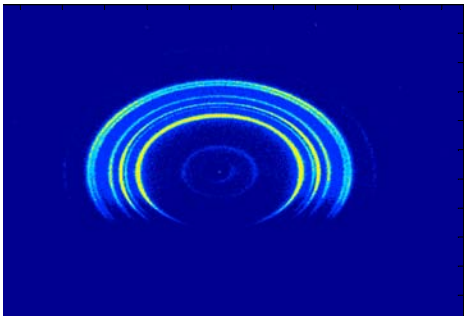

Fig. 8. Diffraction pattern obtained from the clay-polymer composite.

We see a clear change in the line integral (Fig. 9.) when the diffraction peak of the clay in the composite is studied (Fig. 10.)

D. Conclusions

The microprocessor controlled feedback performed well, being capable of detecting small changes in the environmental conditions and adjusting the resonant frequency according to them. This allows reliable generation of a resonant pattern in the sample during long measurements (e.g. wood samples, which take appr. 30 minutes to complete).

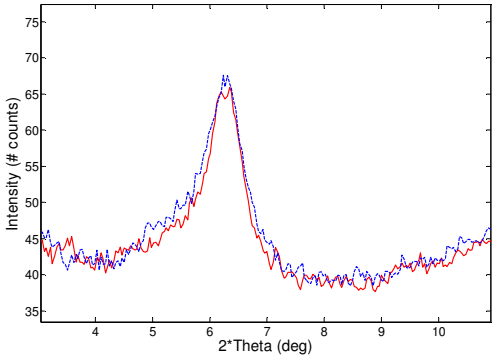

Fig. 10. 001 diffraction peak of the organoclay11[14] in the clay-polymer composite. There is a clear change in FWHM when ultrasound is applied (red solid line) compared to the reference (blue dashed line).

The diffraction peaks obtained for the wood sample matched those published earlier, and the X-ray beam could be aimed to the edge of the transducer. This allows

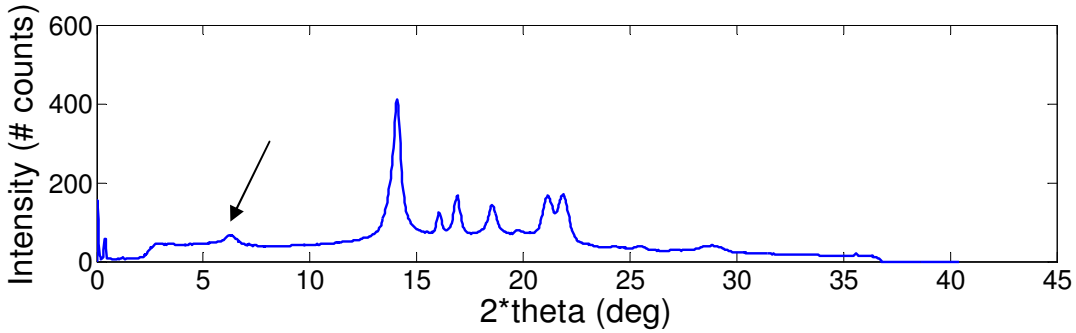

Fig. 9. The intensity curve of the clay-polymer composite obtained by integration over polar angle. 001 reflection of the organoclay11[14] is marked with an arrow.

doi:10.3728/ICUltrasonics.2007.Vienna.1625_salmi

application of the maximum pressure with no displacement of the cellulose fibres according to basic acoustic theory and our FEM simulations.

However, no consistent change in the FWHM of the reflection 004 in the wood sample was detected. This is most probably due to the low power applied to the sample. The longitudinal modulus of elasticity of cellulose fibres is appr. 160 GPa [19], and thus a 1‰ change (close to the low limit of the X-ray) in the strain would require 16 MPa of stress.

In the clay-polymer samples, a clear change in the FWHM of the line integral of the diffraction pattern was detected. This implies that the resonant wave pattern was able to 'strengthen' the structure, since the coherent length of the crystalline structure was increased. There are similar results obtained with non-resonant ultrasound using 20 kHz power ultrasound.

In the future, a 300 watt power amplifier will be used in conjunction with the microprocessor controlled feed-back loop to create additional pressure in the sample. In addition, since the elastic modulus of cellulose fibres is decreased by ten-fold in radial direction, a study of effects of radial ultrasonic resonance will be performed.

The clay-polymer composite demonstrated another application of our technique: ultrasound induced treatment of materials. The resonant pattern allows creation of localized effects on e.g. self-forming layers or composite materials. This will be studied in the future.

E. References

[1] Huang, C.-L., H. Lindström, R. Nakada, and J. Ralston, "Cell wall structure and wood properties determined by acoustics - a selective review", Holz als Roh- und Werkstoff, 2003. **61**(5): p. 321-335.

[2] Kopac, J. and S. Sali, "Wood: an important material in manufacturing technology", Journal of Materials Processing Technology, 2002. **133**(1-2): p. 134-142.

[3] Perré, P. and E. Badel, "Predicting of oak wood properties using X-ray inspection: representation, homogenisation and localisaion. Part II: Computation of macroscopic properties and microscopic stress fields", Ann. For. Sci., 2003. **60**: p. 247-257.

[4] Sippola, M. and K. Frühmann, "*In situ* Longitudinal Tensile Tests of Pine Wood in an Environmental Scanning Electron Microscope", Holtzforschung, 2002. **56**: p. 669-675.

[5] Gindl, W., H.S. Gupta, and C. Grünwald, "Lignification of spruce tracheid secondary cell walls related to longitudinal hardness and modulus of elasticity using nano-indentation", Canadian Journal of Bothanics, 2002. **80**: p. 1029-1033.

[6] Gindl, W. and T. Schöberl, "The significance of the elastic modulus of wood cell walls obtained from nanoindentation measurements", Composites: Part A, 2004. **35**: p. 1345-1349.

[7] Evans, R. and J. Ilic, "Rapid prediction of wood stiffness from microfibril angle and density", Forest Products Journal, 2001. **51**(3): p. 53-58.

[8] Clair, B., R. Arinero, G. Lévèque, M. Ramoda, and B. Thibaut, "Imaging the mechanical properties of wood cell layers by atomic force modulation microscopy", IAWA Journal, 2003. **24**(3): p. 223-230.

[9] Petryna, Y.S., D. Pfanner, F. Stangenberg, and W.B. Krätzig, "Reliability of reinforced concrete structures under fatigue", Reliability Engineering and System Safety, 2002. **77**(3): p. 253-261.

[10] Yang, J.L. and R. Evans, "Prediction of MOE of eucalypt wood from microfibril angle and density", Holz als Roh- und Werkstoff, 2003. **61**: p. 449-452.

[11] Peura, M., I. Grotkopp, H. Lemke, A. Vikkula, J. Laine, M. Müller, and R. Serimaa, "Negative Poisson Ratio of Crystalline Cellulose in Kraft Cooked Norway Spruce", Biomacromolecules, 2006. **7**(5): p. 1521-1528.

[12] Hoffmeyer, P. and R.W. Davidson, "Mechano-sorptive creep mechanism of wood in compression and bending", Wood Science and Technology, 1988. **23**(3): p. 215-227.

[13] Wypuch, G., "Handbook of Fillers, 2nd ed." ChemTec: Toronto, 1999: p. 890.

[14] Ristolainen, N., U. Vainio, S. Paavola, M. Torkkeli, R. Serimaa, and J. Seppälä, "Polypropylene/Organoclay Nanocomposites Compabilized with Hydroxyl-Functional Polypropylenes", Journal of Polymer Science: Part B: Polymer Physics, 2005. **43**: p. 1892-1903.

[15] Souza, P.E.N.d., C. Cusatis, C.K. Saul, A.R.D. Rodrigues, and P.C.d. Camargo, "Thick-mode resonance of a PZT/Si wafer stack investigated by X-ray diffraction in Bragg geometry", Journal of Applied Crystallography, 2003. **36**: p. 1144-1147.

[16] Beall, F.C., "Overview of the use of ultrasonic technologies in research on wood properties", Wood Science and Technology, 2004. **36**(3): p. 197-212.

[17] Sandoz, J.L., "Moisture content and temperature effect on ultrasound timber grading", Wood Science and Technology, 1993. **27**(5): p. 373-380.

[18] S. Andersson, "A study of the nanostructure of the cell wall of the tracheids of conifer xylem by x-ray scattering", Academic dissertation, University of Helsinki, 2007

[19] Bergander, A and Salmén, L, "Variability in cell wall properties and their effects on the mechanical properties of fibers", Wood and wood fiber composites, Otto-Graf-Institute, 2000: p. 3-13

F. Acknowledgements

We would like to acknowledge the valuable work done on the measurement setup by Mr. Pekka Pihkala and his crew.