VALIDATING THE APPLICABILITY OF POLARIZED LIGHT MICRSOCOPY FOR FIBER-FIBER BONDED AREA MEASUREMENT

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Introduction

Paper strength depends on the strength of single fibers and the strength of fiber-fiber bonds. These fiber-fiber bonds again depend on the size of the bonded area and on the specific bonding strength. Non-destructive measurement of the bonded area in a fiber-fiber bond is a key goal to determine the specific bonding strength (bonding force per unit area) between fibers.

In this study we will demonstrate that polarized light microscopy is a suitable tool for measuring the bonded area of individual fiber-fiber bonds. Polarized light microscopy, as developed by Page [1], was applied for fiber-fiber bonded area measurement. Page states that under polarized vertical illumination bonded areas appear dark while unbonded areas are bright. In earlier studies it has already been shown that polarized light microscopy shows the bonded area correctly, if one of the fibers in the fiber-fiber bond is dyed black [2]. Still, it is not applicable for undyed fibers [3]. We compare the results from polarized light microscopy to a measurement where the bonded area is determined using a 3D-representation of the fiber-fiber bond obtained with a microtome serial sectioning technique [4].

Material and Methods

Measuring the three-dimensional bonded area

The method for measuring the three-dimensional bonded area is based on microtome serial sectioning and image analysis [4]. The major steps of the method are shown in Fig 1.

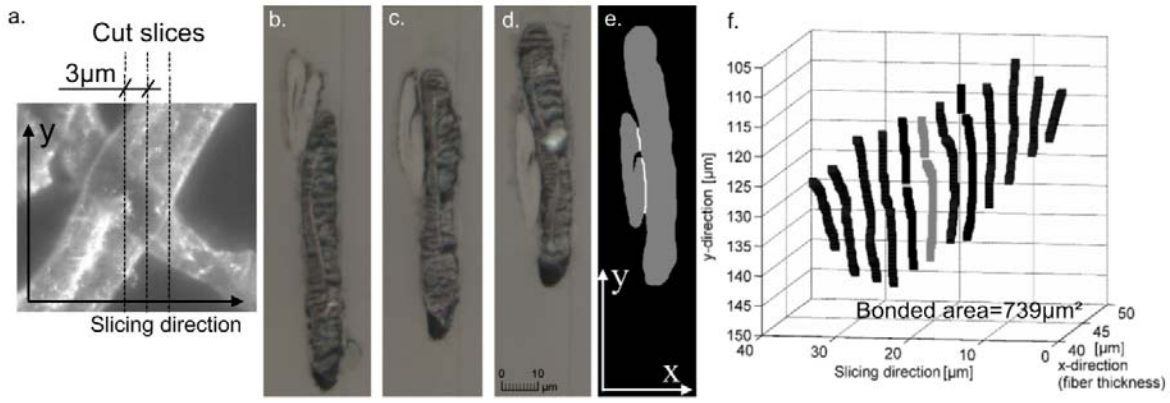

Fig 1: The same fiber-fiber bond under polarized light microscopy (a), in serial sectioning (b-d), after image analysis of one slice (e) and visualization of the 3D bonded area (f).

A single fiber-fiber bond (Fig 1(a)) is prepared according to Forsström [5], embedded in resin and clamped in a microtome. Slices with a thickness of 3 µm are cut off the embedded sample and the cutting area is imaged using an automated light microscope with an optical resolution of 0.161 µm/ pixel [6]. Fig 1(bcd) show images of the bond cross sections at different cutting positions. Fiber-fiber contact is determined image analytically, yielding a bonding line for every cut, as it is indicated by the white line in Fig 1(e). We are aware that the resolution of the optical microscope is too low to quantify whether the fibers really are in contact on a nanometer scale, we measure optically bonded area. Bonded area is calculated from bond line length and the cut thickness (3 µm). A three dimensional visualization of the bonding region is obtained by plotting these consecutive bonding lines (Fig 1 (f)).

The size and 3-dimensional structure of the optically bonded area is assessed together with cross sectional fiber morphology (fiber cross sectional area, fiber perimeter, fiber wall thickness, fiber collapse and fiber width). Holes and overlapping but unbonded regions of the fiber edges are also identified.

Measuring the bonded area with polarized light microscopy

Page [1] introduced polarized light microscopy for bonded area measurement of fiber-fiber bonds. The phenomenology of the method can be described as follows. Light is linearly polarized, so waves with only one plane of oscillation are directed through the objective and to the surface of the sample, where it is reflected. The second polarizing filter is rotated 90° towards the first one, so that only optically modified light is able to pass it. Light that is reflected at surfaces under vertical illumination does not change its state of polarization. Therefore reflexes at surfaces are of negligible intensity. The geometry of the optical system at the fiber-fiber bond yields very small reflected over-all intensities, so that the fiber-fiber bonds appear dark, while the inner reflection at the back surface of a single fiber yields reflected light with quite a high intensity and a changed polarization direction. Thus single fibers appear bright.

The polarized light microscopy investigations were performed with a Leica Leitz DMRX microscope, which was equipped with crossed polarizing filters. A 20-fold objective with an optical resolution of 0.255 µm/Pixel was used. The images were taken with a Leica DFC290 camera. The fiber-fiber bonds were imaged in eight different rotary positions, the image with the highest contrast was chosen for further analysis.

The dark bonded area was drawn manually and the amount of pixels in this area was determined using image analysis. Then the size of bonded area was calculated with the pixel size.

The bonded area was measured with polarized light microscopy and with microtomy. The size and the shape of the bonded area were compared for the beaten and for the unbeaten pulp.

Explaining the deviations between the methods

Based on the measured morphological parameters several factors that may explain the deviations between the bonded areas measured with the two different methods were investigated. Multiple linear regression was performed to quantify the significance (F*-statistics) and impact (ANOVA) of the predictor variables [7]. The deviation between the measurement results was the response variable, all morphological parameters and the degree of bonding (holes and overlapping but unbonded regions of the fiber edges) were the predictor variables.

Used Pulp

All experiments were performed with an unbleached pulp from spruce and pine wood. Fiber-fiber bonds were prepared from unbeaten pulp as well as from pulp that was beaten with 9000 revolutions in the PFI mill. 50 % of the pulp was dyed with Chlorazol Black and bonds between one dyed and one undyed fiber were formed.

Results

Comparison of bonded area for unbeaten pulp

Bonded area of 73 fiber-fiber bonds was measured with polarized light microscopy and with the microtome serial sectioning method, the results are compared in Fig 2. The diagonal indicates equal values. In general the values show quite good agreement, although bonded area is overestimated more often by polarized light microscopy. On average the values measured with polarized light microscopy were 12.5 % higher for the investigated bonds.

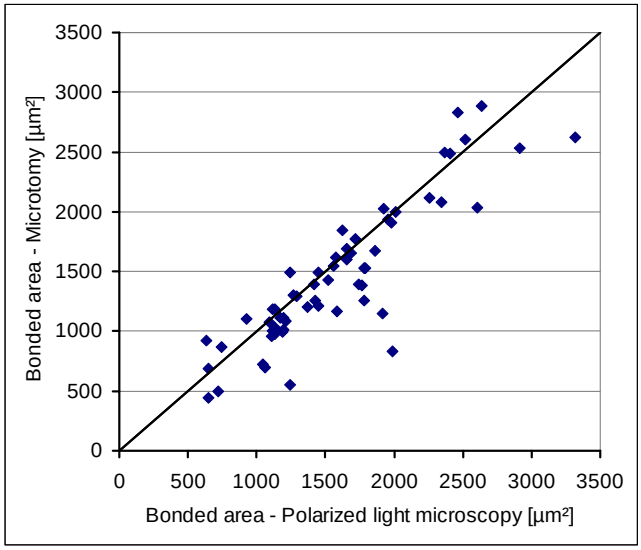

Fig 2: Comparison of bonded area measured with polarized light microscopy and with microtomy – unbeaten pulp

To explain this overestimation of bonded area a multiple linear regression model was built. Table 1 shows the significance (p-value of the F*-statistics) and the impact (ANOVA) of the variables. The results of the model are interpreted on basis of the stepwise R^2 values, i.e. the ANOVA. Because of redundancies and interactions the R^2 values alone are not really meaningful and are not used for interpretation of the results. The p-value shows the significance on a 95 % confidence level. Only the significant parameters are given in Table 1. It can be shown that unbonded fiber edges account for 56.7 % of the deviations (the R^2 value is 0.567). By adding the fraction of holes in the bond to the model, the stepwise R^2 value is increases from 0.567 to 0.590, indicating that the holes in the bond explain further 2.3 % of the deviations. All other morphological parameters are not significant on a 95 % confidence level (i.e. $p>0.05$).

Table 1: Results of multiple linear regression and ANOVA with the deviation of bonded area between microtomy and polarized light microscopy as the response variable

Variable	R^2 stepwise	R^2 alone	p-Value	Sign of Coeff.
Unbonded fiber edges	0.567	0.567	$<10^{-5}$	-
Holes in the bond	0.590	0.092	0.03	-

The model results indicate that unbonded fibers cannot always be identified correctly with polarized light microscopy and lead to overestimation of the bonded area. The reason why unbonded fiber regions can only be identified in some cases is still under study.

Fig 3 shows an example where the fiber-fiber bonded area is overestimated with polarized light microscopy. The microtome image (left) shows that the left fiber is folded in the lower part of the bond. Here the fibers are not bonded, a small gap between the fibers is evident (see scale up in

Fig 3, left). This unbonded fiber edges do not show under the polarized light microscope. It is rather suggested that the bonded area reaches over the border of the fiber-fiber crossing region, as the fold also appears as bonded fiber (marked with the white arrow, right).

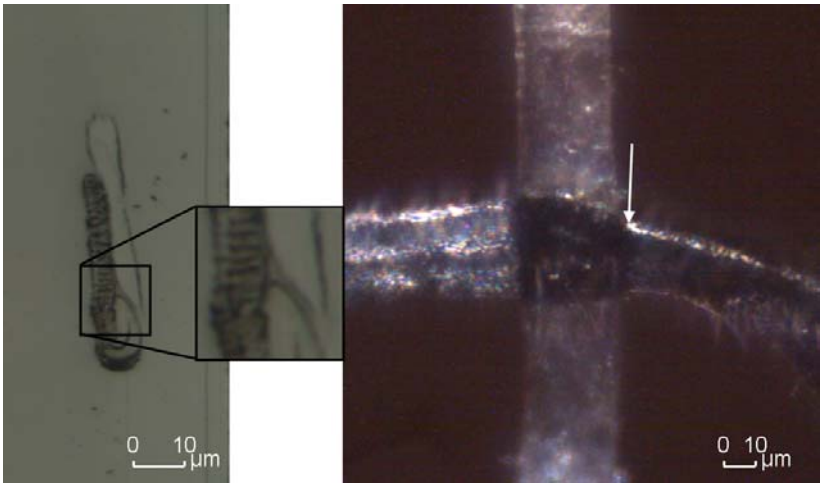

Fig 3: Overestimation of bonded area with polarized light microscopy: the fold appears dark under polarized light, although the fibers are unbonded in this region (marked with white arrow)

Comparison of bonded area for beaten pulp

Fiber-fiber bonds of a highly beaten pulp were analyzed first with the polarized light microscopy and then with the microtome. The size and the shape of bonded area of 68 fiber-fiber bonds were compared. In Fig 4 bonded area measured with polarized light microscopy is plotted versus bonded area measured with the microtome. The diagonal indicates equal values. In contrast to the unbeaten case, now bonded area is underestimated by polarized light microscopy in most cases (on average by 15.9 %).

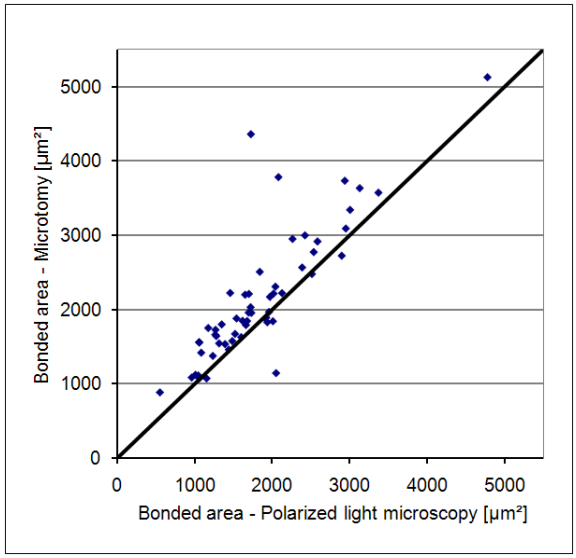

Fig 4: Comparison of bonded area measured with polarized light microscopy and with microtomy – beaten pulp

To explain the deviations between the measurement results again multiple linear regression modeling was performed. The mathematical model showed that not any of the morphological parameters has a statistically significant influence on the deviations on a 95 % confidence level.

It seems as if most of the deviations between the measurement results can be explained only with the fibrils on the beaten pulp fiber surfaces, which cannot be reproduced with any of the applied measurement methods. The fibrils cause additional reflections, so that the polarized light microscope images are brighter in total. In the crossing region these bright reflections are evaluated as unbonded fiber edges and holes in the bond.

An exemplary comparison of a polarized light microscope image with the fiber-fiber bond cross section is shown in Fig 5. The center of the bond appears brighter under the polarized light microscope (right), suggesting that the fibers are unbonded in this area. Also the fiber edges appear as unbonded under the polarized light. However, the microtome image on the left shows that the fibers are optically bonded over the entire length. This comparison confirms the assumption that the additional reflections caused by the fibrils are the origin of the underestimation of the bonded area in the case of beaten pulp.

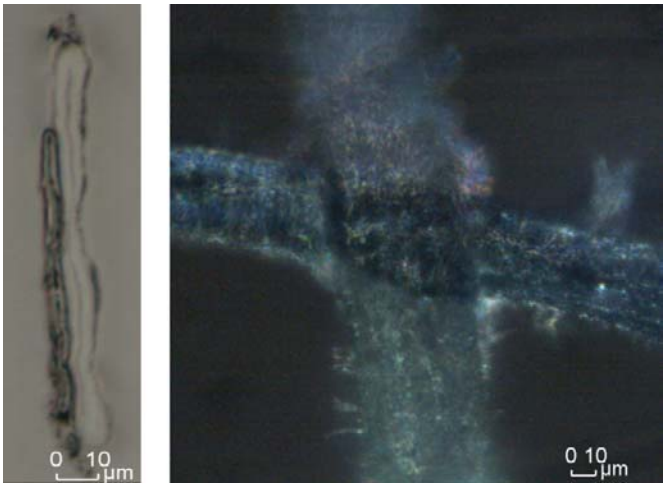

Fig 5: Underestimation of bonded area with polarized light microscopy: fibrils on the fiber surface are evaluated as holes in the bond and as unbonded fiber edges

Conclusions

The comparison of the two different methods showed that polarized light microscopy is an appropriate tool for fiber-fiber bonded area measurement. However, for unbeaten pulp bonded area is over- and for beaten pulp it is underestimated. If these deviations are considered polarized light microscopy provides a non-destructive method for bonded area measurement. It can be applied in combination with micro tensile testing of fiber-fiber bonds in order to determine the specific bonding strength.

Acknowledgements

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