

A Proposed Failure Mechanism for Pulp Fiber-Fiber Joints

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Due to stress concentration at the edges, fiber-fiber bonds under load are known to fail gradually inwards from the edges. In this paper, we propose a failure mechanism for fiber-fiber joints under load, based on the peak stresses occurring at the bond edges. We have modeled the mechanical testing of individual fiber-fiber joints using a finite element method (FEM) framework. The model is based on experimental results of fiber-fiber joint strength tests designed to induce each of the three modes in fracture mechanics: opening, sliding, and tearing. A parametric study of the peak load at the edges of the fibers was carried out in order to identify a failure mechanism. The peak stresses were not directly taken from the FEM models, as these values are highly discretization-dependent. Instead, the peak stresses were estimated from resultant forces and moments in the bond and an idealized geometry of the bonding region. The literature has, up to now, focused on shear load as a failure mechanism for fiber-fiber bonds. However, our findings indicate that pulp fiber joints are sensitive to normal stresses and insensitive to shear stresses. Hence, we suggest utilizing failure criteria related to normal stress in future work.

Keywords: Failure criteria; Interfiber joint strength; Fiber-fiber bond; Shear stress; Normal stress

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INTRODUCTION

The bonding strength between pulp fibers in paper is one of the key parameters determining the strength of the paper. It is not possible to measure fiber-fiber bond strength reliably from paper sheets because paper strength also depends on other factors *e.g.* fiber length, fiber tensile strength, paper density, and straining during drying of the sheet. Therefore, fiber-fiber bond strength is usually investigated by measuring the bond strength of individual fiber-fiber joints (Schniewind *et al.* 1964; Saketi and Kallio 2011; Fischer *et al.* 2012; Schmied *et al.* 2012; Saketi *et al.* 2012; Magnusson *et al.* 2013b). It might be intuitive to think that the breaking load (in N) of a fiber-fiber joint is composed of a specific bond strength (bonding force per unit area, N/m²) times the bonded area (in m²) of the fiber-fiber joint. This, however, is not the case. Stress concentrations occur at the edges of the bonding area (Button 1979; Uesaka 1984; Page 2002), which leads to a progressive failure of the fiber-fiber bonds starting at the peak stress regions. This progressive failure has also been observed in fiber-fiber joint testing, where sudden drops in loading force indicate local failure of the bond (Uesaka 1984; Magnusson *et al.* 2013b; Schmied *et al.* 2013).

52 There is considerable evidence that failure in paper also occurs due to progressive
53 failure of fiber-fiber bonds. Nordman *et al.* (1952) found that the light scattering coefficient
54 of paper increases upon straining. The increase in light scattering can be attributed to new
55 surface area created in the paper due to the separation of previously bonded fiber regions
56 (Page 2002). Investigations of fiber-fiber bonds in paper using polarized light microscopy
57 have shown that the bonds indeed fail progressively from the edges inward under dynamic
58 load (Page *et al.* 1962) as well as under constant load, *i.e.* creep testing (DeMaio *et al.*
59 2006).

60 It is the aim of this work to propose a key mechanism of fiber-fiber bond failure
61 based on the peak stresses occurring at the edges of the bonds. Progressive failure is always
62 initiated by the peak stresses in the structure. Therefore, failure theories give a criterion for
63 yield or fracture in the material by providing a scalar representation of a multiaxial state of
64 stress, *i.e.* the normal and shear stress are combined into a single value (Brinson and
65 Brinson 2008; Pruitt and Chakravartula 2011). It is important to understand that, in many
66 respects, the behavior of pulp fiber is fundamentally different from classical engineering
67 materials. Typically, pulp fibers possess a sophisticated hierarchical micro-structure
68 (Bodig and Benjamin 1993). Therefore, classical failure theories may not directly apply.
69 Collagen, like pulp fibers, is a viscoelastic, fibril-based biomaterial. It has been well
70 researched, because of its relevance regarding defects and surgery of blood vessels. Still,
71 no conclusive failure mechanism has been worked out for this material, although several
72 different failure mechanisms have been discussed (Wang *et al.* 1997; Gasser 2011).
73 Recently, a comprehensive finite element method (FEM) framework to model the behavior
74 of fiber-fiber joints during mechanical testing was presented by Magnusson *et al.* (2013).
75 The work focused on resultant forces and moments in the bonding regions and did not
76 consider local stress concentrations. Based on that, they discussed a failure criterion
77 according to which the bonds are more sensitive to shear load than to normal load. For
78 further work they recommended incorporation of local stress variations, *e.g.* by cohesive
79 zone modeling. In a recent review (Da Silva and Campilho 2012) on cohesive zone
80 modeling, several different failure models are discussed for fiber-based composites, the
81 literature reviewed there also does not permit a general recommendation for the case of
82 pulp fibers.

83 In this work, we will propose a key mechanism of pulp fiber-fiber bond failure
84 based on the analysis of peak stresses inferred from FEM models of fiber-fiber bond
85 mechanical testing. We have conducted three different types of fiber-fiber bond strength
86 measurements, each one designed to predominantly load the fiber-fiber joint in one of the
87 three fracture modes, see Fig. 1. The parameters for the FEM models are taken from
88 previous experiments and literature (Magnusson *et al.* 2013b). Several parameters that
89 represent the characteristic features of the pair of bonded fibers are defined. These are fiber
90 thickness t , fiber width w , fiber fibril angle ψ , and crossing angle Φ of the fiber-fiber joint.
91 These parameters are varied in physically meaningful ranges in a parametric study for three
92 different types of loading, which correspond to mode 1, mode 2, and mode 3 types of
93 fracture. The applied loading in the numerical model was taken from the corresponding
94 experimental results at rupture. For each parameter set and type of loading, the arising
95 resultant shear and normal forces as well as the resultant opening, twisting, and tearing
96 moments in the bond region were obtained with the help of a FEM model in ABAQUS
97 (2012). These resultant forces and moments are employed to calculate estimated normal
98 and shear stress distributions in the interfiber joint based on a simplified model of the fiber-

99 fiber joint geometry. Based on that, one can identify peak values for normal and shear stress
100 for each parameter set.

101 The paper is organized as follows. First, the experiments on fiber-fiber joints are
102 described, which provide the experimentally obtained parameters for the FEM model.
103 Next, the methods section gives details about the FEM discretization and the computation
104 of the estimated normal and shear stress distributions in the bonding region. Furthermore,
105 the obtained peak stresses for the three types of loading are presented. The results section
106 presents a surprising behavior: while the obtained peak values for normal stress are within
107 the same range for the three types of loading, the peak values for shear stress are clearly in
108 different ranges. This suggests that normal stress plays an important role in the failure of
109 pulp fiber-fiber bonds.

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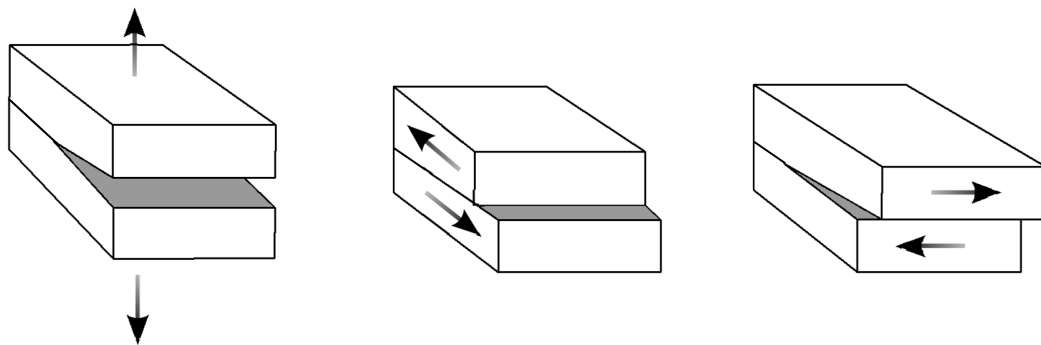
111

112 EXPERIMENTAL

113

114 In fracture mechanics, there are different modes of fracture (see Fig. 1). Cracks may
115 propagate in the plane perpendicular to normal stress (mode 1, opening), in the plane with
116 shear stresses with the crack line perpendicular to the stresses (mode 2, sliding), or in the
117 plane with shear stress with the crack line parallel to the shear stress (mode 3, tearing). In
118 single fiber testing, we have performed experiments to specifically address these different
119 fracture modes.

120



121

122 (a) Mode 1 (opening)

123

122 (b) Mode 2 (sliding)

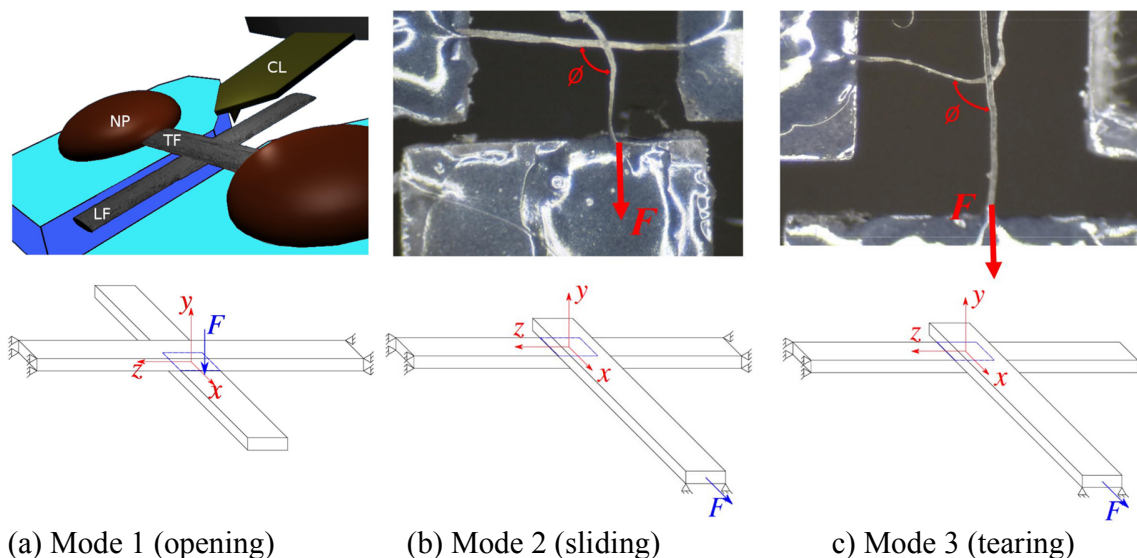
122 (c) Mode 3 (tearing)

124 **Fig. 1.** Illustration of the three fracture modes in fracture mechanics

125

126 The experimental setup for this work is shown in Fig. 2. The details for the
127 experimental procedure for mode 1 are described by Schmied *et al.* (2012) and for modes
128 2 and 3 by Fischer *et al.* (2012). In short the setups are as follows. For mode 1 an atomic
129 force microscope (AFM) is used. The fiber-fiber bond to be tested is fixed via the top fiber
130 (TF in Fig. 2(a)) on two sides using nail polish (NP in Fig. 2(a)). The lower fiber (LF in
131 Fig. 2(a)) is therefore only held by the fiber bond. Then the AFM cantilever (CL in Fig.
132 2(a)) is used to push down the lower fiber (LF). The loading force on the lower fiber is
133 measured with the AFM, with recording of force-distance curves. The load is applied in
134 closest possible proximity to the fiber-fiber bonding region, thus leading to a loading
135 situation very similar to opening mode. For mode 2 and 3 testing the fiber bonds are glued
136 to an acrylic holder. For mode 2 the vertical fiber is glued on both sides of the holder (top

137 part in Fig. 2(b)) and the vertical free fiber is glued to the moving part (lower part in Fig.
 138 2(b)) to apply the force. The force distance data is obtained via a linear table, a microscope
 139 camera, and a strain gauge. For mode 3 the horizontal fiber is only fixed on one side (left
 140 side in Fig. 2(c)). Otherwise the system is identical to the mode 2 tests. The setup in Fig.
 141 2(a) gives rise to a predominantly mode 1 load, the setup in Fig. 2(b) gives a predominantly
 142 mode 2 load, and the setup in Fig. 2(c) creates a predominantly mode 3 load. Please note
 143 that the configurations shown in Fig. 2 do not result in pure loadings according to modes
 144 1, 2, and 3. Due to the curved geometry of the fibers, fiber twisting during the experiment,
 145 and the tilting of the fibril angle to the fiber axis, there is a large amount of opening,
 146 twisting, and tearing load on the bonding region in all three experiments (Magnusson *et al.*
 147 2013a, b).
 148



149 (a) Mode 1 (opening)

(b) Mode 2 (sliding)

c) Mode 3 (tearing)

150
 151 **Fig. 2.** The experimental setup for the three modes

152
 153 The geometry of the specimens was captured by micrographs. Furthermore, the
 154 applied force at rupture was measured. For all experiments unbleached and unrefined
 155 softwood kraft pulp fibers were used. The fiber bonds were made from highly diluted
 156 suspension put between Teflon foils in a standard lab sheet former. Therefore, all the fibers
 157 tested were collapsed. This was also checked by microscopy. For further details, we refer
 158 to (Kappel 2009).
 159

160

161

161 METHODS

162

163 The objective of this work is to study the essential characteristic of fiber-fiber
 164 bonds. As mentioned in the introduction, the numerical investigation of real fibers is very
 165 challenging due to the uniqueness of each real fiber. Hence, our goal is to develop a
 166 numerical model that keeps the principal characteristics, but neglects superfluous details.
 167 The proposed numerical model is still based on experimental data, but avoids the
 168 interference with random characteristics of individual fibers. Furthermore, it allows us to
 169 make predictions on the basis of features that all fibers share.

170 Geometric Discretization, Material Behavior, and Loading

171 The cell wall of pulp fibers consists of four major layers; the primary wall and three
 172 secondary layers (S_1 , S_2 , S_3), as shown in Fig. 3. All layers are composed of cellulose,
 173 hemicellulose, and lignin in varying compositions (Bodig and Benjamin 1993).
 174 Furthermore, each secondary layer shows a micro-fibril wrapped helically along the fiber
 175 at a specific angle. The fiber's cell wall is made of up to 80-85% of the S_2 layer (Page
 176 1969a), and it is commonly assumed in literature that this layer has the highest influence
 177 on the fiber's mechanical behavior (e.g. Magnusson and Östlund 2013). Therefore, the pulp
 178 fiber will be modeled by the S_2 layer only.

179 Each real pair of bonded fibers is unique. It will differ from any other pair in terms
 180 of geometry and material properties. Therefore, a system that is reduced to a minimal set
 181 of parameters is chosen to study the distinct influence of the model parameters. The fiber-
 182 fiber cross was modeled as two straight beams. Use of such a model is tantamount to
 183 neglecting the curvature and the twist along the fiber direction (Seth 2006). The model
 184 parameters were chosen to be the width w , the thickness t , and the fibril angle ψ of the
 185 fibers (Fig. 4). Furthermore, the crossing angle Φ of both fibers is investigated (Fig. 2 b,c).
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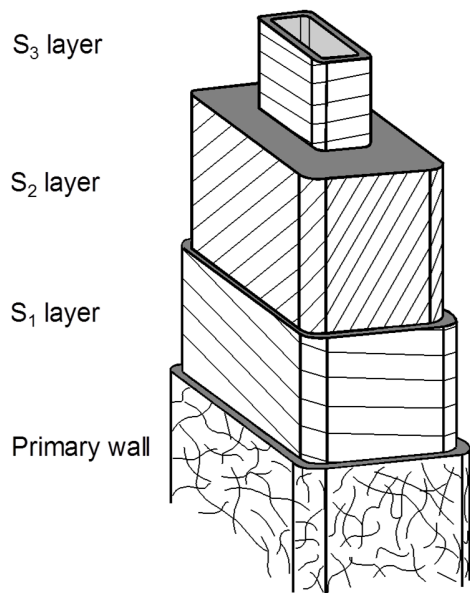


Fig. 3. The layered structure of a single pulp fiber

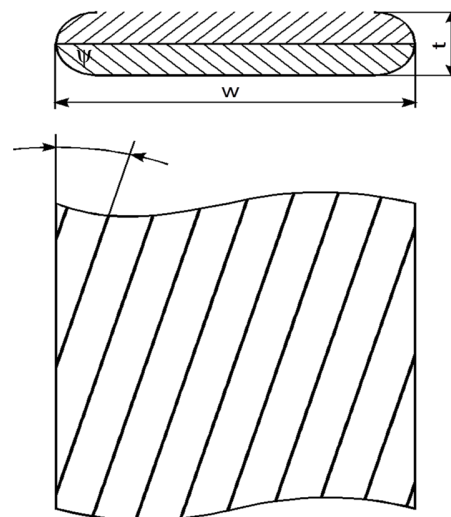


Fig. 4. Cross section and geometry of the idealized fiber structure

187
 188 The fibers were considered as fully collapsed volumetric bodies. The cross section
 189 of the idealized fiber model is given in Fig. 4. Each fiber consisted of two parts with the
 190 micro-fibril pointing in opposite directions in each part. If the upper part showed an angle
 191 of $\psi = 30^\circ$, then the lower part had -30° . The micro-fibril angle was expected to be constant
 192 along the fiber length. Furthermore, the length of the fibers was 1 mm, to be in close
 193 agreement with the previously described experiments. In all performed computations, the
 194 loaded fiber was positioned right in the middle of the fixed fiber.

195 The material behavior of the fiber (modeled by the S_2 layer only) was chosen to be
 196 transversely isotropic in the model. This material law considered the effect of the micro
 197 structure of the fiber. The micro-fibrils acted as a reinforcement in the matrix of lignin and

198 hemicellulose. The axis of transverse isotropy was aligned with the direction of the micro-
199 fibril.

200 The material constants as used in the simulation are shown in Table 1. The modulus
201 of elasticity $E_1=30\text{GPa}$ was chosen as an average of the data given by Magnusson and
202 Östlund (2013). It has to be mentioned that the material properties of the S_2 layer are subject
203 to wide variations (Page *et al.* 1977; Groom *et al.* 1995; Neagu *et al.* 2004). As there is not
204 enough material data available for such a model, we are neglecting the viscoelastic nature
205 of pulp fibers and we assume the fiber will behave according to the previously described
206 anisotropic elastic model.

207

208 **Table 1.** Material Constants of the Cell Wall (Magnusson and Östlund 2013).

Coefficient	E_1	$E_2 = E_3$	$G_{12} = G_{13}$	G_{23}	$\nu_{12} = \nu_{13}$	ν_{23}
Value	E_1	$\frac{E_1}{11}$	$\frac{E_1}{23}$	$\frac{E_2}{2(1 + \nu_{23})}$	0.022	0.39

209

210 Three different modes of loading were tested according to the experiments
211 described in the previous section. The three models of the various modes, their boundary
212 conditions, and the direction of the applied force can be seen in Fig. 2. In modes 2 and 3,
213 the load was applied in x-direction. If the crossing angle Φ was different to 90° and thereby
214 the axis of the loaded fiber was not aligned to the x-direction, then the force was still
215 applied in x-direction. In mode 1, the applied force pointed into the negative y-direction.
216 We assumed the load to rupture the bonding region to be much smaller than the load to
217 plastically deform or even rupture the fiber (Burgert *et al.* 2003). Hence, the bonding region
218 was the predetermined breaking point of the structure.

219

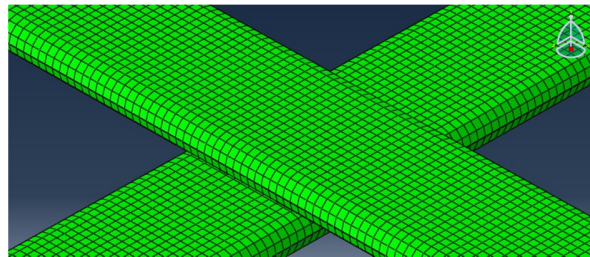
220 Finite Element Discretization

221 The commercial FEM software ABAQUS (2012, version 6.11-2) and its scripting
222 interface in Python were used to perform the non-linear quasi-static FEM model
223 simulations. The pair of bonded fibers was discretized using a mesh consisting of 8-noded
224 hexahedral elements with reduced integration (C3D8R in the ABAQUS element library).
225 A mesh size dependency check was performed, and the elements' size was chosen to render
226 the deviation in the results to be practically insignificant.

227 The FEM model assumed the contact area to be fully bonded, which was considered
228 unlikely for real bonded fibers (*e.g.* Page 1960). Regions close to the edge of the bonding
229 region, or even in the interior of the bonding region, may not be molecularly bonded (Page
230 1960; Kappel *et al.* 2009).

231 It is discussed in Torgnysdotter *et al.* (2007a, b) that the degree of contact is of great
232 importance for the maximum stress in the bonding region. In contrast, recent results show
233 that there is a high degree of bonding between fiber surfaces (Persson *et al.* 2013, Hirn *et al.*
234 *et al.* 2013, Hirn and Schennach 2015). Therefore, we neglected possible flaws in the bonding
235 for reasons of simplification. Furthermore, we assumed that the contact zone did not change
236 before rupture. As a result, the two surfaces of the fibers in contact were tied to each other
237 by a surface-to-surface contact discretization using tie constraints in the FEM software
238 ABAQUS. An example of the meshed pair of bonded fibers used for all three loading types
239 is given in Fig. 5.

240



241
242 **Fig. 5.** Finite element model of the fiber-fiber bond.

243
244 **Resultant Forces and Moments in the Bonding Region**

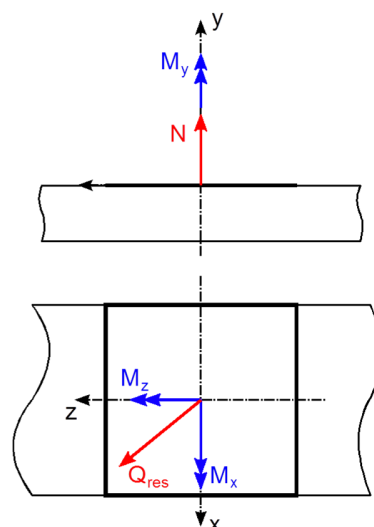
245 The applied loading caused resultant reaction forces and moments in the bonding
246 region compared with the similar treatment in Magnusson and Östlund (2013). These were
247 described in a local coordinate system, the origin of which was defined at the centroid of
248 the interface region. As already shown in Fig. 2, the y -axis was defined by the outward unit
249 normal, z was defined in direction of the fixed fiber, and x was orthogonal to the previous
250 two directions. The resultant reaction forces and moments in coordinate directions were
251 computed. The resultant forces N (normal force), Q_x and Q_z (shear forces in x - and z -
252 direction) were calculated by adding up the the nodal forces in the bonding region (NFORC
253 in ABAQUS) as follows. The quantities N_i , Q_{xi} , and Q_{zi} were the nodal forces at node i (for
254 n nodes in the bonding region) in y -, x -, and z -directions, respectively. The resultant
255 reaction forces were computed using the three equations:

$$256 \quad N = \sum_{i=1}^n N_i, \quad Q_x = \sum_{i=1}^n Q_{xi}, \quad Q_z = \sum_{i=1}^n Q_{zi} \quad (1)$$

257 The three resultant moments M_x , M_y , and M_z in the local coordinate system were then
258 obtained from the three relations:

$$259 \quad M_x = \sum_{i=1}^n -z_i N_i, \quad M_z = \sum_{i=1}^n x_i N_i, \quad M_y = \sum_{i=1}^n z_i Q_{xi} - x_i Q_{zi} \quad (2)$$

260 The quantities x_i and z_i were the perpendicular distances of the nodal forces to the origin
261 of the coordinate system. Figure 6 gives a visualization of the resultant forces and moments
262 in the bonding region.



267
268 **Fig. 6.** Resultant forces and moments in the bonding region

269 Q_{res} in Fig. 6 was obtained from the equation:

$$270 \quad Q_{res} = \sqrt{Q_x^2 + Q_z^2} \quad (3)$$

272

273 Resultant Stresses in the Bonding Region

274 Although it may appear straightforward, the peak stresses extracted directly from
 275 the FEM model of the fiber-fiber joints needed to be treated with care. For a detailed
 276 discussion on this topic, please refer to Da Silva and Campilho (2012). The peak stresses
 277 were typically found to be close to the stress discontinuities of the model, *i.e.* sharp corners
 278 or interfaces with different material properties. This was also shown by Magnusson *et al*
 279 (2013a). In the present case, this was where the rounded edge of one fiber touched the
 280 surface of the other fiber (compare Fig. 5). The magnitude of the peak stresses in the FEM
 281 model strongly depended on how well the stress field was modeled around these
 282 discontinuities. Specifically, it was very sensitive to both the mesh size used and the
 283 considered geometrical details in the model. In particular, the latter could not be
 284 appropriately met in any simplified fiber-fiber model. Therefore, we refrained from
 285 extracting the peak stresses directly from the model. Instead we applied the resulting forces
 286 and moments, as described in the previous section, to estimate the peak stresses using an
 287 idealized model of the bonding region.

288 The actual stress situation in fiber-fiber joints was simplified by neglecting local
 289 unbonded regions and irregularities in the fiber geometries. These simplifications were
 290 expected to lead to deviations from the reality in terms of absolute stresses. It was, however,
 291 not the present goal to correctly model the absolute values of the peak stresses or fit the
 292 experimental results to the FEM model. Instead the goal was to extract the general behavior
 293 of the peak stresses and the relation between shear- and normal stresses. This generalization
 294 was achieved, on the one hand, by simplifying the geometry of the model and, on the other
 295 hand by varying the parameters for fiber-fiber bond configurations in a wide range (see
 296 Table 2). Nevertheless, it is worth pointing out that the simplification only relates to the
 297 rectangular geometry of the bonding zone and the negligence of edge effects creating stress
 298 discontinuities, the calculation of the stresses followed standard procedures in mechanics.
 299 The presented approach computed idealized stress distributions (constant for tensile and
 300 shear loading, linear for bending and torsion) and obtained a single estimated peak value
 301 for the normal stress and a single estimated peak value for the shear stress for each pair of
 302 fibers. This allowed for an easy comparison of very different geometrical settings.

303 The interfacial region between the fibers in a joint was defined by the area A of the
 304 bonding region, the second area moment of inertia I for bending, and the polar section
 305 modulus W_p for torsion. As can be seen in Fig. 4, a single fiber had a radius at the edge.
 306 This had to be taken into account when the length of the bonding region was determined.
 307 Therefore, the length of the bonding region had the value $w-t$. For two orthogonal fibers
 308 (crossing angle $\Phi=90^\circ$) it was found that:

309

$$310 \quad A = (w - t)^2, \quad I = \frac{(w-t)^3(w-t)}{12}, \quad W_p = 0.208(w - t)^3 \quad (4)$$

311

312 The presented formula for W_p was valid only for a square section area (Grote and Feldhusen
 313 2011). If the crossing angle Φ was different from 90° , the bonding region A changed to a
 314 rhomboid. For this case, the area A and the second area moments of inertia I_1 and I_2 were

315 found analytically, and the torsion constant W_p was numerically computed for principal
316 axes.

317 The estimated normal stress distribution σ_N , according to the resultant normal force,
318 was constant:
319

$$320 \quad \sigma_N = \frac{N}{A} \quad (5)$$

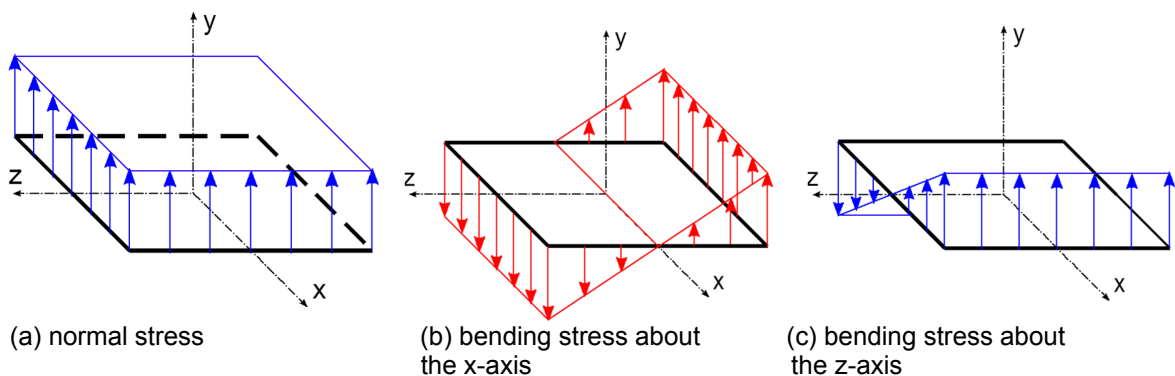
322 Next, the contribution to the normal stress due to bending σ_B for orthogonal fibers was
323 computed as follows with M_z and M_x being the moments defined above:
324

$$325 \quad \sigma_B = \frac{M_z}{I} x - \frac{M_x}{I} z \quad (6)$$

326
327 If the crossing angle Φ was different to 90° , σ_B was set up in principal axes. The total
328 normal stress distribution σ_{res} was given as,
329

$$330 \quad \sigma_{res} = \sigma_N + \sigma_B \quad (7)$$

332 and is visualized in Fig. 7. The maximum of the total normal stress was obtained by
333 computing its value at the corresponding corner of the bonding region.
334



335 (a) normal stress

336

337

338 **Fig. 7.** Components of normal stress

339

340 The estimated shear stress distribution τ_{Qres} according to the resultant shear forces
341 was assumed to be constant over the bonding region:
342

$$343 \quad \tau_{Qres} = \frac{1}{A} \sqrt{Q_x^2 + Q_z^2} \quad (8)$$

344

345 Furthermore, the maximum shear stress due to torsion τ_T was computed as:
346

$$347 \quad \tau_T = \frac{M_y}{W_p} \quad (9)$$

348

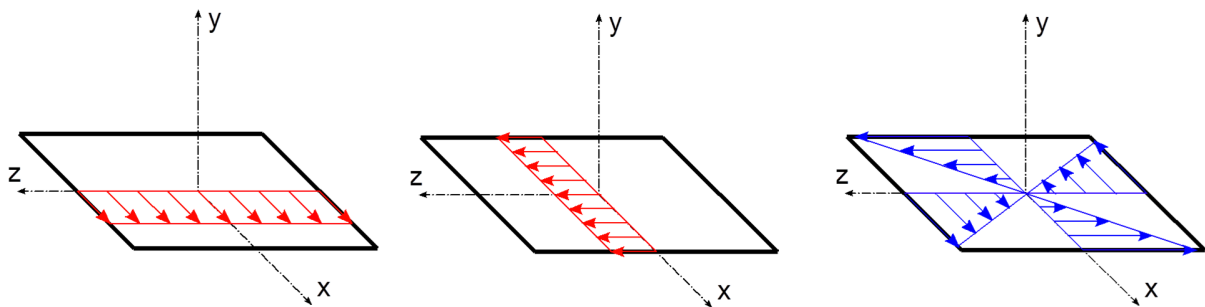
349 If the crossing angle Φ was different to 90° , τ_T was numerically computed. The maximum
350 value of the total shear stress τ_{res} was found as,
351

$$352 \quad \tau_{res} = \tau_{Qres} + \tau_T$$

353

354 and is visualized in Fig. 8.

355



356

357 (a) shear stress in x-direction (b) shear stress in z-direction

358

(c) torsional shear stress
about the y-axis359 **Fig. 8.** Components of shear stress.

360

361

362 **RESULTS AND DISCUSSION**

363

364

365 A parametric study was performed with the numerical model. The mean fiber width
366 of the pulp was found to be 32.00 μm , and the mean fiber thickness equaled 7.45 μm . The
367 experimentally obtained mean force in mode 1 equaled 0.33 mN (Schmied *et al.* 2013).
368 The mean force for mode 2 was 6.45 mN, and for mode 3 it was 1.06 mN (Fischer *et al.*
369 2012). The ranges of the varied parameters in the numerical model are listed in Table 2
370 (fibril angle ψ , fiber thickness t , fiber width w , and bonding angle Φ). The applied load
371 was taken according to the experimental results. All the conclusions drawn in the following
372 paragraphs refer to the unbleached unrefined softwood fibers used in the experiments.

372

373 **Table 2.** Ranges of Modified Parameters.

	t [μm]	w [μm]	ψ [$^\circ$]	Φ [$^\circ$]
Range	4.65-10.25	25.20-45.60	0-45	60-120
Increments	0.70	3.40	5	5

374

375

376 The obtained peak values for normal and shear stresses for each parameter set and
377 type of loading were collected and are presented in Fig. 9. The occurrence of each symbol
378 (\times , $+$, \square) in this figure stands for a parameter set, where the shear stress in the bond is
379 plotted against the normal stress in the bond. The three types of loading are denoted as M1,
380 M2, and M3 (by referring to the corresponding fracture mode). The different points show
381 the varied parameters fibril angle ψ (\times in Fig. 9), fiber thickness t ($+$ in Fig. 9), fiber width
382 w (\diamond in Fig. 9), and bonding angle Φ (\square in Fig. 9). Furthermore, a dependency check on
383 the applied load, Young's modulus, and Poisson's ratio (see Table 1) was performed by
384 varying a single quantity and keeping the remaining parameters unchanged. The results
385 were essentially equivalent to Fig. 9, and thus we refrained from reproducing them here.
386 For each mode a "core region" of the estimated peak stresses, which contains most of the
387 data points, can be identified in the plane of normal stress and shear stress. These core
388 regions are marked by circles in Fig. 9.

388

389 These results showed that the obtained peak values for normal stress were within
390 the same range between about 2 to 10 MPa for all three types of loading (all three core
391 regions in Fig. 9 are in this range).

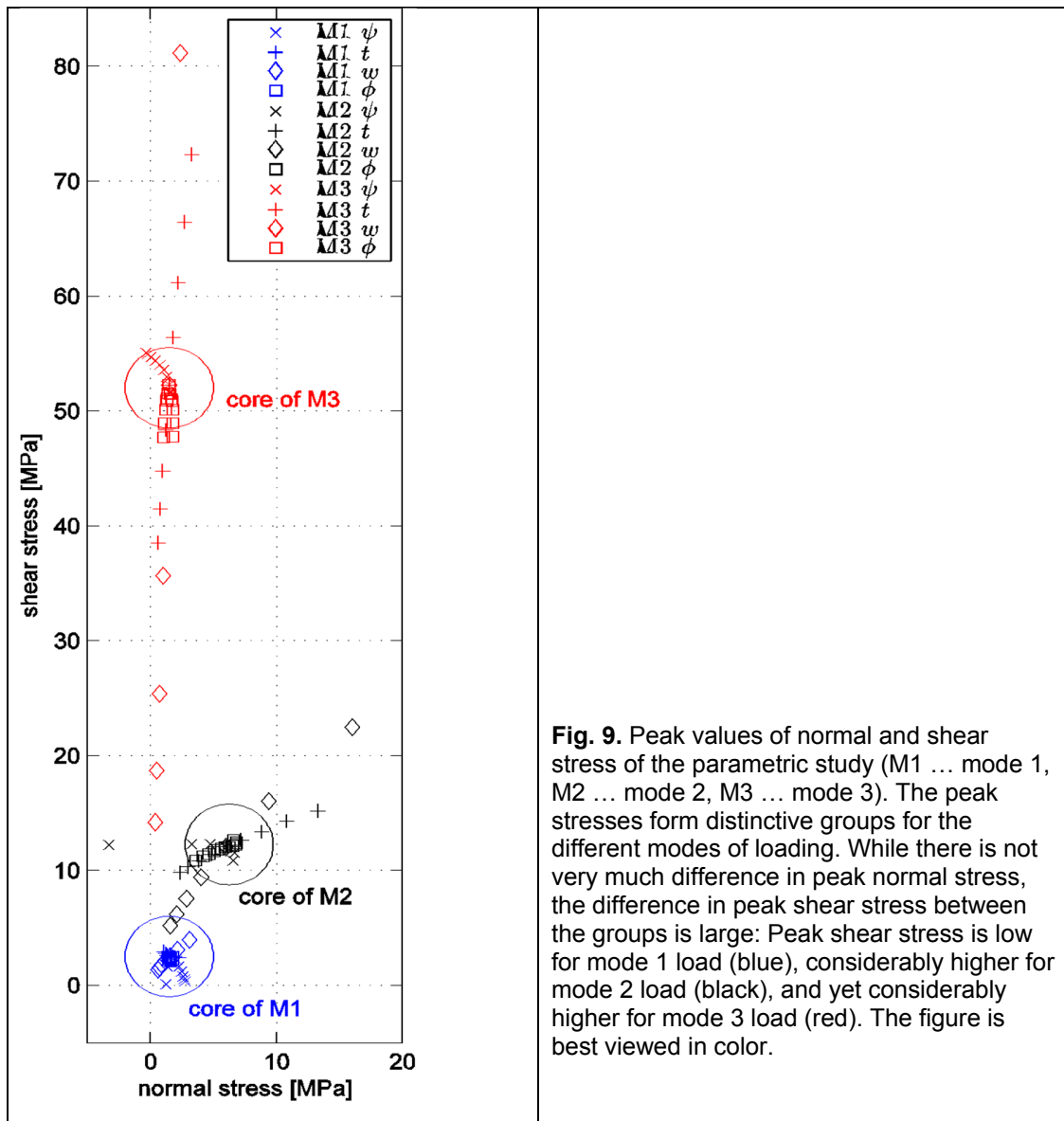


Fig. 9. Peak values of normal and shear stress of the parametric study (M1 ... mode 1, M2 ... mode 2, M3 ... mode 3). The peak stresses form distinctive groups for the different modes of loading. While there is not very much difference in peak normal stress, the difference in peak shear stress between the groups is large: Peak shear stress is low for mode 1 load (blue), considerably higher for mode 2 load (black), and yet considerably higher for mode 3 load (red). The figure is best viewed in color.

391

392

393

In contrast, the core regions for shear stress were found to be in very different
 394 ranges (see Fig. 9). Mode 1 shows shear stresses around 2 to 5 MPa, Mode 2 has the core
 395 region at around 12 to 15 MPa, and Mode 3 shows values around 49 to 55 MPa. Please
 396 note that the fiber bond testing setups specifically designed to apply shear forces to the
 397 fiber-fiber bond (mode 2 and 3 in Fig. 2, M2 and M3 in Fig. 9) had the same (M3 in Fig.
 398 9) or even higher (M2 in Fig. 9) peak normal stresses, as compared to the mode 1 (M1 in
 399 Fig. 9) configuration. The low variation in peak normal stresses and the high variation in
 400 peak shear stresses indicates that the critical factor for fiber-fiber bond strength in the
 401 experiments is the normal stress. It is not likely that the peak shear stress is the reason for
 402 failure because its core values vary from 2 to 52 MPa. It is more likely that the true limiting
 403 factor in fiber-fiber bond strength is the normal stress, which was found to have core values
 404 between 2 and 7 MPa for all experiments. The present findings thus lead to a new
 405 interpretation of the single fiber-fiber testing experiments described in the literature. The

406 common explanation that shear stress dominates failure in fiber-fiber bonds was not found
407 in the present results; instead the simulation results suggest that in all three types of
408 experiments made, the bonds failed due to peak normal stresses above 2 to 7 MPa.

409 To place that suggestion into the right context, it is important to briefly discuss the
410 common failure criteria applied to various materials. Material failure strongly depends on
411 whether the material microstructure renders it ductile, brittle, or semi-brittle (Bartenev and
412 Zuyev 1968; Collins 1981; Pruitt and Chakravartula 2011). While ductile materials yield
413 before failure, brittle materials will instantly fracture. A semi-brittle system shows a small
414 amount of plastic deformation prior to failure. Metals are commonly considered as ductile
415 (Tresca or von Mises failure criteria, which are based on shear stress), and ceramics as
416 brittle (normal stress failure criterion). The mechanical behavior of polymer structures is
417 known to depend on many variables in a complex manner: chain chemistry, configuration
418 and length, meso structure, and others. The failure characteristics of polymer biomaterials
419 can exhibit both, ductile (shear stress failure) as well semi-brittle (normal stress failure)
420 behavior (Pruitt and Chakravartula 2011). Thus our interpretation that fiber-fiber joints are
421 more likely to be sensitive to normal loading than shear loading can be aligned with known
422 fracture behavior of composite biomaterials from the literature.

423 The ideas presented in this paper have the potential to shift the understanding of
424 how the fiber-fiber bonds in paper are failing. The fibers and the fiber-fiber bonds in paper
425 under tensile load are subjected to shear stress because they are aligned predominantly in
426 the paper plane. That has intuitively led to the idea that the shear stresses are responsible
427 for the paper failure. Also, the most common theory on paper tensile strength, the equation
428 of Page (1969b), employs shear stress as the key mechanism for fiber-fiber bond strength.
429 As a consequence, shear load is usually regarded to be the tensile failure mechanism in
430 paper (Page 2002). The present results, however, suggest that normal stress failure may be
431 predominant in fiber-fiber bonds, which is a new perspective on the mechanical failure of
432 paper under tensile load. In recent work (Magnusson and Östlund 2013; Magnusson *et al.*
433 2013a), it is shown that normal stresses are of considerable magnitude and present in all
434 three different modes of loading in fiber-fiber bonds. Recently Magnusson concluded that
435 an increase of the strength in the normal direction has the largest effect on the load carrying
436 capacity of fiber fiber bonds (Magnusson 2016), which fits well with our finding that fiber-
437 fiber bond failure initiates due to normal stresses.

438 Future work needs to expand our findings on a single fiber-fiber bond to network
439 structures. The modes of loading and the loading history experienced by bonds in a paper
440 network and the interaction of many pulp fibers may be different from the present model
441 of two crossed fibers. Next to that, further experiments focusing on failure criteria of fiber-
442 fiber joints related to normal stress are certainly required.

443

444

445 CONCLUSIONS

446

- 447 1. Fiber-fiber bonds fail gradually due to the peak stresses at the edges of the bond. A
448 parametric study of the peak stresses in fiber-fiber joints was conducted using FEM
449 models. The models showed characteristic core regions for the three fracture modes
450 investigated experimentally (opening, sliding, and tearing mode). While the normal
451 stresses are almost the same in all three cases, the shear stresses are significantly
452 different.

453 2. Therefore, it was concluded that fiber-fiber joints are more likely to be sensitive to
454 normal loading than shear loading. Hence, it is proposed that a failure criterion for
455 fiber-fiber joints should be related to normal stress. This result brings a new perspective
456 to the theory of fiber-fiber bond failure in paper which, in literature usually is attributed
457 to shear failure.

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460 REFERENCES CITED

461

- 462 ABAQUS FEA (2012). ABAQUS/CAE User's Manual. Dassault Systèmes.
- 463 Bartenev, G. M., and Zuyev, Y. S. (1968). "Strength and failure of visco-elastic
464 materials," Pergamon Press Ltd., Oxford.
- 465 Bodig, J., and Benjamin, A. J. (1993). "Mechanics of wood and wood composites,"
466 Krieger Publishing Company, Florida.
- 467 Brinson, H. F., and Brinson, L. C. (2008). *Polymer Engineering Science and
468 Viscoelasticity: An Introduction*, Springer, New York.
- 469 Burgert, I., Frühmann, K., Keckes, J., Fratzl, P., and Stanzl-Tschegg, S. E. (2003).
470 "Microtensile testing of wood fibers combined with video extensometry for efficient
471 strain detection," *Holzforschung* 57, 661-664. DOI: 10.1515/HF.2003.099
- 472 Button, A. F. (1979). "Fiber-fiber bond strength: A study of a linear elastic model
473 structure," Ph.D thesis, IPST, Georgia Institute of Technology.
- 474 Collins, J. A. (1981). *Failure of Materials in Mechanical Design: Analysis, Prediction,
475 Prevention*, John Wiley & Sons, Inc., New York.
- 476 Da Silva, L. F. M., and Campilho, R. D. S. G. (2012). "Advances in numerical modeling
477 of adhesive joints," SpringerBriefs in Applied Sciences and Technology. Springer,
478 Berlin.
- 479 DeMaio, A., Lowe, R., Patterson, T., and Ragauskas, A. (2006). "Direct observations of
480 bonding influence on the tensile creep behavior of paper," *Nordic Pulp & Paper
481 Research Journal* 21, 297-302. DOI: 10.3183/NPPRJ-2006-21-03-p297-302
- 482 Fischer, W., Hirn, U., Bauer, W., and Schennach R. (2012). "Testing of individual fiber-
483 fiber joints under biaxial load and simultaneous analysis of deformation," *Nordic
484 Pulp & Paper Research Journal* 27, 237-244. DOI: 10.3183/NPPRJ-2012-27-02-pp.
485 237-244
- 486 Gasser, C. (2011). "An irreversible constitutive model for fibrous soft biological tissue: A
487 3-d microfiber approach with demonstrative application to abdominal aortic
488 aneurysms," *Acta Biomaterialia* 7(6), 2457-2466. DOI: 10.1016/j.actbio.2011.02.015
- 489 Groom, L. H., Shaler, S. M., and Mott, L. (1995). "Characterizing micro and
490 macromechanical properties of single wood fibres," in: International Paper Physics
491 Conference, Niagara-on-the-Lake, pp. 11-14
- 492 Grote, K. H., and Feldhusen, J. (2011). "Dubbel," Springer Verlag, Berlin, 23rd edition.
- 493 Hirn, U., Schennach, R., Ganser, C., Magnusson, M., Teichert, C., and Östlund, S.
494 (2013). "The area in molecular contact in fiber-fiber bonds," in: Trans. of the 15th
495 Fundamental Research Symposium, Cambridge, pp. 201-226.
- 496 Hirn, U., Schennach, R. (2015). "Comprehensive analysis of individual pulp fiber bonds
497 quantifies the mechanisms of fiber bonding in paper," *Scientific Reports* 5 10503 (9
498 pp.) DOI: 10.1038/srep10503

- 499 Kappel, L., Hirn, U., Bauer, W., and Schennach, R. (2009). "A novel method for the
500 determination of bonded area of individual fiber-fiber bonds," *Nordic Pulp & Paper*
501 *Research Journal* 24, 199-244. DOI: 10.3183/NPPRJ-2009-24-02-pp. 199-205
- 502 Magnusson, M. S., and Östlund, S. (2013). "Numerical evaluation of interfibre joint
503 strength measurements in terms of three-dimensional resultant forces and moments,"
504 *Cellulose* 20, 1691-1710. DOI: 10.1007/s10570-013-9939-x
- 505 Magnusson, M. S., Fischer, J. W., Östlund, S., and Hirn, U. (2013a). "Interfibre joint
506 strength under peeling, shearing and tearing types of loading," in: *Trans. of the 15th*
507 *Fundamental Research Symposium*, Cambridge, pp. 103-124.
- 508 Magnusson, M. S., Zhang, X., and Östlund, S. (2013b). "Experimental evaluation of the
509 interfibre joint strength of papermaking fibres in terms of manufacturing parameters
510 and in two different loading directions," *Experimental Mechanics* 53, 1621-1634.
511 DOI: 10.1007/s11340-013-9757-y
- 512 Magnusson, M. S. (2016) "Investigation of interfibre joint failure and how to tailor their
513 properties for paper strength," *Nordic Pulp & Paper Research Journal* 31(1), 109-
514 122. DOI: 10.3183/NPPRJ-2016-31-01-p109-122
- 515 Neagu, R. C., Gamstedt, E. K., and Lindström, M. (2004). "Influence of wood-fibre
516 hygroexpansion on the dimensional instability of fibre mats and composites,"
517 *Composites Part A* 36, 772-788. DOI: 10.1016/j.compositesa.2004.10.023
- 518 Nordman, L., Gustafsson, C., and Gustafsson, G. (1952). "On the strength of bondings in
519 paper," *Paperi ja Puu* 3, 47.
- 520 Page, D. H. (1960). "Fibre-to-fibre bonds part 1 - A method for their direct observation,"
521 *Paper Technology* 1, 407-411.
- 522 Page, D. H. (1969a). "A method for determining the fibrillar angle in wood tracheids,"
523 *Journal of Microscopy* 90, 137-143. DOI: 10.1111/j.1365-2818.1969.tb00701.x
- 524 Page, D. H. (1969b). "A theory for the tensile strength of paper," *Tappi Journal* 52(4),
525 674-681.
- 526 Page, D. H. (2002). "The meaning of Nordman bond strength," *Nordic Pulp & Paper*
527 *Research Journal* 17(1), 39-44. DOI: 10.3183/NPPRJ-2002-17-01-p039-044
- 528 Page, D. H., Tydeman, P. A., and Hunt, M. (1962). "A study of fibre-to-fibre bonding by
529 direct observation," in: *Fund. Res. Symp 1961, The Formation and Structure of*
530 *Paper*, Vol. 1, pp. 171-194.
- 531 Page, D. H., El-Hosseiny, F., Winkler, F., and Lancaster, A. P. S. (1977). "Elastic
532 modulus of single wood pulp fibres," *Tappi Journal* 60, 114-117.
- 533 Persson, B. N. J., Ganser, C., Schmied, F., Teichert, C., Schennach, R., Gilli, E., and
534 Hirn, U. (2013). "Adhesion of cellulose fibers in paper," *J. Phys.: Condens. Matter*
535 25 045002 (11 pp.). DOI: 10.1088/0953-8984/25/4/045002
- 536 Pruitt, L. A., and Chakravartula, A. M. (2011). *Mechanics of Biomaterials: Fundamental*
537 *Principles for Implant Design*, Cambridge University Press, Cambridge, UK.
- 538 Saketi, P., and Kallio, P. (2011). "Microrobotic platform for making, manipulating and
539 breaking individual paper fiber bonds," in: *IEEE International Symposium on*
540 *Assembly and Manufacturing (ISAM)*, pp. 1-6.
- 541 Saketi, P., von Essen, M., Mikczinski, M., Heinemann, S., Fatikow, S., and Kallio, P.
542 (2012). "A flexible microrobotic platform for handling microscale specimens of
543 fibrous materials for microscopic studies," *Journal of Microscopy* 248, 163-171.
544 DOI: 10.1111/j.1365-2818.2012.03660.x
- 545 Schmied, F., Teichert, C., Kappel, L., Hirn, U., and Schennach, R. (2012). "Joint strength
546 measurements of individual fiber-fiber bonds an atomic force microscopy based

- 547 method,” *Review of Scientific Instruments* 83, 073902-1-073902-8. DOI:
548 10.1063/1.4731010.
- 549 Schmied, F., Teichert, C., Kappel, L., Hirn, U., Bauer, W., and Schennach, R. (2013).
550 “What holds paper together: Nanometre scale exploration of bonding between paper
551 fibres,” *Scientific Reports* 3, 2432 1-6. DOI: 10.1038/srep02432.
- 552 Schniewind, A. P., Nemeth, L. J., and Brink, D. L. (1964). “Fiber and pulp properties - I.
553 Shear strength of single fiber crossings,” *Tappi Journal* 47, 244-248.
- 554 Seth, R. S. (2006). “The importance of fibre straightness for pulp strength,” *Pulp &*
555 *Paper Canada* 107, 34-42.
- 556 Torgnysdotter, A., Kulachenko, A., Gradin, P., and Wågberg, L. (2007a). “Fiber/fiber
557 crosses: Finite element modeling and comparison with experiment,” *Journal of*
558 *Composite Materials* 41, 1603-1618. DOI: 10.1177/0021998306069873
- 559 Torgnysdotter, A., Kulachenko, A., Gradin, P., and Wågberg, L. (2007b). “The link
560 between the fiber contact zone and the physical properties of paper: A way to control
561 paper properties,” *Journal of Composite Materials* 41, 1619-1633. DOI:
562 10.1177/0021998306069875
- 563 Uesaka, T. (1984). “Determination of fiber-fiber bond properties,” in: *Handbook of*
564 *Physical Testing of Paper*, CRC Press, pages 379-402.
- 565 Wang, J. L., Parnianpour, M., Shirazi-Adl, A., and Engin, A. E. (1997). “Failure criterion
566 of collagen fiber: Viscoelastic behavior simulated by using load control data,”
567 *Theoretical and Applied Fracture Mechanics* 27, 1-12. DOI:10.1016/S0167-
568 8442(97)00002-5
- 569
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571