

3
4 **Spruce Wood Flour for Paper Applications – A**
5 **Handsheet Study**
6
7
8

9
10 **ABSTRACT**
11

This study shows that spruce wood flour can be an alternative cellulosic-based wood additive for papermaking. This study used unbleached wood flour with a particle distribution between 200 µm to 500 µm and bleached and unbleached wood flour with particle size distribution between 70 µm to 150 µm. Wood flour was added at levels of 2%, 4%, 6%, 8% and 15% based on oven dry fiber content for the first part of the study. For the second part of the study, starch at a level of 0.25%, 0.50%, 0.75%, 1.00%, 1.25%, and 1.50% based on OD fiber content is added to the suspension. The basis weight of the handsheet manufactured was 75 g/m². Bulk increased from 2.20 cm³/g to a maximum of 2.80 cm³/g for 15% wood flour addition. Maximum tensile index achieved was 24.75 Nm/g based on a base sheet value of 20.05 Nm/g. Addition of starch has a positive influence on the tensile index, with a maximum value of 41.41 Nm/g at 1% addition. Brightness value of the manufactured handsheets decreased gradually for the unbleached wood flour. Bleached wood flour showed a 1%-point increase above the base sheet brightness of 88.51%. Addition of starch increased the brightness value from 88.51% of the base sheet by up to 4.5%. An opacity increase was achieved for all wood flour additions with the highest opacity value of 95.68% at an addition of 15% wood flour. Addition of starch can decrease the opacity value of up to 1.5% points. Addition of wood flour resulted in a decrease in smoothness by increasing the airflow from the base sheet value of 2564 ml/min by 385 ml/min. at 8% wood flour addition. Adding a line pressure of 1.673 kN/m to simulate calendering resulted in an improved smoothness by reducing the airflow of up to 447 ml/min. Addition of starch showed an overall increase of smoothness by reducing the airflow number by up to 600 ml/min for sheets with and without line pressure.

Comment [HSMD1]: Wrong format. It should contain aims, study design, place and duration of study, methodology, results and conclusion.

12
13 *Keywords: Wood flour, additive, papermaking, handsheets, paper properties*
14
15

16 **1. INTRODUCTION**
17

18 Paper today is produced from renewable hardwood and softwood materials. Recycling of
19 paper products has improved the environmental footprint of the paper industry in the past
20 decades [1]. Despite this, the paper industry is increasing their efforts in making paper more
21 sustainable, biodegradable and eco-efficient. However, ever rising production cost for paper
22 and board products and their application demand new solution of utilizing raw materials for
23 the production process. Tighter environmental regulations demand an increasing use of
24 sustainable chemical and additives. This will result in an increasing use of renewable
25 materials in the future [2].

26 One of these renewable materials that can be added to the papermaking process as additive
27 is Wood Flour (WF). WF is known since the early 1900s [3]. The first WF patent was issued
28 for the production process of phonographic records and other articles. The US. Patent
29 No.1,406.938 was granted to John Cunningham, a resident in Glens Falls in New York State
30 on Feb. 14, 1922 [4]. According to Reineke (1966) [5], WF are wood particles manufactured
31 by grinding selected wood residues. WF can be produced by various grinding and sieving
32 processes of sawdust to sizes between 20 μ m to 500 μ m with a size ratio of 1:1 [6].
33 Karinkanta et al. describes that the manufacturing process today can consist of a thermal,
34 chemical and enzymatic pretreatment before wet milling, dry milling and sieving techniques
35 are applied [7]. Commercial applications for WF today are mainly in the area of Wood Plastic
36 Composites (WPC) and moulding technology applications for articles such as furniture parts,
37 dishes and toys (Hogan et al. 2011) [8].
38 Recently WF with a size of 200 μ m o 450 μ m has been investigated in a handsheet
39 laboratory study by Dongmei et al. [9]. He showed that bulk can be improved, and
40 mechanical pulp be replaced. Lee et al. [10] showed that wood powder added to duplex
41 board increase bulk of the produced board paper. Sung et al. [11] showed that powder
42 produced from conifer leaves can be an alternative organic filler source to wood flour in
43 paperboard applications.
44 Park et al. [12] investigated flour from wood and ground agricultural byproducts for a
45 paperboard application, showing that bulk and drying can be improved, but paper strength is
46 decreasing. However, WF has not been the focus in recent investigations as an alternative
47 cellulosic-based wood additive for papermaking. This handsheet study compares three
48 commercial varieties of spruce WF at an addition of 2%, 4%, 6%, 8% and 15% to a 75 g/m²
49 paper product.

51 52 **2. MATERIAL AND METHODS**

53
54 This section describes the materials, standardized TAPPI test methods, and procedures,
55 used for this study. Repeatability of the results stayed in between the allowable margins of
56 the TAPPI testing standards.

57 58 **2.1 TAPPI Methods**

59 Pulp refining was done according to T 200 sp-06 "Laboratory beating of pulp (Valley beater
60 method) [13]. Handsheets for physical testing were prepared in accordance with T 205 sp-
61 06, "Forming handsheets for physical tests of pulp" [14]. Physical testing of handsheets was
62 performed in accordance to T 220 sp-06, "Physical testing of pulp handsheets" [15], the
63 freeness of pulp was measured as Canadian Standard Freeness (CSF) according to T 227
64 om-09 "Freeness of pulp (Canadian standard method)" [16]. "Forming handsheets for
65 physical tests of pulp". Conditioning of the paper samples was done according to T 402 sp-
66 08, "Standard conditioning and testing atmospheres for paper, board, pulp handsheets, and
67 related products" [17]. Tensile strength was measured in accordance with T404 cm-92,
68 "Tensile breaking strength and elongation of paper and paperboard" [18]. Basis weight was
69 measured with T 410 om-08. "Grammage of Paper and Paperboard (weight per unit area)"
70 [19]. The paper thickness was measured by T 411 om-10 "Thickness (caliper) of paper,
71 paperboard, and combined board" [20]. Moisture content of pulp was determined by T412
72 om-06 "Moisture in pulp, paper and paperboard" [21]. Opacity of paper handsheets was
73 performed according to T 425 om-06, "Opacity of paper (15/d geometry, illuminant A/2", 89%
74 reflectance backing and paper backing) [22]. Brightness of pulp was measured according to
75 T 452 om-08, "Brightness of pulp, paper and paperboard (directional reflectance at 457 nm)"
76 [23]. Tensile strength was performed following T494 om-06, "Tensile properties of paper and
77 paperboard (using constant rate of elongation apparatus)" [24]. Smoothness/Roughness of

78 the manufactured handsheets was tested according to T 538 om-08, "Roughness of paper
79 and paperboard (Sheffield Method)" [25].

80
81

82 2.1 Materials

83

84 For this study 75 g/m² handsheets are produced from 80% Elemental Corine Free (ECF)
85 Eucalyptus bleached Kraft pulp, and 20% Northern Bleached Softwood Kraft (NBSK) pulp.
86 Prior to handsheet forming the pulp is refined to a Canadian Standard Freeness (CSF) level
87 of 360 ml following T 200 sp-06 method [16]. WF was added based on Oven Dry (OD) fiber
88 content prior to handsheet forming following T 220 sp-06 method [18]. Spruce WF was
89 obtained from J. Rettenmaier & Söhne, Rosenberg, Germany. WF1 and WF2 were
90 unbleached with a particle size distribution of 200µm to 500µm and 70µm to 150µm
91 respectively. WF3 was bleached with a particle size distribution of 70 µm to 150 µm.
92 Starch used in this study was cationic starch cooked at a 3% solution at 90°C for 20 minutes
93 prior to handsheet making, cooled down to 30°C and added to the pulp WF suspension prior
94 to **handsheet** forming.

95

96

97 3. RESULTS AND DISCUSSION

98

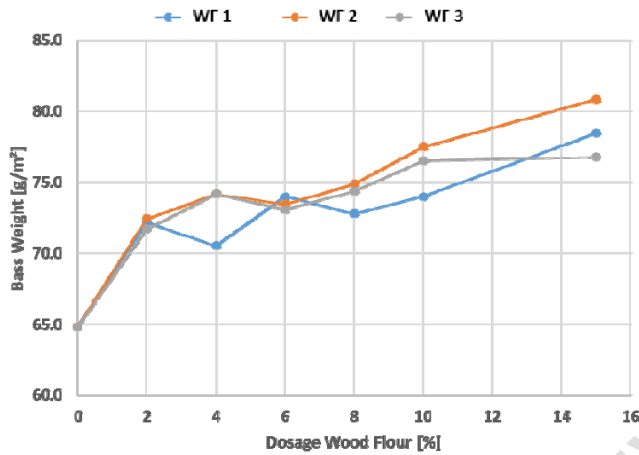
99 All handsheets were made and tested according to TAPPI standards. In the first part of the
100 study WF1, WF2, and WF3 was added at levels of 2%, 4%, 6%, 8% and 15% based on OD
101 pulp. In the second part, handsheets were prepared with the addition of starch at a level of
102 0.25%, 0.50%, 0.75%, 1.00%, 1.25%, and 1.50% based on OD fiber content. Handsheets
103 with and without starch for the smoothness measurement were exposed to a **line** pressure of
104 1.673 kN/m to simulate calendering.

105 Fig.1 shows that the basis weight of the base sheet was 64.5 g/m². Manufactured
106 handsheets with WF1 had a basis weight range of 72.20 g/m² to 76.50 g/m², WF2 resulted in
107 a basis weight range of 72.40 g/m² to 80.30g/m², and WF3 in a basis weight range of 71.70
108 g/m² to 76.90 g/m². The basis weight increase for all WF follows the same pattern except for
109 WF1 at a dosage of 4%, 8%, and 10% were a 3.6 g/m², 2.1 g/m², 3.5 g/m² lower basis
110 weight was achieved respectively compared to WF 2 which had the highest basis weight at
111 all WF dosage levels. WF3 basis weight levels are very comparable to WF 2 except for the
112 15% dosage were a 1.7 g/m² lower basis weight was the result for WF 1 and 4.1 g/m² for
113 WF3.

114

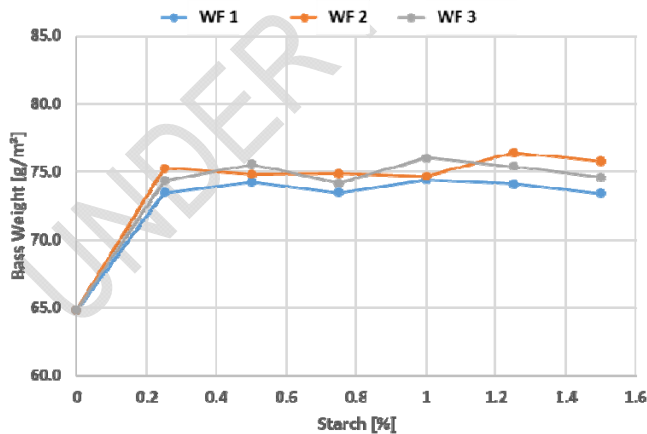
Comment [HSMD2]: grammatical error.

Were added at levels.....



115
116 **Figure 1: Basis weight**
117

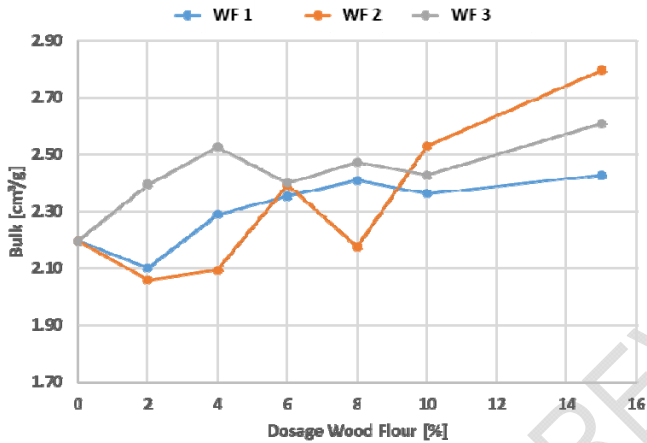
118 The graph in Fig.2 shows the basis weight achieved for WF1 to WF3 with the addition of
119 starch at 0.25%, 0.50%, 0.75%, 1.00%, 1.25%, and 1.50% based on OD fiber content. The
120 percentage of the WF addition was chosen based on Fig.1. WF1 addition was 2%. The
121 addition of WF2 unbleached and WF3 bleached with the same particle size distribution was
122 4% and 8% respectively in order to compare unbleached and bleached WF at the same
123 basis weight for the starch addition. Fig. 2 shows, that starch serves as a good retention aid,
124 bonding the fine fibers and WF into the produced handsheet. As a result, the basis weight of
125 the handsheet increases from the base sheet of 65.00 g/m² of about 10.00 g/m² at a starch
126 addition of 0.25%. For a starch addition of 0.50%, 0.75%, 1.00%, 1.25%, and 1.50% the
127 basis weight stays constant at around 75.00 g/m².
128



129
130 **Figure 2: Basis weight with starch**
131

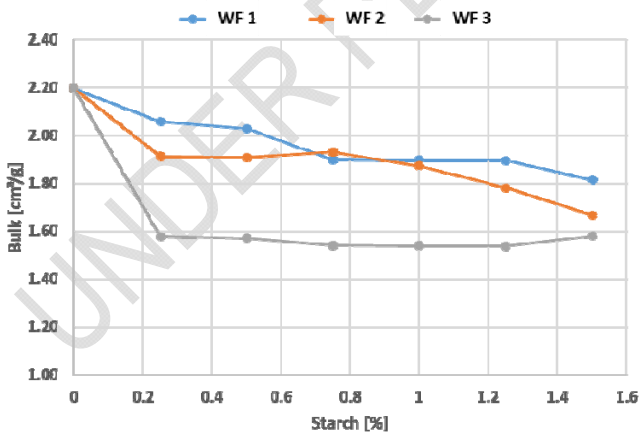
132 Fig. 3 shows that addition of WF increases the bulk from 2.20 cm³/g of the base sheet to a
133 maximum of 2.43 cm³/g, 2.80 cm³/g, and 2.61 cm³/g for the 15% WF addition of the
134 manufactured handsheets for WF1, WF 2, and WF3 respectively. For WF1 and WF2 a bulk

135 reduction resulted for the 2% addition to 2.10 cm³/g and 2.06 cm³/g respectively. WF2
 136 showed in addition lower bulk value for the 4% and 8% addition at 2.09 cm³/g and 2.17
 137 cm³/g respectively.
 138



139
 140 **Figure 3: Bulk without starch**
 141

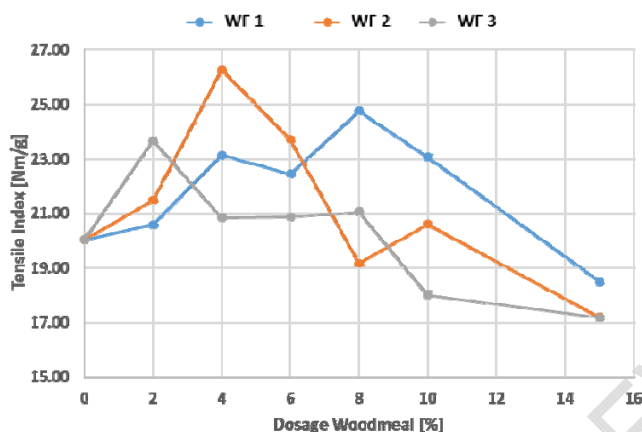
142 Addition of starch reduces the bulk, as shown in Fig.4 from 2.20 cm³/g of the base sheet to a
 143 minimum of 1.61 cm³/g, 1.67 cm³/g, and 1.58 cm³/g for the 1.5% starch addition
 144 of the manufactured handsheets for WF1, WF 2, and WF3 respectively. Bulk reduction for WF3
 145 was identical for all starch additions. WF1 and WF2 had the lowest reduction at 0.25%
 146 starch addition with 2.06 cm³/g and 1.91 cm³/g respectively.
 147



148
 149 **Figure 4: Bulk with starch**
 150

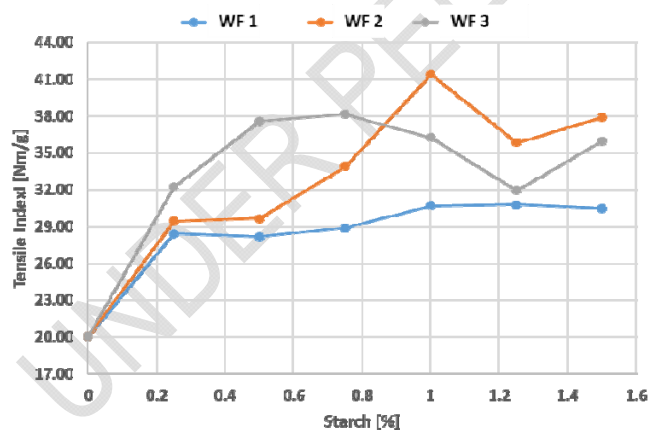
151 Fig. 5 shows that WF3 has an increase in tensile index only for an addition of 2% from the
 152 base value of 20.05 Nm/g to a value of 23.67 Nm/g. WF2 had its maximum tensile index at
 153 an addition of 4% with a value of 23.13 Nm/g. At an addition of 8%, 10%, and 15% the
 154 tensile index was lower at 19.17 Nm/g, 20.60 Nm/g, and 17.20 Nm/g respectively. WF1 had

155 its maximum tensile index at an addition of 8% with a value of 24.75 Nm/g. At an addition of
 156 15% the tensile index was lower at 18.50 Nm/g.
 157



158
 159 **Figure 5: Tensile index without starch**
 160

161 Addition of starch and WF increases the tensile index for all WF1 to WF3 as shown in Fig. 6.
 162 Above the base sheet value of 20.05 Nm/g. For WF1 has its peak at a starch addition of 1%
 163 with a tensile index value of 30.70 Nm/g. WF2 and WF3 have their maximum tensile index at
 164 1% with 41.41 Nm/g and 0.75% with a value of 36.26 Nm/g respectively.
 165



166
 167 **Figure 6: Tensile index with starch**
 168

169
 170 Fig. 7 and Fig. 8 show the brightness value of the manufactured handsheets for different
 171 additions of WF1, WF2, and WF3. A gradually decreasing brightness value with increasing
 172 WF content can be observed for WF1 and WF2, with the lowest brightness of 83.27% and
 173 85.92% respectively, based on the base sheet brightness of 88.51%. WF3 showed a up
 174 to 1% point brightness gain compared to the base sheet brightness of 88.51.

Comment [HSMD3]: Is it correct??
 It could be « an up to»....
 Manuscript should be written in concise English.

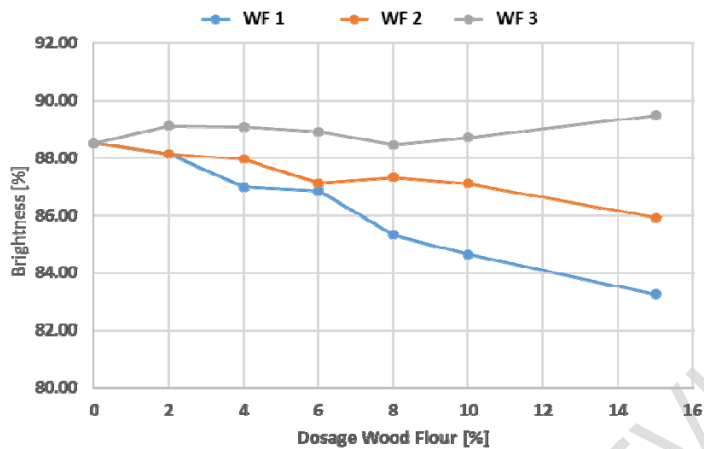


Figure 7: Brightness

Addition of starch increased the brightness value from 88.51% of the base sheet by up to 4.5% for the bleached WF3 and up to 4% for WF2. WF1 resulted in a up to 2.5- points brightness increase. For all WF, a starch addition of 0.25% resulted in the highest brightness increase. For starch additions of 0.55, 0.75%, 1.00%, 1.25%, and 1.50%, except for WF1 and WF2 which had a brightness increase of 3.5%-points at a starch addition of 1.5%.

Comment [HSMD4]: Is it correct?
It should be an up to

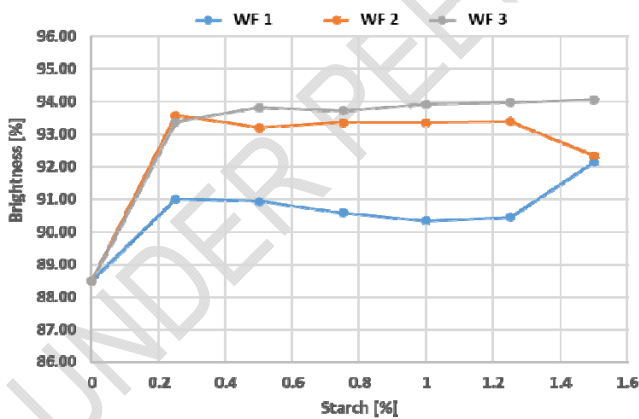
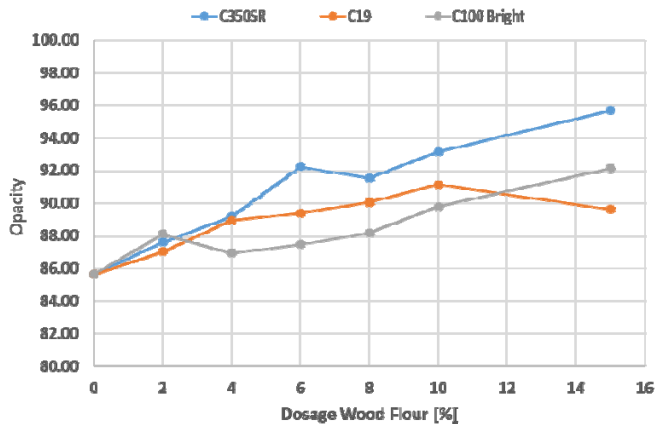


Figure 8: Brightness with starch

Fig. 9 and Fig. 10 show the opacity value of the manufactured handsheets for different additions of WF1, WF2, and WF3. The opacity value of the base handsheet was 85.64%. A gradually increasing opacity value with increasing WF content can be observed for WF1, WF2, and WF3, with the highest opacity of 95.68% and 92.14% for WF1 and WF3 respectively at the addition of 15%. WF 2 had its highest opacity value at an addition of 10% with an opacity value of 91.13%.

175
176
177
178
179
180
181
182
183

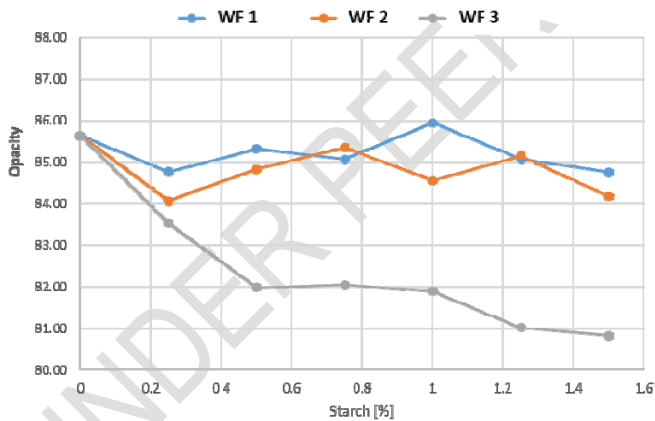
184
185
186
187
188
189
190
191
192
193



194
195
196
197
198
199
200
201

Figure 9: Opacity

Addition of starch decreased the opacity value from 85.64% of the base sheet by up to 0.9% points for WF1, up to 1.5% points for WF2, and up to 3.2% points for WF3. At a starch addition of 1% WF1 showed a 0.3%-point opacity increase based on the base value of 85.64%.



202
203
204
205
206
207
208
209

Figure 10: Opacity with starch

Fig. 11 shows the smoothness of handsheets for WF1, WF2, and WF3. The initial value for smoothness was 2564 ml/min. for the base sheet. Addition of WF1, WF2, and WF3 decrease the smoothness of the paper due to the higher airflow value. WF1 increase up to 385 ml/min. at 8% WF addition, WF2 and WF3 showed an increase of 314 ml/min. and 301ml/min. at 15% WF addition respectively.

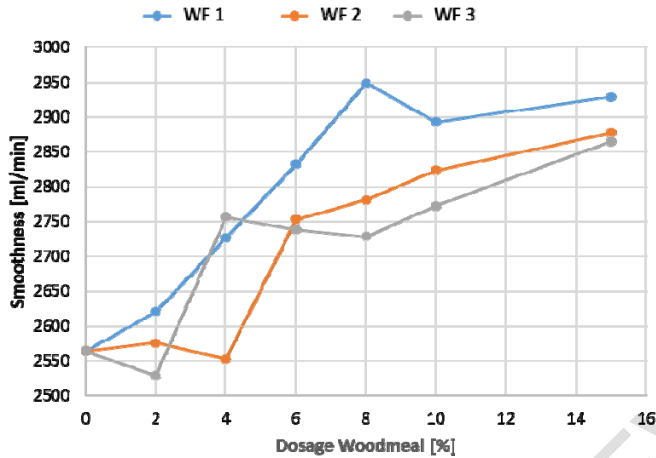


Figure 11: Smoothness without line pressure

Fig. 12 shows smoothness values with a line pressure of 1.673 kN/m applied to simulate calendaring. The line pressure reduces the airflow and improves smoothness of the manufactured handsheets containing WF. Applying the line pressure reduces airflow by 362 ml/min. to 2202 ml/min. for the base sheet, WF1 had a reduction of 31 ml/min. to 118 ml/min., WF2 a reduction between 77 ml/min. to 447 ml/min, WF3 a reduction of 104 ml/min. to 335 ml/min.

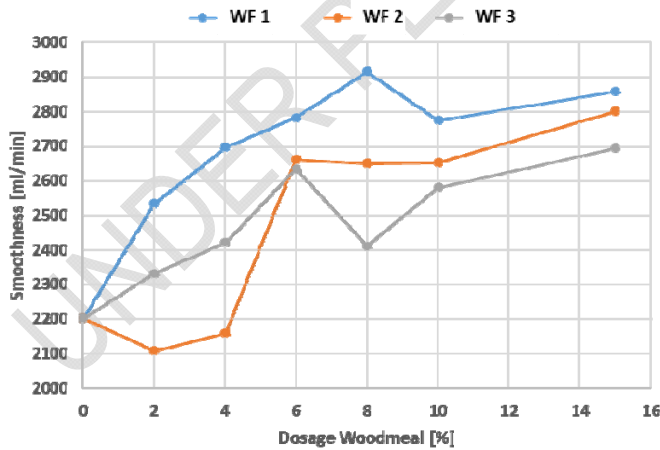
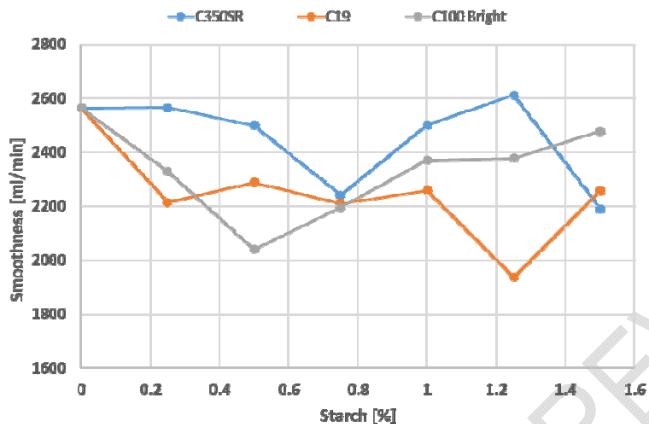


Figure 12: Smoothness with 1.673 kN/m line pressure

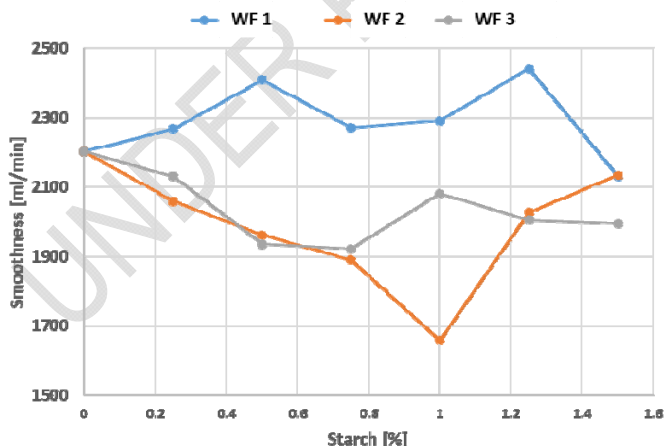
Graphs of Fig. 13 and Fig. 14 show the smoothness value if starch is applied for handsheets containing WF1, WF2, and WF3. The initial value for smoothness was 2564 ml/min. for the base sheet. Addition of starch showed an overall increase of smoothness by reducing the airflow number. WF1 had a maximum decrease below the air flow number of the base sheet

228 of 298 ml/min., WF2 showed a decrease of 600 ml/min., and WF3 showed a 484 ml/min. air
 229 flow decrease.
 230



231 **Figure 13: Smoothness with starch and without line pressure**

232 Fig. 14 shows smoothness values with a line pressure of 1.673 kN/m to the starch containing
 233 handsheets to simulate calendaring. The line pressure reduces the airflow and improves
 234 smoothness of the manufactured handsheets containing WF. Applying the line pressure
 235 reduces airflow by 362 ml/min. to 2202 ml/min. for the base sheet. WF1 had a reduction of
 236 up to 298 ml/min., WF2 a reduction of up to 600 ml/min., and WF3 a reduction of up to 484
 237 ml/min. of air flow.
 238
 239
 240



241 **Figure 14: Smoothness with starch and line pressure of 1.673 kN/m**

242
 243
 244

245 4. CONCLUSION

246
247
248
249
250
251
252
253
254
255
256
257
258
259
260
261
262
263
264
265
266
267
268
269
270
271
272
273
274
275
276
277
278
279
280
281
282
283
284
285
286
287
288
289
290
291
292
293
294
295

This study shows that WF can be considered an alternative cellulosic-based wood additive for papermaking. This handsheet study showed that spruce WF with a particle distribution between 200 µm to 500 µm and bleached and unbleached WF with a particle distribution of 70 µm to 150 µm respectively could have benefits for paper production. WF added at levels of 2%, 4%, 6%, 8% and 15% to a 75 g/m² handsheet increases bulk from 2.20 cm³/g of the base sheet to a maximum of 2.80 cm³/g for the 15% WF addition. Increase of tensile index can be achieved at up to 8% WF addition but is dependent on the WF type used. Maximum tensile index achieved was 24.75 Nm/g based on a base sheet value of 20.05 Nm/g. Addition of starch has a positive influence on the tensile index, with a maximum value of 41.41 Nm/g at 1% addition. Bulk values decreased by the addition of starch at all levels. Brightness value of the manufactured handsheets decreased gradually for the unbleached WF. Bleached WF showed a 1%-point increase above the base sheet brightness of 88.51%. Addition of starch increased the brightness value from 88.51% of the base sheet by up to 4.5%. An opacity increase was achieved for all WF addition with the highest opacity value of 95.68% at an addition of 15% WF. Addition of starch can decrease the opacity value of up to 1.5% points. Addition of WF resulted in a decrease in smoothness by increasing the airflow from the base sheet value of 2564 ml/min by 385 ml/min. at 8% WF addition. Adding a line pressure of 1.673 kN/m to simulate calendering resulted in an improved smoothness by reducing the airflow of up to 447 ml/min. by WF addition. Addition of starch showed an overall increase of smoothness by reducing the airflow by up to 600 ml/min. based on the WF used. By applying a line pressure of 1.673 kN/m to the starch, containing handsheets smoothness can be improved by an additional 600 ml/min. in airflow reduction.

275 7. REFERENCES

1. Doelle K, Amaya JJ, Application of calcium carbonate for uncoated digital printing paper from 100% eucalyptus pulp. TAPPI JOURNAL, Jan. 2012;11(1):41-49.
2. Lyon SW, Quesada-Pineda HJ, Crawford SD. Reducing electrical consumption in the forest products industry using lean thinking. *BioRes.* 2014;9(1):1373-1386.
3. Clemons CM, Wood flour. In: Xanthos M (ed) Functional fillers for plastics, 2nd ed. Wiley-VCH, Weinheim, 2010:269–290
4. Cunningham JJ.: Method for Producing Flour, US Patent No. 1,406,938, patented February 14, 1922.
5. Reineke LH, Wood Flower, U.S. Department of Agriculture, Forest Products Laboratory, U.S. Forest Service Research Note FPL-0113., 1966.
6. Korte K, Ofte S, Hansmann H. Compression, relaxation and swelling behavior of solid wood, wood powder and wood-plastic composites (WPC), *Holztechnologie* 2016;57(6):5-11.
7. Karinkanta A, Ämmälä MI, Jouko N, Fine grinding of wood – Overview from wood breakage to applications, *Biomass and Bioenergy*, 2018;113:31-44.
8. Hogan US, Akpan GA, Essien OA, Wood Flour Moulding Technology: Implications for Technical Education in Nigeria, *African Research review*, 2011;5(2): 233-242.
9. Dongmei Y, Chuanshan Z, Chaojun W, Daiqi W, Wood Powder Used in Paper Making to Improve Bulkness, *Advanced Materials Research*, 2015;550-553: 3352-3355.

Comment [HSMD5]: the conclusion can be short.

Comment [HSMD6]: Wrong format.

Reference to a journal:
For Published paper:

1. Hilly M, Adams ML, Nelson SC. A study of digit fusion in the mouse embryo. *Clin Exp Allergy.* 2002;32(4):489-98.

Comment [HSMD7]: Journal name should be written either in full name or abbreviated name, but not a mixture of both.

Comment [HSMD8]: Journal name should be written either in full name or abbreviated name, but not a mixture of both.

- 296 10. Lee JY, Kim CH, Seo DJ, Lim GB, Kim SY, Park JH, Kim EH, Fundamental study on
297 developing wood powder as an additive of paperboard, TAPPI Journal, 2013;13(11):17-
298 21.
- 299 11. Sung JY, Kim DS, Lee JY, Seo YB, Im CK, Gwon WO, Kim JD, Application of Conifer
300 Leave Powder to Papermaking Process as an Organic filler, Journal of Korea TAPPI,
301 2014;46(4):62-68.
- 302 12. Park, JH, Lee, JY, Kim, CH, Kim EH, Effects of Lignocellulosic Bulk agents Made from
303 Agricultural Byproducts on Physical Properties and Drying energy Consumption of
304 Duplex Board, BioResources, 2015;10(4):7889-7897.
- 305 13. TAPPI T 200 sp-06. Laboratory beating of pulp (Valley beater method).
- 306 14. TAPPI T 205 sp-12. Forming handsheets for physical tests of pulp.
- 307 15. TAPPI T 211 om-02. Ash in wood, pulp, paper and paperboard: combustion at 525°C.
- 308 16. TAPPI T220 sp10. Physical testing of pulp handsheets.
- 309 17. TAPPI T227 om-09. Freeness of pulp (Canadian standard method).
- 310 18. TAPPI T 402 sp-13. Standard conditioning and testing atmospheres for paper, board,
311 pulp handsheets.
- 312 19. TAPPI T404 cm-92 Tensile breaking strength and elongation of paper and paperboard
- 313 20. TAPPI T 410 om-08. Grammage of Paper and Paperboard (weight per unit area).
- 314 21. TAPPI T 411 om-10. Thickness (caliper) of paper, paperboard, and combined board.
- 315 22. TAPPI T412 om-06. Moisture in pulp, paper and paperboard.
- 316 23. TAPPI T414 om-12. Internal tearing resistance of paper (Elmendorf-type method).
- 317 24. TAPPI T425 om-06. Opacity of paper (15/d geometry, illuminant A/2°, 89% reflectance
318 backing and paper backing).
- 319 25. TAPPI T 452 om-08. Brightness of pulp, paper and paperboard (directional reflectance
320 at 457 nm).
- 321 26. TAPPI T494 om-06. Tensile properties of paper and paperboard.
- 322 27. TAPPI T 538 om-08. Roughness of Paper and Paperboard (Sheffield method)
- 323