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Tensile Strength of Handsheets from Recovered Fibers Treated with *N*-Methylol Melamine and 1,3-Dimethylol-4,5-dihydroxyethyleneurea

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ABSTRACT: The main objective of this study was to assess the effect of two amino resins, *N*-methylol melamine (NMM) and 1,3-dimethylol-4,5-dihydroxyethyleneurea (DMDHEU), on the intrafiber and interfiber strengths and water absorption of two types of waste paper categories, office paper (OP), and old corrugated containers (OCCs). The tensile strength of individual fibers measured at zero span was reduced by increases in the resin concentrations. The dry tensile strengths of the recovered handsheets measured at a finite span were enhanced with increases in the weight percentage gain of the resins. The increasing of the resin concentration also significantly improved the intrabonding of the OP and OCCs in moist measuring conditions. The water absorption of the handsheets considerably decreased at the higher concentration of the thermosetting resins, especially with NMM. The results are promising for the use of NMM- and DMDHEU-treated recovered fibers as an alternative fiber resource for the production of laminated paper and also for the use of DMDHEU as a new *N*-methylol compound for laminated paper. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2015**, *132*, 41290.

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INTRODUCTION

The impregnation of paper sheets with an aminoplastic thermosetting resin has been widely used for the surface protection and decoration of medium-density fiberboards and particleboards for furniture[1–3] and for laminate flooring.[4,5]

The manufacturers of these sheets face challenges in the retention of the high performance of the products and the reduction of the production costs to make appropriate cover papers for overlaying wood-based panels (WBPs). Therefore, it is inevitable that producers of impregnated papers will need to minimize their production costs and increase productivity by substituting costly raw materials and increasing the material output.[6] Moreover, the limitation in the availability of natural sources and the search for an inexpensive solutions has forced WBP producers and the paper industry to shift toward alternative raw materials such as nonwood and recycled fibers.[7]

Recycled fiber sources are highly heterogeneous and differ greatly in their characteristics and morphological properties. Fiber analysis is one suitable technique that the paper industry can use to address the variability of the recycled raw materials used in paper manufacturing.[8,9] Moreover, the recycling process affects the morphology and properties of the fibers[10–14] and, thus, influences paper performance. Among the various mechanical, chemical, and enzymatic treatments,[15–20] chemical modification has been proved to be suitable for improving the performance of fibers and paper.

Laminated paper is normally produced from kraft pulp and is saturated by resin because of capillary action. Melamine formaldehyde resin, which is cured to form a hard composite with the fiber structure of the paper, is mostly used for laminated paper.[21] Also, melamine formaldehyde is traditionally recommended as a wet-strength agent in the paper industry but at very low concentrations.[22,23]

The application of crosslinking agents can enhance fiber bonding and, hence, improve the mechanical properties of paper.[24–26] 1,3-Dimethylol-4,5-dihydroxyethyleneurea (DMDHEU) is commonly used as a wrinkle-resistance additive for cotton fabrics in the textile industry[27–32] and has also found applications in wood modification.[33,34] The modification of wood products such as veneer and paper with *N*-methylol melamine (NMM)

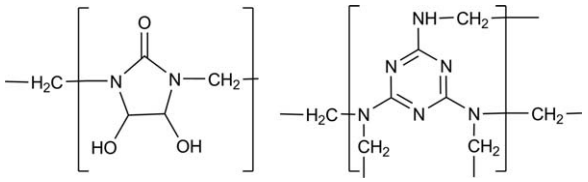

Figure 1. Schematics of the DMDHEU (left) and NMM (right) building blocks.

and DMDHEU reduces the flexibility of the cell wall and increases the embrittlement.[35,36]

The two main objectives of this study were (1) to investigate the effect of *N*-methylol resins on the interfiber and intrafiber tensile strengths and the water repellency of fibers recovered from waste papers [office paper (OP) and old corrugated containers (OCCs)], which may serve as alternative resources for the production of laminated paper, and (2) to evaluate DMDHEU as a new *N*-methylol compound for impregnating paper.

EXPERIMENTAL

Fibers and Chemicals

Two types of waste papers, OCCs and OP, from the monthly usage of the University of Göttingen (Göttingen, Germany) were used throughout this study. The OCC waste category was used with the mixture of liner and fluting grades. Both types of waste papers (OCCs and OP) were manually cut into small pieces, mixed with tap water, and then dispersed by a laboratory disintegrator (Ika Ultra-Turrax T18 Basic, IKA, Staufen, Germany) to produce pulp. Then, to gradually remove the excess water, the wet pulps were placed in an oven at 40°C for 72 h and stored in a climate chamber at 23°C and 50% relative humidity (RH) until handsheet preparation occurred.

A commercial ready-to-use solution of DMDHEU (Fixapret CP Konz) was supplied by BASF SE (Ludwigshafen, Germany), whereas magnesium chloride hexahydrate ($MgCl_2{\cdot}6H_2O$) was used as a catalyst. The NMM (Madurit MW840/75WA, Ineos Melamines GmbH, Frankfurt, Germany) was supplied as an aqueous stock solution with a solid content of approximately 75% and a specific gravity of 1.245–1.260 kg^{-1} at 23°C.

Fiber Analysis

To get information on the fibrous raw materials from the waste paper categories, fiber furnish analysis was performed according to ISO 9184-1.[37] For testing, three unmodified handsheets were randomly selected. The qualitative and quantitative determinations of the fiber components of the waste papers were carried out with the Herzberg staining test method (ISO 9184-3).[38] The stained fibers were examined under a Nikon ECLIPCE E600 light microscope (Nikon, Düsseldorf, Germany) equipped with a digital camera and a crosshair eyepiece. Identification of the pulping processes was based on the colors developed by the Herzberg stain.[9] The fibers were classified into softwood, hardwood, and nonwood categories according to their morphology. The weight percentages of the pulp constituents were calculated after the conversion of the fiber counts through the use of appropriate predetermined weight factors recommended by ISO 9184-1.[37] The total fiber count of each category was multiplied by its respective weight factor to obtain the equivalent weights, and then, their percentages by weight of the total weight were calculated and reported as the nearest whole number.

Preparation of the Handsheets and the Resin Treatments

For handsheet preparation, 2.65 g of dry fibers was dispersed in 1000 mL of tap water for 2 min with an Ultra-Turrax (Ika T18 basic, IKA, Staufen, Germany) stirring device at 22,000 rpm. Handsheets with an average grammage of 80 g/m^2 were prepared with a Rapid Köthen sheet former (Kleinblattbildner KBB-1, Estanit GmbH, Muehlheim an der Ruhr, Germany) according to the German standard DIN 54358-1.[39] The handsheets were dried at 93°C with a vacuum of 0.95 bar for 10 min.

The resin treatments were performed by the immersion of the handsheets in aqueous solutions of 2, 5, 10, 20, and 40% w/w separately for NMM and DMDHEU (on the basis of the stock solution) for 2 min under ambient conditions. Control handsheets were also submerged in demineralized water. Subsequently, the handsheets were dried in an oven at 120°C for 120 min. The treated and untreated handsheets were stored at 23°C and 50% RH until testing.

Weight Percentage Gain (WPG) and Nitrogen Analysis

The WPG was calculated from the oven-dried weight of the handsheet before and after the resin treatments as follows:

$$\mathrm{WPG}=\frac{W_1\text{-}W_0}{W_0}\times 100$$

where W_1 is the oven-dried weight of the treated handsheet and W_0 is the oven-dried weight of the untreated handsheet.

To determine the carbon and nitrogen content, the handsheets were ground in a ball mill, and subsequently, 6 mg of dry powder was analyzed in a LECO CHN 2000-Analyzer (LECO Instrumente GmbH, Mönchengladbach, Germany).

The contents of DMDHEU ($C_4N_2O_3H_6$) and NMM ($N_6C_6H_7$) in the treated handsheets were calculated from the nitrogen content and the molecular weight of the building blocks of DMDHEU and NMM with estimated one and three methylene groups in the resin (Figure 1) as follows:

$$q=\frac{a}{\rho}\times n$$

where q is the resin content in the treated handsheet (%), a is the molecular weight of one building block in the resin (g/mol), ρ is the molecular weight of nitrogen in one building block in the resin (g/mol), and n is the nitrogen content (%).

Zero-Span Tensile Strength (Z-Strength) Assessment

The Z-strength of the handsheets was assessed with a Pulmac paper tester (Pulmac International, Inc., Middlesex, United Kingdom) as described previously.[36,40] The clamping pressure was set to 0.6 MPa, and the force was gradually increased by 70 kPa/s. The dimensions of the samples (paper strips) were 50 × 15 mm^2, and the test procedure was carried out according to Tappi T231.[41] For each treatment, 20 samples were used for both dry and wet conditions.

For the wet measuring condition, a sponge was filled up with water up to approximately 1/2 of the sponge thickness. The

Table I. Weight Proportions of Fiber Components in the Waste Paper Categories

Waste paper category	Weight (%)							
	Chemical pulp				Semichemical and chemimechanical pulp			
	SW	HW	NW	Total	SW	HW	NW	Total
OCC	20	9	1	30	49	21	0	70
OP	33	11	1	44	33	22	1	56

SW, softwood; HW, hardwood; NW, nonwood.

surface of the sponge was moist but not saturated throughout the experiment. Then, the paper strips were laid on the top of the sponge and wetted with a spray bottle. A roller was used to saturate the sample properly. After 5 s, we transferred each strip to the wet sample inserter by laying the inserter on the strip and pressing down slightly. The inserter was then lifted and turned so that the test strip was on top.

Finite-Span Tensile Strength (F-Strength) Assessment

We determined the F-strength with a Zwick Z010 universal testing machine (Zwick, Ulm, Germany) according to BS ISO 1924-3[42] by keeping a 70-mm distance between the clamps at an elongation rate of 100 mm/min. The dimensions of the samples (paper strips) were 100 × 15 mm^2. For each treatment, 20 samples were used in dry and wet conditions and at 90% RH (24 h, conditioned at 23°C). The testing procedure under wet conditions was the same as that used for the wet Z-strength test.

Water Absorption (Cobb Test)

Water absorption was determined with a Cobb sizing tester (Gurley Precision Instruments, New York) according to ISO 535.[43] Test pieces with dimensions of 140 ×140 mm^2 were conditioned at 23°C and 50% RH and weighed. For each treatment, 10 samples were tested. After 20 s of contact, the cylinder was emptied and released. After another 10 s (for a total of 30 s), the wet specimen was placed between two bloating papers, and a 10-kg roller was rolled once in each direction over the sample. The specimen was weighed again, and the water absorption was calculated.

Statistical Analysis

A one-way analysis of variance (ANOVA) of the properties of handsheets was performed with Statistica10 (StatSoft, Inc.). Statistical differences between the values were evaluated by Tukey's honestly significant difference (HSD) as a *post hoc* test at an error probability of $\alpha = 0.05$.

RESULTS AND DISCUSSION

Fiber Composition

Fiber analysis revealed differences in the composition for both the origin (softwood, hardwood, or nonwood) and processing of the fibers (type of pulp) between the two waste paper categories (Table I). It is well known that inherent fiber characteristics, including the wood species, morphological features, and chemical composition, significantly influence the final properties of paper.[44,45] Also, the production methodology affects the fiber bonding ability and, as a result, the strength properties of paper. For example, chemical pulps (kraft and sulfite) have better and

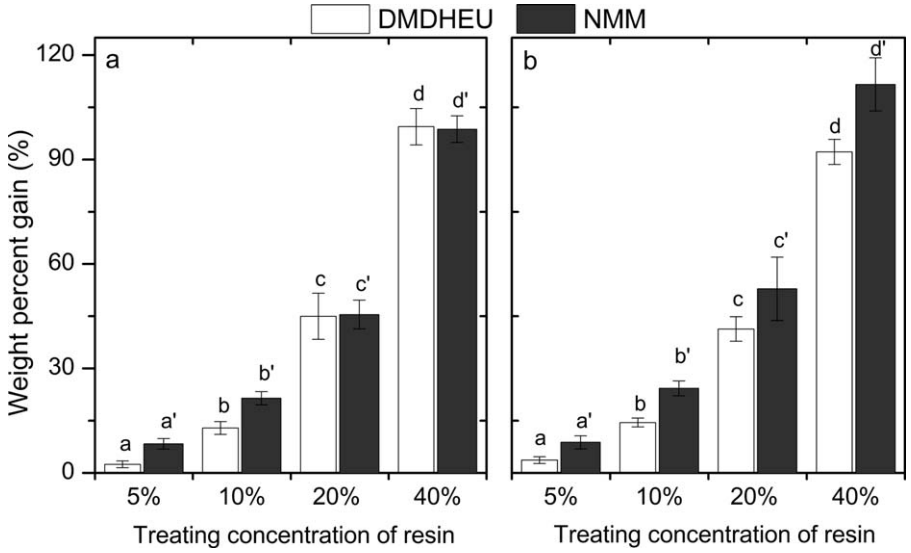

Figure 2. WPGs of the NMM- and DMDHEU-treated handsheets: (a) OCC and (b) OP. The standard deviation was based on the entire population, and the statistical differences were tested with ANOVA and a *post hoc* Tukey's HSD test. The labeled values were statistically equal at an error probability of $\alpha = 0.05$.

Table II. *n* and *q* Values of the NMM- and DMDHEU-Treated Hand-Sheets from the OCC and OP Waste Categories

	n (%)[a]		*q* (%)[b]	
Treatment/concentration	OCC	OP	OCC	OP
DMDHEU				
2%	0.9	0.9	4.2	4.2
5%	1.9	1.8	8.8	8.3
10%	3.1	3.1	14.4	14.4
20%	5.6	4.9	26.0	22.7
40%	7.3	6.8	33.9	31.6
NMM				
2%	2.9	2.7	5.5	5.2
5%	6.7	7.1	12.7	13.7
10%	10.4	11.7	19.7	22.7
20%	14.7	15.2	27.9	29.5
40%	21.3	22.1	40.4	42.7

[a] Mean values of two repetitions.
[b] Calculated NMM and DMDHEU contents in the treated sample.

more uniform fiber quality with generally less lignin and proportionately more intact fibers than mechanical and semichemical pulps.[46]

In the OCC waste paper category, the share of chemical pulp was much lower (30 wt %) than those of semichemical and chemimechanical pulp (70 wt %), whereas in the OP category, differences in weight were not that great between the pulp types (44 and 56 wt %, respectively).

The OP category had a balanced share of softwood fibers per pulp type. In the case of OCC, the amount of softwood semichemical and chemimechanical pulp exceeded considerably (more than double) that of softwood chemical pulp. Softwood fibers of chemical origin were more abundant in the OP than in the OCC category, and the opposite occurred for the semichemical and chemimechanical pulp. The weight proportion of hardwood fibers was not found to differ between the waste paper categories but the semichemical and chemimechanical fibers were almost double in weight compared to the chemical fibers. A trivial amount of nonwood fibers of 0–1 wt % was found with no differences between the waste paper categories and types of pulp.

WPG and Resin Content

The weight gain of the treated handsheets increased linearly with increasing concentration of NMM and DMDHEU (Figure 2). The OCC and OP waste categories exhibited only minor differences in weight gain. The NMM treatment caused a slightly higher WPG than the DMDHEU treatment in both waste paper categories at given concentrations. However, the differences were not statistically significant. It should be noted that both

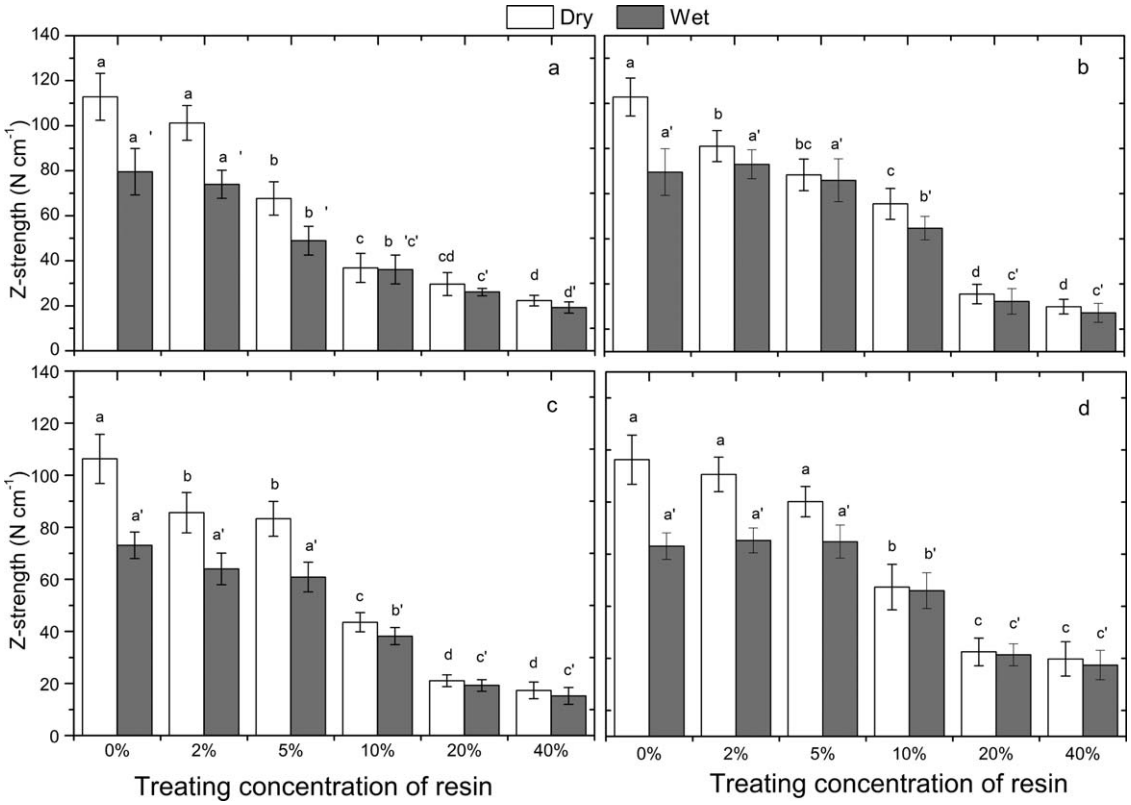

Figure 3. Z-strength of the recovered fibers treated with different concentrations of resins in dry and wet measuring conditions: (a) OCC treated with DMDHEU, (b) OCC treated with NMM, (c) OP treated with DMDHEU, and (d) OP treated with NMM. The standard deviation was based on the entire population, and the statistical differences were tested with ANOVA and a *post hoc* Tukey's HSD test. Values labeled with the same letter were statistically equal at an error probability of $\alpha = 0.05$.

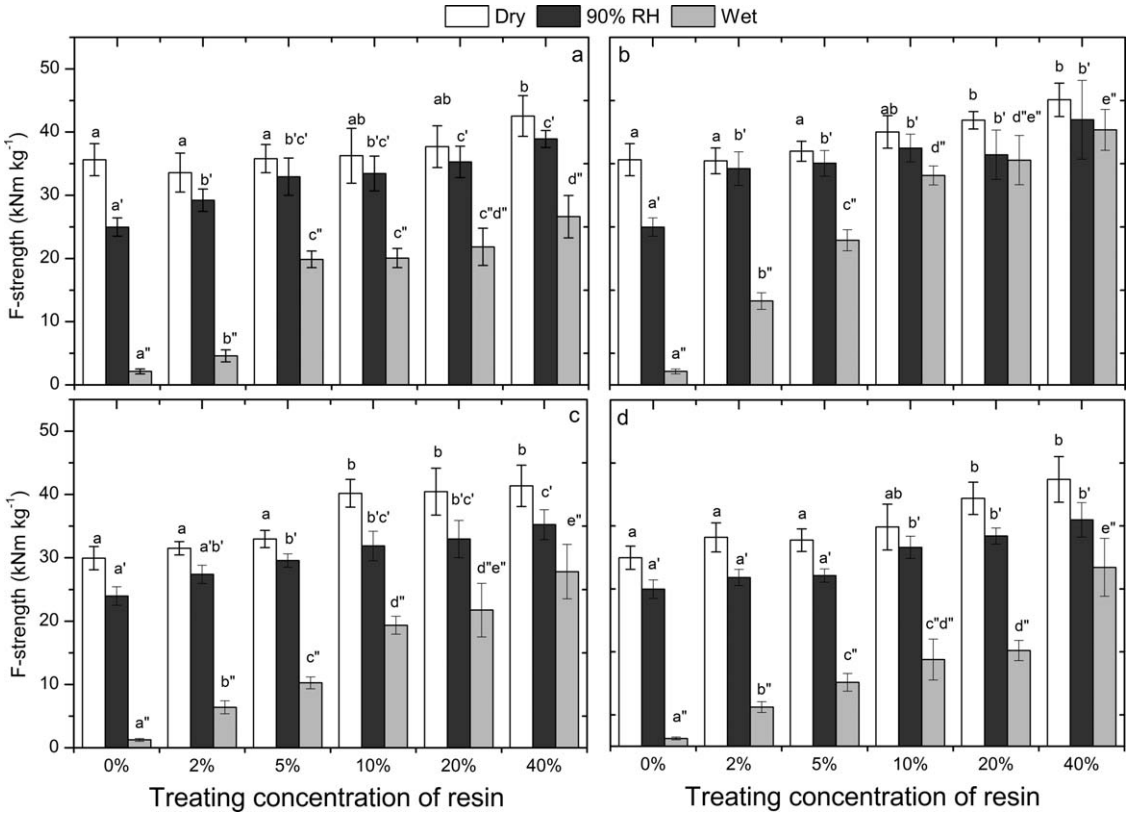

Figure 4. F-strength of OCC and OP treated and untreated handsheets in dry, wet, and 90% RH measuring conditions: (a) OCC treated with DMDHEU, (b) OCC treated with NMM, (c) OP treated with DMDHEU, and (d) OP treated with NMM. The standard deviation was based on the entire population, and the statistical differences were tested with ANOVA and a *post hoc* Tukey's HSD test. Values labeled with the same letter were statistically equal at an error probability of $\alpha = 0.05$.

the NMM and DMDHEU treatments at the lowest concentrations of 2% did not cause a noteworthy weight gain, and thus, these results are not presented in Figure 2.

The nitrogen content was similar in the untreated sample and the low-concentration (2%) DMDHEU-treated OCC and OP samples (Table II). The NMM-treated handsheets showed a considerably higher nitrogen content than those treated with DMDHEU for both the OCCs and OP. The higher nitrogen content of the NMM-treated samples was attributed to the higher amino content of NMM rather than DMDHEU (Figure 1). In the higher concentrations of the NMM-treated samples (10, 20, and 40%), OP had a slightly higher nitrogen content than the OCCs. The calculated resin content showed that the amount of NMM and DMDHEU increased gradually with increasing concentration (Table II). At the highest concentrations of 20 and 40%, the content of DMDHEU was slightly higher in the OCCs than in the OP. On the other hand, the NMM-treated OCC samples exhibited a generally lower resin content than the OP samples.

Z-Strength

The Z-strength is based on the individual fiber strength. The untreated OCC samples exhibited a Z-strength of 113 and 80 N/cm in dry and wet measuring conditions, respectively (Figure 3). A similar reduction in the Z-strength under wet conditions was observed in the untreated and treated OCC samples [Figure 3(a,b)]. The treatment of the OCC fibers with low concentrations (2%) of DMDHEU and NMM slightly decreased the individual fiber strength. The influence of the DMDHEU and NMM resins on the Z-strength of the OCC-treated fibers clearly depended on the concentration of the chemicals. The OCC handsheets treated with 10, 20, and 40% of DMDHEU lost 67, 74, and 78% of their dry Z-strengths, respectively [Figure 3(a)]. Like DMDHEU, NMM treatment also reduced the Z-strength of the OCC fibers under wet and dry conditions, but the strength loss was lower. In comparison to DMDHEU, the NMM treatment showed the relatively lower reduction in the dry Z-strength at 5 and 10% concentrations. The NMM-treated OCC fibers, however, showed considerably higher wet Z-strengths at 5 and 10% concentration than the DMDHEU-treated ones [Figure 3(b)].

The OP recycled fibers displayed Z-strengths of 107 and 73 N/cm in dry and wet measuring conditions; this was lower than that of the OCC fibers [Figure 3(c,d)]. The lower Z-strength of the untreated OP fibers might have been related to the bleaching process; this caused some damage to the fibers.[47] Like the OCC-treated fibers, the Z-strength of the OP fibers decreased more strongly with the DMDHEU treatment than with NMM treatment under dry and wet conditions.

Generally, strength loss in resin-treated cellulosic fibers is caused by two main factors: (1) enhanced stiffness and brittleness of

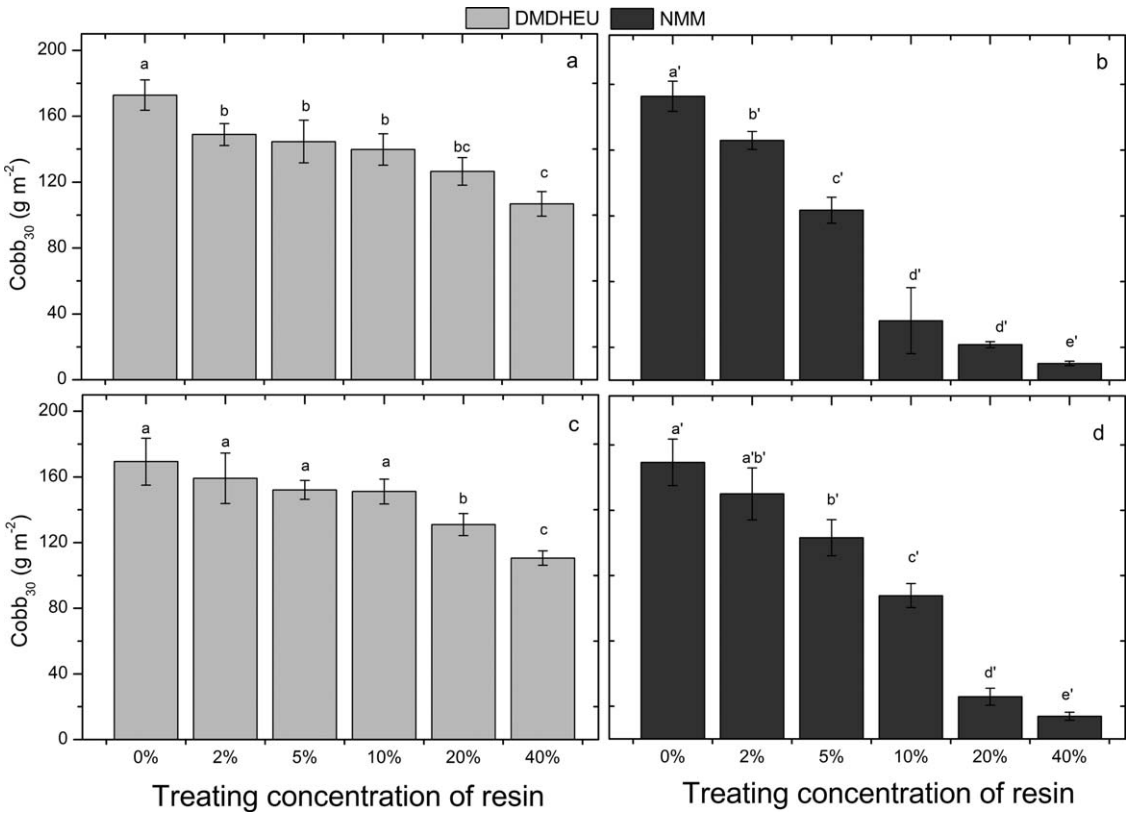

Figure 5. Water absorption ($Cobb_{30}$) of the OCC and OP treated and untreated handsheets: (a) OCC treated with DMDHEU, (b) OCC treated with NMM, (c) OP treated with DMDHEU, and (d) OP treated with NMM. The standard deviation was based on the entire population, and the statistical differences were tested with ANOVA and a *post hoc* Tukey's HSD test. Values labeled with the same letter were statistically equal at an error probability of $\alpha = 0.05$.

the fibers, which is induced by the embedding of the fibers in a rigid matrix and by the crosslinking of cell wall polymers and (2) the degradation of cellulose molecules with an acid catalyst in the curing process.[48–51] Increased stiffness caused by thermosetting resins inhibits the untwisting of the fibers.[52] It was also conceivable that crosslinking agents impede any slippage and reduce the flexibility of single microfibrils; this is necessary to give a more uniform sharing of applied stress and thus decreases the fiber strength.[52,53] Moreover, the higher strength reduction of the DMDHEU-treated samples might have been due to the hydrolysis of cell wall polymers, which resulted in a reduced degree of polymerization of the polysaccharides and tensile strength loss. Hydrolysis was catalyzed through proton acids or Lewis acids, which were applied in combination with DMDHEU.[36,54]

F-Strength

The F-strength assesses the bonding between the fibers. The untreated OCC fibers showed F-strengths of about 34, 25, and 2 kNm/kg in dry, 90% RH, and wet measuring conditions, respectively (Figure 4). Lower concentrations of DMDHEU and NMM (2 and 5%) did not change the dry F-strength in comparison to that of the control. The treatment of the OCC fibers with low concentrations of resins, however, significantly increased the F-strength in the 90% RH and wet measuring conditions. There were no obvious differences found in the wet F-strengths of the 10, 20, and 40% DMDHEU-treated OCC fibers. High concentrations of NMM (10, 20, and 40%) considerably enhanced the F-strengths under moist conditions. In comparison to DMDHEU, NMM treatment improved the moist F-strength of the OCC fibers more strongly at given concentrations.

The control OP fibers had F-strengths of 30, 25, and 1 kNm/kg in dry, 90% RH, and wet measuring conditions, respectively (Figure 4). The higher F-strength of the untreated OCC samples compared to that of OP might have been related to the lower fiber damage on the unbleached pulp.[55] Indeed, after the recycling process, the fibers became less flexible and conformed. The result of this loss in flexibility and conformability was weaker interfiber bonding[56] and, consequently, a lower strength in both types of handsheets.

Higher concentrations of DMDHEU and NMM apparently improved the dry F-strength of the OP samples. The wet F-strength of the OP handsheets increased with increasing concentrations of DMDHEU and NMM.

The improvement in the wet F-strength of the handsheets from recovered fibers at the lowest NMM and DMDHEU concentrations (2%) could be explained by their action as wet strength agents. After the absorption of the resins and possible penetration into the cell walls of the fibers (depending on the contact

time and the molecular weight of the resin), the resins condensed. The condensation reaction was slightly different in the resins; normally, NMM undergoes an autocondensation reaction,[57] whereas the condensation reaction in DMDHEU was attributed to a combination of autocondensation and crosslinking.[58] The condensed resins formed an interpenetrating network within the cell wall of the fibers[26,59] and improved the wet strength of the paper.

At higher concentrations of the resins, however, the fibers were embedded in the polymer and acted as reinforcements in the resin composite, whereas the interfacial adhesion between the fibers was improved by the resin.[60–64] Thus, the improvement in the strength of the handsheets at high concentrations of resins might have been related to the structure of the resin matrix during the condensation and bonding between the fibers and resin.

Water Absorption

The water absorption properties of the paper depended on the fiber type and the coating material,[65] and laminate papers, which are used as a surface layer, must be water resistant. The Cobb values of the untreated OCC and OP handsheets, which reflected the amount of water absorption, were 173 and 169 g/m^2, respectively. Cobb tests performed on the treated handsheets showed that the water absorption capacity decreased with increasing resin concentration (Figure 5). The OCC samples showed lower water absorption than the OP samples at the same concentrations.

The NMM-treated samples displayed a lower water absorption than the DMDHEU-treated ones. The treatment of the OCC and OP handsheets with 40% DMDHEU decreased the water sorption by 38 and 35%, respectively, as compared to the controls. The Cobb value of the NMM-treated handsheets gradually reduced with increasing NMM concentration. The OCC and OP samples treated with 40% NMM exhibited the strongest reduction of the Cobb value by 94 and 92%, respectively, in comparison to the controls. The lower improvement in the waterproof performance of the DMDHEU treatment in comparison to NMM might have been associated with the hydrophilicity of DMDHEU because of the two hydroxyl groups in the building block, whereas the presence of amino groups in the structure of NMM, which led to stronger crosslinking, induced the resin to be more hydrophobic.[66]

CONCLUSIONS

This study revealed that recovered fibers modified with high concentrations of thermosetting resins have the potential to be applied as an alternative raw material for laminated paper in the WBP industry. The application of NMM and DMDHEU revealed comparable effects on the mechanical properties of the resulting laminates: it decreased the individual Z-strength of the fibers, and it increased the tensile strength of the whole compound measured at finite span, particularly under wet conditions. NMM was considerably more effective in reducing the water absorption of the handsheets because of the more hydrophobic character of the resin as compared to that of DMDHEU. Thus, NMM could be (partly) replaced for the production of laminates in applications where hydrophobicity plays a minor role, such as for indoor applications.

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