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Turning low-cost recycled paper into high-value binder-free all-cellulose panel products

Raquel Arévalo

School of Engineering and Materials Science and Materials Research
Institute, Queen Mary University of London, London, UK

Nattakan Soykeabkaew

Center of Innovative Materials for Sustainability (iMatS), School of Science,
Mae Fah Luang University, Chiang Rai, Thailand

Ton Peijs

Materials Engineering Centre, WMG, University of Warwick, Coventry, UK
(corresponding author: t.peijs@warwick.ac.uk)

In this work, the feasibility of producing low-cost, recyclable and biodegradable binderless all-cellulose fibreboards was demonstrated through the successful manufacturing of self-binding composites based on recycled paper and kraft fibres. These all-cellulose composites were made by a simple and environmentally friendly solvent-free mechanical fibrillation method using water as a processing aid. Kraft fibres and recycled paper pulp were mixed in a 30/70 w/w ratio and refined simultaneously in a Valley beater for different periods of time. The mechanical properties of the composites, consisting of a 'matrix' of recycled paper reinforced with kraft fibres, were highly dependent on the level of fibrillation of the kraft fibres. Optimal levels of fibrillation resulted in a good balance of self-binding properties and reinforcing efficiency. The hot-pressed all-cellulose panels exhibited a flexural strength of 75 MPa and a modulus of 5.9 GPa, which are impressive values compared with those of conventional fibreboard materials. In addition, these panels showed better water resistance than some existing wood panel products. These binder-free all-cellulose composites have potential as cost-effective sustainable alternatives to existing panel board products, as they are entirely made from renewable and recycled materials, which at the end of life can potentially be recycled or composted, making them perfectly fit for a circular economy.

Notation

P	pressure
T	temperature
t	time
W_d	weight of the dry sample
W_w	weight of the wet sample
ρ_c	density of cellulose
ρ_p	density of the panel
$^{\circ}\text{SR}$	Schopper–Riegler degree

1. Introduction

The early idea of green composite materials started with the use of natural fibres to replace glass fibres as a reinforcement for polymeric resins.^{1,2} These materials were of interest since they originate from a renewable resource, are biodegradable and lightweight and exhibit good specific strength and stiffness; they can also be incinerated with high energy recovery. However, a notable feature of lignocellulosic fibres is their hydrophilicity, which makes them compatible with hydrophilic polymers but mostly incompatible with hydrophobic polymer matrices, such as polypropylene.³ This issue leads directly to poor fibre–matrix adhesion and promotes void formation, which are both major strength-reducing factors in natural fibre composites.^{3,4} One of the potential solutions to enhancing fibre–matrix adhesion in cellulose-based composites, while at the same time creating a green material, is through the development of self-reinforced or all-cellulose composites.^{2,5,6}

Prior works in the area of all-cellulose-based composites generally involved the use of solvents to dissolve the cellulose fibre surface

selectively to form a matrix phase and fuse the cellulose fibres together.^{7–11} Despite the remarkable mechanical properties of these all-cellulose composites, with moduli up to 20 GPa and tensile strengths in excess of 200 MPa, the need for solvents in their processing makes this solution not really an environmentally benign one. Moreover, the solvent exchange process allows only for the development of relatively thin two-dimensional sheet-like materials. Hence, there is a need for a green solvent-free manufacturing process for all-cellulose composites, which allows for the possibility of creating thicker panel board products.

The recent years have seen a growing interest in producing binder-free or binderless fibreboards and composites made from different sources of lignocellulosic materials, as they can have both environmental and economic benefits. Binder-free fibreboards can provide fully recyclable, biodegradable and renewable green products while requiring no synthetic resins, binders or solvents in the production process. Moreover, as the physical properties of these materials make use of the self-binding capacity of nanocellulose networks through van der Waals interactions and/or hydrogen bonds as in paper, these materials are in principle recyclable similarly as paper. This makes these materials fit for a circular economy, in which it is aimed to minimise waste and make the most of resources by recovering and recycling products at the end of their life.

The optimisation of the self-binding capacity of cellulose networks relies on increasing the surface area of the cellulose fibres through micro- or nanofibrillation. Bacterial cellulose^{12,13} and cellulose

nanopaper,^{2,14} consisting of either cellulose nanofibres directly obtained from bacteria or cellulose nanofibres extracted from wood, are prime examples of self-binding cellulose materials. Initially, most research into cellulose networks was devoted to thin films and nanopaper exhibiting impressive tensile strength values of around 250 MPa. However, in recent years, more studies have been aimed at developing construction materials such as panel board products based on self-binding nanocellulose. For example, with the refinement of lignocellulosic materials, such as flax, into self-binding nanofibrillated cellulose, the authors recently showed that the strength of such binderless all-cellulose panels can be raised to 120 MPa with moduli of up to 17 GPa.¹⁵ These mechanical properties are quite impressive and outperform those of most conventional natural-fibre-reinforced plastics (NFRPs) or wood fibre panel products.^{15–17} However, the use of natural fibres, such as flax and hemp, for the manufacturing of these products at the industrial scale could pose problems related to their availability, storage and particularly price. More recently, cellulose nanofibre boards made from fibrillated softwood with a nominal thickness of 3 mm were prepared by using a press apparatus to dewater the cellulose nanofibre network prior to drying.¹⁸ The maximum flexural and tensile strengths of these cellulose nanofibre boards were 162 and 85 MPa, respectively.

The uses of flax fibres,¹⁵ cotton stalks,¹⁶ hardwood and softwood pulps,¹⁷ softwood sawdust,¹⁹ kraft lignin²⁰ and banana bunch²¹ as raw materials to prepare binder-free all-cellulose panels have all been demonstrated. From these studies, many processing parameters have been identified as crucial for mechanical and physical properties of such materials, including pretreatment and refinement of the raw biomass materials and pressing and drying conditions of the moulded panels. The strength and modulus values of such binder-free cellulose panels are typically around 20–30 MPa and 2–4 GPa, respectively.^{17,19–21} More recently, with the self-binding potential of nanocellulose recognised, a number of studies have shown its use as a sustainable adhesive replacement in paper, fibreboard, formaldehyde-free hybrid composites and wood-based panels.^{22–26} For instance, a laminate system composed of stacked sheets of paper impregnated with cellulose nanofibrils was pressed and bonded together. It was found that the flexural modulus and strength of these composite laminates were as high as 3.6 GPa and 39 MPa, respectively.²⁷ In another approach, a nanocomposite laminate was produced by dipping strips of paper in a cellulose nanofibril suspension, cross-folding the soaked strips into a wet laminate and hot-pressing it in a hydraulic press to obtain a stiff laminate. This laminated paper sample showed a flexural strength of around 100 MPa and a flexural modulus of 8 GPa.²⁸ Interestingly, the mechanical performance of this laminate exceeded that of short glass- or natural-fibre-reinforced plastics, as well as that of some wood fibre panel boards.

To endorse a circular economy further, the use of alternative cellulose sources from waste streams such as recycled paper could prove an interesting cost-effective alternative for binderless all-cellulose composites due to their relatively low cost and availability

throughout the year, thus eliminating possible problems associated with storage. However, it is well known that recycled paper fibres present inferior strength properties and lower bonding capability compared with virgin or freshly pulped fibres as a consequence of chemical and physical changes undergone during pressing, drying, printing, storage, re-pulping and de-inking.²⁹ Most of the changes that reduce the strength properties of recycled paper fibres arise from the drying process since it brings about partially irreversible closure of small pores in the fibre wall, as well as an increased resistance to swelling during rewetting.³⁰ The phenomenon associated with this partially irreversible loss of flexibility and swelling capacity of fibres after drying is known as hornification. Hornification occurs when hydrogen bonds formed between cellulose chains in the cell wall during drying cannot be broken during the rewetting process, resulting in only partial swelling since some chains remain bonded.^{31,32} The loss of the swelling capability of recycled paper fibres leads to reduced fibre flexibility, less fibre–fibre contacts and therefore reduced inter-fibre network bonding.³³ Therefore, drying conditions affect cellulose fibre strength, fibre swelling and binding potential.³⁴ Intrinsic fibre strength, inter-fibre bonding and inter-fibre contacts are all known parameters that affect the mechanical properties of fibrous cellulose networks.^{35–37} Different approaches can be followed for the recovery of the binding potential of recycled fibres including re-refining and blending with virgin fibre pulps.³⁸

In an attempt to produce binder-free all-cellulose composites at a reduced cost compared with those reported by the authors previously,^{15,39} the use of highly fibrillated recycled paper fibres as a raw material was explored in this work. Due to the lower strength and bonding capability of these recycled paper fibres compared with those of lignocellulosic fibres such as flax and hemp, bleached kraft fibres were incorporated in the recycled paper pulp matrix with the aim of enhancing their overall mechanical performance. The reinforcing effect of fibrillated kraft fibres in the recycled paper matrix was studied as a function of refinement time. After dewatering and hot-pressing, the mechanical properties and water absorption (WA) behaviour of the resulting binderless kraft fibre/recycled paper composite panels were assessed.

2. Experimental section

2.1 Materials and characterisation

In this work, recycled paper pulp was used for the production of binder-free all-cellulose composite panels. This de-inked recycled wet paper pulp was kindly provided in 25 l plastic containers by Drumparam (Austria). In order to improve the mechanical performance of these recycled paper materials, bleached kraft pulp was used as a reinforcement to create binderless kraft fibre/recycled paper composites. Bleached kraft fibres with a fibre length between 2 and 3 mm were provided in sheet form (80 × 80 cm) by Norske Skog (Norway). Figure 1 shows images of the as-received recycled paper pulp (Figure 1(a)) and bleached kraft fibre sheet (Figure 1(b)).

Figure 1. Images of (a) as-received recycled paper pulp in a 25 l plastic container and (b) bleached kraft fibre sheet (80 cm × 80 cm)

To improve inter-fibre binding between the recycled paper pulp matrix and the bleached kraft fibre, a high degree of microfibrillation of the cellulose fibres is required, as previously demonstrated in many works.^{15–17,19,20} Pieces of kraft paper were first soaked in water and disintegrated in a Valley beater for 15 min. After disintegration, the kraft fibres were refined in the Valley beater for different periods of time (5, 10, 15, 20, 25 and 30 min). The level of microfibrillation was first indirectly evaluated by determining the drainability of the refined pulp suspensions through the Schopper–Riegler (SR) method.⁴⁰ This test measures the rate of water drainage from the pulp fibres and gives an indication of the so-called freeness of the pulp and degree of fibrillation and hydration of the pulp fibres.

Figure 2 shows the SR apparatus employed for performing such tests together with a working schematic diagram. The functioning of this

device is quite simple, as can be deduced from the schematic diagram. The technology is based on the principle that finer pulp suspensions are more hydrated, slowing down the water drainage through the filter and allowing most of the water to pass through orifice B. The small amount of water collected from orifice A indicates the SR degree ($^{\circ}\text{SR}$) of the pulp in the collector vessel. Hence, the $^{\circ}\text{SR}$ value of high-fibrillated pulp will be higher than that of low-fibrillated pulp. For lightly beaten pulp suspensions, the water drainage through the filter is fast, and, consequently, water accumulates in the cone. In this case, most of the drained water will pass through orifice A instead of B, leading to a lower $^{\circ}\text{SR}$ value. Pulp suspensions of kraft fibres for SR testing were prepared and tested according to ISO standard 5267.⁴¹ At least five suspensions of each pulp were tested, and the $^{\circ}\text{SR}$ values were averaged. The morphology of the kraft fibres as a function of refinement time was also observed using an Olympus BX60 optical microscope.

Figure 2. Image of the SR test set-up and working principle for the high-fibrillated pulp (a) and low-fibrillated pulp (b)

Table 1. Hot-pressing conditions applied for the successful compaction and drying of recycled-paper-based composites

Hot-pressing parameters		
Phase 1	Phase 2	Phase 3
$T = 140^{\circ}\text{C}$	$T = 140^{\circ}\text{C}$	$T = 20^{\circ}\text{C}$
$P = 20\text{ bar}$	$P = 40\text{ bar}$	$P = 40\text{ bar}$
$t = 1200\text{ s}$	$t = 1200\text{ s}$	$t = 600\text{ s}$

P , pressure; T , temperature; t , time

2.2 Preparation of binderless all-cellulose composite panels

First, the kraft paper sheets were soaked in water and disintegrated in a Valley beater for 15 min prior to the addition of the recycled paper pulp. After disintegration, recycled paper was added to create a 30/70 w/w kraft fibre/recycled paper blend, which was refined in the Valley beater for different periods of time (5, 10, 15, 20, 25 and 30 min), with the resulting blends used for the production of self-binding all-cellulose composite panels. In this way, the effect of the degree of fibrillation of the kraft fibres and bonding of both types of fibres on the mechanical properties of the kraft fibre/recycled paper composites can be studied. To form binderless all-cellulose composite panels, after mixing and refining, the blended fibre pulp was subsequently filtered and pressed at room temperature for initial dewatering. Finally, this partially dewatered precursor cake was dried in a hot-press under conditions listed in Table 1. Hot-pressing conditions for the kraft/recycled paper composites were based on the authors’ previous work using microfibrillated flax fibres,^{20,35} albeit using a somewhat lower maximum pressure to avoid cracking during moulding. The compression-moulding cycle consisted of a drying, a compacting and a cooling phase using a maximum pressure of 40 bar at 140°C and resulted in panels with a nominal thickness of around 8 mm.

2.3 Characterisation of binderless all-cellulose composite panels

Three-point bending tests on the panels were performed using an Instron 5584 universal tester at room temperature with a 1 kN load cell. The cross-head speed was 5 mm/min. In order to account for the potential anisotropic behaviour of the panels, specimens were cut into strips (25 mm × 160 mm) in both longitudinal and transverse directions according to the ASTM D790-02⁴² standard (procedure A) (Figure 3(a)). At least eight specimens of each sample were tested (Figure 3(b)), and values were averaged.

The microstructure of the binderless all-cellulose composites was analysed by scanning electron microscopy (SEM), using a Jeol JSM 6300 microscope at an accelerating voltage of 10 kV. Prior to examination, the surface of the samples was sputtered with a thin layer of gold to make them conductive.

The density of the all-cellulose panels was measured directly by weighing the hot-pressed panels. Porosity can then be calculated from the measured density following the equation

Figure 3. (a) Image of an all-cellulose panel showing the location of test samples cut in both longitudinal and transverse directions according to the ASTM D 790-02 standard (procedure A); (b) image of an all-cellulose composite sample subjected to a three-point bending test

1. $\text{porosity (\%)} = \left(\rho_c - \rho_p \right) / \rho_c \times 100$

where ρ_c is the density of cellulose (1.6 g/cm³) and ρ_p corresponds to the density of the panel.

The WA of the best-performing binderless all-cellulose composite panels was monitored at regular intervals. Prior to carrying out these tests, the samples were dried in an oven at 60°C until a constant mass. The dried specimens were then submerged in a Grant Y-38 distilled water bath at 23°C, removed, wiped dry with a paper towel and weighed using a high-precision balance at selected time intervals. The WA was determined from the weight differences using the following equation

2. $\text{WA(\%)} = (W_w - W_d) / W_d \times 100$

where W_w and W_d are the weight of wet and dry samples, respectively. Times ranging from 0 to 48 h were studied.

3. Results and discussion

First, the refinement of pure kraft fibres in the Valley beater was studied separately in order to characterise the level of fibrillation and morphology as a function of the refinement time. Before actual refinement, the kraft paper was first disintegrated in the Valley beater. The final degree of fibrillation of the kraft fibres was determined by the SR method for different refinement times, as shown in Figure 4. For the first 15 min of refining, only a slight increase in the degree of fibrillation of the kraft fibres can be observed. However, a further increase in refinement time leads to a drastic improvement in the °SR value, indicating greater fibre fibrillation. The morphology of the kraft fibres as a function of refinement time as observed by optical microscopy (OM) is presented in Figure 5. The OM images clearly support the results obtained through the SR method, evidencing that effective microfibrillation is obtained only after 15 min of refinement. This implies that effective inter-fibre bonding between kraft fibres and recycled paper may be achieved only after 15 min of refinement.

Figure 4. Effect of the refining time on the °SR value of kraft fibre pulp. The optimum level of microfibrillation of the kraft fibres for the production of self-binding kraft/recycled paper composites – as will be shown later – is encircled

It is worth highlighting that 30 min of refining leads to excessive fibrillation and damage of the kraft fibres, with little remaining of the original kraft fibre microstructure (Figure 5(f)).

Next, the influence of refinement time on the physical properties of the hot-pressed all-cellulose composite panels was studied. Again, kraft paper was first disintegrated in the Valley beater before wet recycled paper was added for final refinement of the blend in the Valley beater for different periods of time. The influence of refinement time on the flexural properties of the kraft fibre/recycled paper composites (30/70 wt.% ratio) is shown in Figure 6. As expected from the separate kraft fibrillation studies,

the effect of microfibrillation on the flexural properties of the self-binding kraft fibre/recycled paper composites was not evident up to 15 min of refinement, after which a strong improvement of both flexural modulus and flexural strength was observed. A further increase in refinement times leads to a drastic improvement in microfibrillation, as shown in Figures 4 and 5, explaining the sudden improvement in composite properties for refinement times of 20 and 25 min. This sudden enhancement of panel properties is associated with an increase in panel density from around 1.15–1.20 g/cm³ for panels with refinement times of up to 15 min and densities of 1.35 g/cm³ for panels with refinement times of ≥ 20 min. At 25 min of refinement time, the flexural properties reach maximum values of 5.9 GPa and 75 MPa for stiffness and strength, respectively. Compared with the values for conventional panel board products such as oriented strand board (OSB) or medium-density fibreboard (MDF) and wood fibre plastic (WFP) or NFRP, these values are very favourable.^{43,44} The current low-cost binder-free all-cellulose composites show a slightly higher or comparable stiffness but significantly greater strength compared with OSB, MDF, WFP or NFRP with typical strength values ranging from 20 to 50 MPa.^{15,43,44} The lower strength of conventional panel board materials can again be linked to the relatively low density of such panels. Typical board densities for MDF are 0.4–0.8 g/cm³ and that for OSB is 0.7 g/cm³, significantly lower than those for the microfibrillated all-cellulose composite panels.

Compared with the authors' previously reported self-binding all-cellulose composites based on 100% microfibrillated flax fibres,¹⁵ the current composites have slightly weaker mechanical properties. Flax-fibre-based self-binding all-cellulose panels based on fibres with optimised fibre lengths exhibited a maximum panel density of 1.41 g/cm³ and maximum flexural strength and modulus of 120 MPa and 17 GPa,²⁰ respectively, compared with values of 1.35 g/cm³, 75 MPa and 5.9 GPa for the current panels,

Figure 5. Effect of refinement time on the morphology of microfibrillated kraft fibres. The OM images show kraft fibres refined in a Valley beater for (a) 5, (b) 10, (c) 15, (d) 20, (e) 25 and (f) 30 min

Figure 6. Effect of refinement time of kraft fibre/recycled paper (30/70 w/w) blend on the (a) flexural modulus and (b) flexural strength of dewatered and hot-pressed binderless recycled paper composites reinforced with 30 wt.% bleached kraft fibres. For comparison, the mechanical properties of materials made solely from recycled paper are also included (green bars)

respectively – that is, a 65% and a 37% reduction in mechanical properties. However, from a circular economy and raw material cost point of view, the current panels are still of great interest. The recycled-paper-based composites have significant environmental benefits, as they are based on waste stream materials that are upgraded into a high-quality construction material. Moreover, this material can again be recycled in principle into all-cellulose panels or other paper-based products. Likewise, the current all-cellulose composite materials may have some significant cost advantages, as they are 70% based on recycled paper and 30% kraft paper, both of which are considerably cheaper than flax fibres. Based on an estimated price for recycled paper of €0.10/kg and €1/kg for kraft paper, the raw material price of the blend works out at €0.37/kg, which is significantly less (78%) than that of flax fibres with an estimated price of €1.7/kg.⁴⁵

Excessive fibrillation and damage of the kraft fibres ($^{\circ}\text{SR}$ value of 58) after 30 min of refining lead to a significant loss of reinforcing efficiency. The optimum degree of fibrillation of kraft fibres for use as reinforcements in recycled-paper-based composites corresponds to an $^{\circ}\text{SR}$ value of around 35 and was achieved after a refining time of 25 min in a Valley beater (see Figure 4).

The sudden improvement in the mechanical properties of the kraft fibre/recycled paper composites after 15 min of refinement could be attributed to the difficulty of re-pulping the once-dried bleached kraft pulp due to the strong hydrogen bonding established between kraft fibres during drying.⁴⁶ Gurnagul *et al.*⁴⁷ studied the physical properties and response to refining of never-dried and dried kraft pulps and reported that never-dried pulps

require less energy to reach a certain degree of fibrillation due to greater swelling and conformability of never-dried fibres. Therefore, pulps should be dried only if they need to be stored or transported, as in this case. In addition, the removal of hemicellulose results in the replacement of relatively flexible cellulose–hemicellulose–cellulose bonds by more rigid cellulose–cellulose bonds, thus increasing the refining energy required for a given degree of fibrillation.⁴⁸ Therefore, the results suggest that effective fibrillation of dried bleached kraft fibres starts only after 15 min refining.

In an attempt to achieve a better understanding of the mechanical behaviour as a function of refinement time, the fracture surfaces of the kraft fibre/recycled paper composites were analysed by SEM, using a Jeol JSM 6300 microscope at an accelerated voltage of 10 kV (Figure 7). The SEM images corresponding to the kraft fibre/recycled paper composites obtained after 5 min (Figure 7(a)) and 10 min (Figure 7(b)) of refining indicate that the fibrillation process has not yet started, thus preventing good inter-fibre bonding between the kraft and paper fibres. After a refinement time of 15 min, a few fibrils appear on the kraft fibre surfaces; however, large voids around these fibres indicate that this degree of fibrillation is still insufficient for achieving effective kraft fibre–recycled paper cross-linking (Figure 7(c)). A further 5 min refining results in an enhancement of the kraft fibre specific surface area (Figure 7(d)), allowing for an increase in fibre–fibre interactions, enhanced cellulose network formation, better consolidation and improved mechanical properties. However, also here the presence of non-fibrillated kraft fibres can still be observed. A highly densified structure, consisting of highly micro- and nanofibrillated kraft fibres surrounded by recycled paper fibres,

Figure 7. SEM images of fracture surfaces of binderless recycled-paper-based composites reinforced with 30 wt.% kraft fibres refined for (a) 5, (b) 10, (c) 15, (d) 20, (e) 25 and (f) 30 min. Arrows indicate the presence of kraft fibres in the recycled paper matrix

is achieved only after 25 min of mechanical refining (Figure 7(e)). At this refinement time, the level of fibrillation of the kraft fibres is sufficient to render strong inter-fibre network formation while still preserving their fibrous microstructure and reinforcing ability, thus leading to an optimal combination of inter-fibre adhesion by the micro- and nanofibrillated network and intrinsic reinforcement by the kraft microfibrils. SEM images of kraft fibre/recycled paper composites obtained after 30 min of mechanical refining (Figure 7(f)) show excessive fibrillation, as well as great microstructural damage and destruction of the kraft fibre structure and aspect ratio, leading to a loss of the intrinsic reinforcing efficiency and weakening of composite properties. The distinction between kraft fibres and recycled paper fibres in this composite structure becomes less clear due to the high degree of fibrillation of the kraft fibres.

Finally, the WA behaviour of well-compacted binder-free kraft fibre/recycled paper composite panels was studied at regular intervals (Figure 8). Moisture absorption of cellulose-based materials is clearly a concern for durable engineering applications.

Figure 8. WA of binderless recycled paper composite reinforced with 30 wt.% kraft fibres refined for 25 min

This is particularly the case for all-cellulose composites, as here protection by a polymer matrix as in the case of wood- or natural-fibre-reinforced plastics is absent.⁴⁹ For this study, a well-densified composite panel with optimal mechanical properties was selected. Based on a panel density of 1.35 g/cm^3 and a cellulose density of 1.6 g/cm^3 , the porosity content of this panel was estimated using Equation 2 to be 16%.

The initial WA of the panels is very rapid, with saturation occurring after some 6 h of immersion, and involves the absorption of water by both pores and cell walls. The fibre saturation point (FSP) of wood fibres is around 25%. Below the FSP, all moisture is considered to be absorbed exclusively by the cell walls, whereas, above the FSP, moisture is exclusively accommodated in empty pores or capillaries.⁵⁰ When the panels reach 43% WA at equilibrium, this means that 18% of the weight is water in pores, whereas 25% is absorbed by the cell walls. These estimates are in good agreement with the calculated porosity of the panel of 16%. Once the pores and cell walls are saturated with water, the WA is significantly reduced, leading to an almost constant weight and a water saturation level of 43%. This level of WA is, as expected, higher than that of a typical NFRP, such as polypropylene/flax ($\sim 20\%$ WA),⁵¹ where lignocellulosic fibres are protected by a hydrophobic polymer matrix, but still lower than those of most conventional wood panel products, such as OSB (75% WA),⁵¹ MDF (65% WA)⁵² and compressed pine wood panels (55% WA).⁵³ This is particularly impressive, as here the entire material is made from cellulose and there is no polymer resin present to bind or protect the fibres. The WA of the current composite panels is also only slightly higher than those reported previously for panels based on 100% microfibrillated flax fibres (37% WA). WA in both types of all-cellulose panels is strongly dominated by porosity and disordered cellulose regions, suggesting that these parameters are

not too dissimilar for both flax-fibre-based and kraft/recycled-paper-based panels. The relatively low moisture absorption of the current kraft/recycled paper composite panels can again be directly related to the highly dense microstructure of compression-moulded microfibrillated cellulose fibres, leading to significantly lower porosity levels compared with conventional panel board products.

4. Conclusions

Potentially low-cost binder-free all-cellulose composite panels were successfully produced using recycled paper as a matrix reinforced with 30 wt.% kraft fibres. Processing involved the mechanical refinement of cellulose fibres using an environmentally friendly processing method with only water as a processing aid. Surface fibrillation of the kraft fibres increased their specific surface area, thus allowing for greater inter-fibre interactions, improved cellulose network formation and better densification of the hot-pressed composite panels. The addition of 30 wt.% optimally refined kraft fibre reinforcements into a recycled paper pulp matrix led to greatly improved mechanical properties of these all-cellulose composite panels. Optimal micro- and nanofibrillation of the kraft fibres was crucial for achieving a good balance between self-binding and reinforcing efficiency of these fibres in the recycled paper matrix. Kraft fibres with a low degree of fibrillation did not possess a sufficient specific surface area to create a strong bond with the recycled paper pulp matrix, leading to poor compaction and low mechanical performance. Conversely, excessive refinement times resulted in the loss of reinforcing efficiency as a result of destruction of the internal kraft fibre structure as well as reduced aspect ratio. An optimum degree of fibrillation of the kraft fibres was achieved at a drainage resistance °SR value of 35 and was reached after 25 min of mechanical refinement in a Valley beater. In summary, the addition of 30 wt.% kraft fibres to recycled paper pulp did result in a 123% improvement in panel strength (75 MPa) and a 119% increase in panel stiffness (5.9 GPa) compared with pure recycled-paper-based panels. The mechanical properties of these fully biobased panels still outperformed many conventional panel board products and wood- or natural-fibre-reinforced plastics. Since these materials mainly use waste streams rather than more expensive lignocellulosic fibre streams, the current approach could lead to highly cost-effective green panel products that can compete with or outperform existing products. Moreover, as these materials are in principle recyclable using a simple re-pulping process or compostable, they should fit well in a circular economy.

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