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Improving Recycled Paper Materials through the Incorporation of Hemp, Wood Virgin Cellulose Fibers, and Nanofibers

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Abstract: A significant increase in the consumption of recycled fiber products has been observed worldwide, and the industry is forced to solve the challenges of recycled fiber quality and add strength agents and/or virgin fibers to reach sufficient properties. In order to investigate whether the mechanical and air permeability properties of waste fiber (WF) material can be significantly improved by adding wood kraft fibers (KF), hemp soda fibers (HF), and ammonium persulfate oxidated cellulose nanofibrils (CNF), different fiber blends were prepared and tested. Results revealed the excellence of hemp fibers over wood fibers regarding the improvement of WF products. The results of WF after the addition of 10% mixed fibers (KF + HF) were higher than the results of a 50% KF addition. The impact of CNF depended on fiber composition and properties. A formula for modeling the CNF impact on different fiber compositions was proposed. Obtained fiber material samples showed suitability for home composting, thus contributing to the goals of the European Green Deal regarding reducing landfill waste and the development of cleaner products.

Keywords: paper; agro-waste; hemp fibers; mechanical properties; air permeability; packaging

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1. Introduction

A significant increase in the consumption of recycled fibers and fiber products has been observed in the last decade, particularly in the packaging industry [1]. There are several main reasons for this increase. The first is the increased online commerce and sharply accelerated need for transportation packaging [2], mostly caused by new shopping habits formed during the COVID-19 pandemic. The second is the changes in social awareness regarding the use of renewable resources and improved waste management [3], which have been guided and intensified by the European Green Deal (EGD) [4] and Sustainable Development Goals (SDG) [5]. The use and recycling of cellulose-based fibers contribute to the main goals of EGD and SDG by lowering the accumulation of landfilled packaging waste, saving water and energy resources, and reducing greenhouse gas emissions [6]. Finally, the increasing concerns about the end of life of packaging have forced the industry to use more recycled fiber and replace synthetic and harmful constituents with "greener" environmentally friendly options to provide home composting packaging in the best case.

Although there are many economic and environmental benefits of using recycled fibers, the main problem of the concept of "old fibers for new paper" is based on the low quality of the recycled fibers [1]. It is known that cellulose fiber loses its quality when it is recycled because of the mechanical and chemical recycling processes, such as deinking, bleaching, repeated heating, wetting, drying, and milling. When pressed and dried, the pulp fibers undergo transformations known as hornification. This process causes the layers within the fibers to compress against each other, resulting in a strong and mostly irreversible bonding network within the fiber. The structural and chemical changes lead to the semipermanent closure of the fibers' cell wall [7]. Consequently, during the recycling process, the porous structure of recycled fibers can only partially reopen with limited swelling, leading to a decrease in fiber quality compared to virgin fibers [8]. Various approaches have been explored to address the subpar performance of recycled fibers, including refining, incorporating dry strength additives, utilizing enzymes, and incorporating nanocellulose into the recycled pulp [9–11].

Besides the creation of new paper materials, waste cellulose fibers can be reused for alternative materials and applications, frequently in quite advanced ones. For example, cellulose-based adsorption materials with high removal efficiency can be obtained from newspaper waste via the functionalization of fibers [12], or office waste paper cellulose fibers can be used for the development and thermal insulation-related property improvement of aerogel made of polysaccharides [13]. Waste paper fibers have been used as reinforcing agents for the graphene–oxide/cellulose nano-crystal hybrid aerogels [14] and as a cross-linking agent for reinforcing polyurethane elastomer composites, resulting in better interfacial compatibility and higher thermal stability [15]. Waste paper cellulose fibers are an appropriate source of nanocellulose [16], enzymatic hydrolysis to sugars [17], the regeneration of cello-oligomers from ionic liquid solutions [18], or textile fibers used as cellulose-based Ag NPs decorated solid substrate for the identification of estrogenic toxicity in contaminated water using surface-enhanced Raman scattering [19]. Newspaper waste fibers magnetized with Fe_3O_4 and processed into composites using different techniques, such as the freeze-drying technique, are used as adsorption material [20,21] or, if chitosan was added, for covalent laccase immobilization [22].

However, the main part of reused cellulose fibers is used for paper and paper products, such as molded fiber products [23]. According to CEPI, in 2022, the recycling rate of the paper value chain reached an impressive 70.5% [24]. Therefore, the task of maintaining the properties of recycled papers at a satisfactory level is a present challenge that has been approached through different methods. Regarding natural ones, cellulose fibers, including micro and nanofibers, from different sources are used in most cases. The mechanical properties of recycled paper have been significantly improved by the addition of nanocellulose varying the sources, such as oil palm empty fruit bunches [25], corn stalks [26], wheat straw [27], wood [9], recycled pulp [28], hemp [29], and others. The addition of wood virgin pulp to recycled pulp to improve mechanical performance has been investigated [30], as well as the addition to different agro-waste, for example, rice straw [31]. Upon conducting a thorough review of the literature regarding the enhancement of recycled paper properties, the authors reached a conclusion that, in essence, no studies have been conducted to explore the impact of the combination of virgin wood and hemp fibers, and additionally CNF, on the enhancement of the paper of recycled fibers.

The advantage of hemp pulping is that there is no need for harsh chemicals associated with pulping (e.g., sulfite or kraft pulping); thus, fibers can be produced more sustainably compared with wood [32]. According to information published on the website of the European Commission, the amount of hemp cultivation increased by 62.4% from 2015 to 2019. The upward trajectory stopped only in 2020, as the COVID-19 pandemic negatively affected the global economy and supply chains; furthermore, the 2022 Russian invasion in Ukraine dealt a major blow to the industry as grain prices skyrocketed, but this factor has contributed to farmers' decisions in favor of growing hemp. The joint agreement on the permissible level of THC and support payments to hemp growers has given a positive impetus to the industry [33].

The CNF has been known as a wet-end additive in the papermaking process for years, and its role in improving the dry and wet strength of paper, also recycled, has been proven by many researchers [10,34]. The CNF can be obtained via the chemi-mechanical method [9,35] from any lignocellulose source, such as wood [36], recycled fibers [16], and agro-waste [37].

The hypothesis of the presented research is that the mechanical properties of materials made of recycled fibers can be significantly improved by adding virgin wood and hemp fibers in different amounts; furthermore, different proportions and blends create a synergistic effect for the improvement of the mechanical performance of paper. Virgin kraft softwood pulp, soda-cooked hemp fibers, and ammonium persulfate-oxidated nanocellulose were added to waste fibers to investigate in detail the effect of each fiber type and amount, their interaction, and interfering factors. Different fiber blends were prepared and tested for their mechanical and air permeability performance, while the biodegradability of the material was proved using a standard method similar to home composting.

2. Materials and Methods

2.1. Materials

Industrially recycled pulp provided by local molded fiber products company SIA "V.L.T." was used in the research as waste fibers (WF). Two batches (WF and WFc) were provided with a distance of two weeks to investigate if properties vary. According to the pulp provider, WF consisted of 60% mixed waste paper (newspaper, journals, books, office paper, cardboard, packaging paper), 30% waste cardboard, 5% printing house waste, and 5% waste from the production process of egg packaging. WF (Figure 1f) and WFc (Figure 1g) pulp was dried at 40 °C until dry and kept in a dry state until used.

Kraft fibers (KF) were obtained from softwood via the kraft pulping process in a 2 L laboratory digester at 170 °C with a cooking time of 3 h and process parameters of 57.4 g/L active alkali as NaOH, a sulfidity of 29.8%, and a liquor-to-wood ratio of 4.5 L/kg. After, cooking fibers were washed with tap water until lignin-containing black liquor was fully removed and washing water had a pH of 7. Fibers were dried at room temperature and kept dry until used (Figure 1e).

Hemp fibers (HF) were extracted from industrial hemp *Cannabis sativa* (USO-31). After the decortication, the hemp stems (Figure 1a) were air-dried and cut into 30 × 7 mm pieces (Figure 1b,c). The hemp biomass then was cooked in 4% NaOH solution at 165 °C for 75 min in a 2 L laboratory digester with a warming up speed of 2 °C/min. After the cooking fibers were washed in tap water to a residual water pH of 7 and refined to 18 °SR freeness using the Blendtec 725 (Orem, UT, USA) at 179 W for 7 min at 1.5% consistency, then dried at room temperature and kept in a dry state until used (Figure 1d). The total yield of fibers was 49.4%, calculated using the mass of the initial stem biomass.

(**a**)

(**b**)

Figure 1. *Cont.*

(**c**) (**d**) (**e**) (**f**) (**g**) (**h**)

Figure 1. Materials used for paper composites: (**a**) Stems of hemp *Cannabis sativa*; (**b**) Hemp stems after the decortication; (**c**) Hemp stems cut for pulping; (**d**) Hemp pulp; (**e**) Kraft pulp; (**f**) Waste pulp WF; (**g**) Waste pulp WFc; (**h**) Cellulose nanofibrils.

Cellulose nanofibrils (CNF) were prepared from industrial bleached hardwood kraft pulp kindly provided by Metsä Fibre (Espoo, Finland) using a combination of chemical and mechanical treatment. After soaking in distilled water and disintegration for 75,000 revolutions in the disintegrator (Frank PTI, Laakirchen, Austria), [38] pulp fibers were oxidated with ammonium persulfate at 70 °C for 4 h, according to Filipova et al. [9], to charge the surface and reduce the size of fibers. Treatment was continued with ultrasonication using the ultrasonic homogenizer SONIC-650W (MRC Ltd., Holon, Israel) for 15 min at 90% power on mode "9 sec on, 1 sec off" to induce fibrillation, which was finalized by processing in the microfluidizer (LM20, Microfluidics, Quadro Engineering, Waterloo, ON, Canada) using combined 200 and 100 μm ceramic chambers and a gradual pressure increase from 300 to 2000 bars, according to the methodology proposed in [39]. The obtained CNF 1.5% w/w solution was kept at 4 °C until it was used (Figure 1h).

The preparation of paper handsheets of different fiber compositions was realized according to ISO 5269-2:2004 [40] with a Rapid Köthen paper machine (Frank PTI, Laakirchen, Austria). The compositions were created by mixing WF with KF and HF in different mass proportions. A certain amount (in g) of dry fiber was placed in a baker and soaked in distilled water for at least 8 h, then disintegrated to 75,000 revolutions in the disintegrator (Frank PTI, Laakirchen, Austria). If the composition contained CNF, a certain amount of CFN suspension was added to the fiber mixture and was disintegrated to more than 25,000 revolutions. Samples were named according to their composition, including the abbreviations of fibers and the mass proportion (in percentage). For example, composition WF 50 KF 25 HF 25 means that it contains 50% waste fibers, 25% kraft fibers, and 25% hemp fibers. CNF was added as an additive in amounts of 1%, 3%, and 5%, calculating to the total dry mass of fibers, thus, composition WF 50 KF 25 HF 25 CNF 5 means that 5% CNF was added.

All the chemicals used for the research NaOH, Na_2S, and $NH_4S_2O_8$ were purchased from Fischer Chemical (Zürich, Switzerland) and were analytical grade.

2.2. Characterization Methods

The fiber length, width, shape, and content of fines were measured with a Fiber tester (Lorentzen and Wettre, Kista, Sweden). To investigate fiber samples by scanning electron microscopy (SEM), they were coated with gold plasma using a K550X sputter coater (Emitech, South Petherton, UK). The examination was performed using Vega TC from Tescan, Czech Republic, with software version 2.9.9.21. Paper thickness was measured according to ISO 534:2011 [41] with a micrometer F16502 (Frank PTI, Austria). The grammage (weight per unit area in g/m^2) of samples was calculated by dividing the paper sample mass by area, according to ISO 536:2019 [42]. Air permeability was tested according to ISO 5636-3:2013 [43] using the Air Permeability Tester 266 (Lorentzen and Wettre, Sweden). Samples for tensile tests were prepared with the Strip Cutter (Frank PTI, Laakirchen, Austria) and tested according to ISO 1924-2:2008 [44] using the Tensile Tester Vertical F81838 (Frank PTI, Laakirchen, Austria). Burst strength was tested according to ISO 2758:2014 [45] using a Burst Tester (Frank PTI, Laakirchen, Austria). Four paper handsheets were tested from each fiber composition.

Biodegradation research was carried out as home composting imitation, according to EN 14045:2003 [46] with minor modifications, in a climate room at 20 °C and 65% RH. Ecologically clean, natural compost (NPK 0.5-0.07-0.05, pH 6–7, organic matter content 16%, moisture > 50%) produced by Zeltabele Ltd. (Jaunauce, Latvia) was used as composting media. Paper samples measuring 5 × 5 cm were kept in compost in individual 0.5 L perforated containers, which were inserted into a bigger box filled with the same compost, with a volume of 130 L. Samples were monitored each week to track their disintegration and to fix the time for total decomposition.

Statistics and visualizations of the data were made using R, a language and environment for statistical computing (R Foundation for Statistical Computing, Vienna, Austria;

available online: https://www.R-project.org/ (accessed on 20 November 2023). R Core Team (2017)).

3. Results and Discussion

To evaluate the possibilities of improving the properties of recycled paper with natural fibers and their mixtures, the research strategy was to gradually replace recycled fibers in the paper composition with KF and HF—both individually and by combining both types—and then add CNF 1%, 3%, and 5% to the compositions. The chosen structure of the article includes the division of the result presentation and discussion into three parts—fiber characterization, fiber blends without CNF, and fiber blends with CNF.

3.1. Fiber Characterization

Waste fibers provided by molded egg packaging production company SIA "V.L.T." were from two different batches. Even though the batches were prepared according to the same technology (60% mixed waste paper (newspaper, journals, books, office paper, cardboard, packaging paper), 30% waste cardboard; 5% printing house waste, and 5% waste from producing process of egg packaging) using incoming raw material, different fiber dimensions and properties were explored (Table 1), thus, representing the challenges of the recycled fiber industry in terms of raw material heterogeneity, which may cause quality problems of final products. The investigation of fiber properties revealed differences in waste fiber length, width, shape, and content of fines, allowing the prediction of the mechanical properties of materials from WF to be more accurate than for that of WFc. The average dimensions of fibers show that they contain both hardwood and softwood fibers. Differences can be seen also visually in Figure 2a,b. WF fibers contained more nonfibrous objects and impurities, whose origin is most likely paper fillers, opacity and coloring pigments, dirt, ink residues, binding components, and other substances present in different types of recycled paper. WFc consists of fibers and fiber fragments with a width of 10–40 µm; also present are microfibrils and some fragments of indeterminate origin. Whereas, in the WF pulp there are a significant number of impurities and fiber-like objects with widths less than 10 µm.

Table 1. Properties of fibers used for paper materials. Average values and standard deviation are given.

Fibers	Fiber Length, mm	Fiber Width, µm	Fiber Shape, %	Content of Fines, %
Waste fibers WF	1.192 ± 0.060	25.7 ± 0.4	89.1 ± 0.1	9.7
Waste fibers WFc	1.141 ± 0.023	25.4 ± 0.1	88.1 ± 0.3	10.0
Kraft fibers KF	2.158 ± 0.016	29.6 ± 0.4	90.9 ± 0.1	3.2
Hemp fibers HF	0.649 ± 0.011	18.8 ± 0.4	91.8 ± 0.1	7.4

Softwood kraft fibers' characteristics were in line with the numbers for ordinary kraft pulp revealed by other authors [47–49]. SEM revealed the smooth, undeveloped surface of chemically released wood fibers (Figure 2c).

Hemp fibers produced from dried hemp stems using the soda pulping method on a laboratory scale were the shortest and the thinnest among other fibers used in the research, with average results of 0.65 mm and 18.8 µm, respectively, which is in accordance with the results obtained for fibers obtained from hemp shives via autohydrolysis, kraft pulping, and carbonate hydrolysis [50]. The length of hemp fiber depends on both the hemp plant species and the part of the stem used, as well as pretreatment and posttreatment. Fibers extracted from the hemp woody core (shives) are usually 2–3 times shorter than hardwood pulp fibers [51]. The whole plant stem was used for the research, so hemp pulp consisted of longer hemp fibers and short fibers from shives, the latter of which were in the majority, as seen in Figure 3, giving more impact on the calculations of average length. SEM images of HF (Figure 2d) revealed fiber fibrillation, which is related to the refining procedure after the pulping.

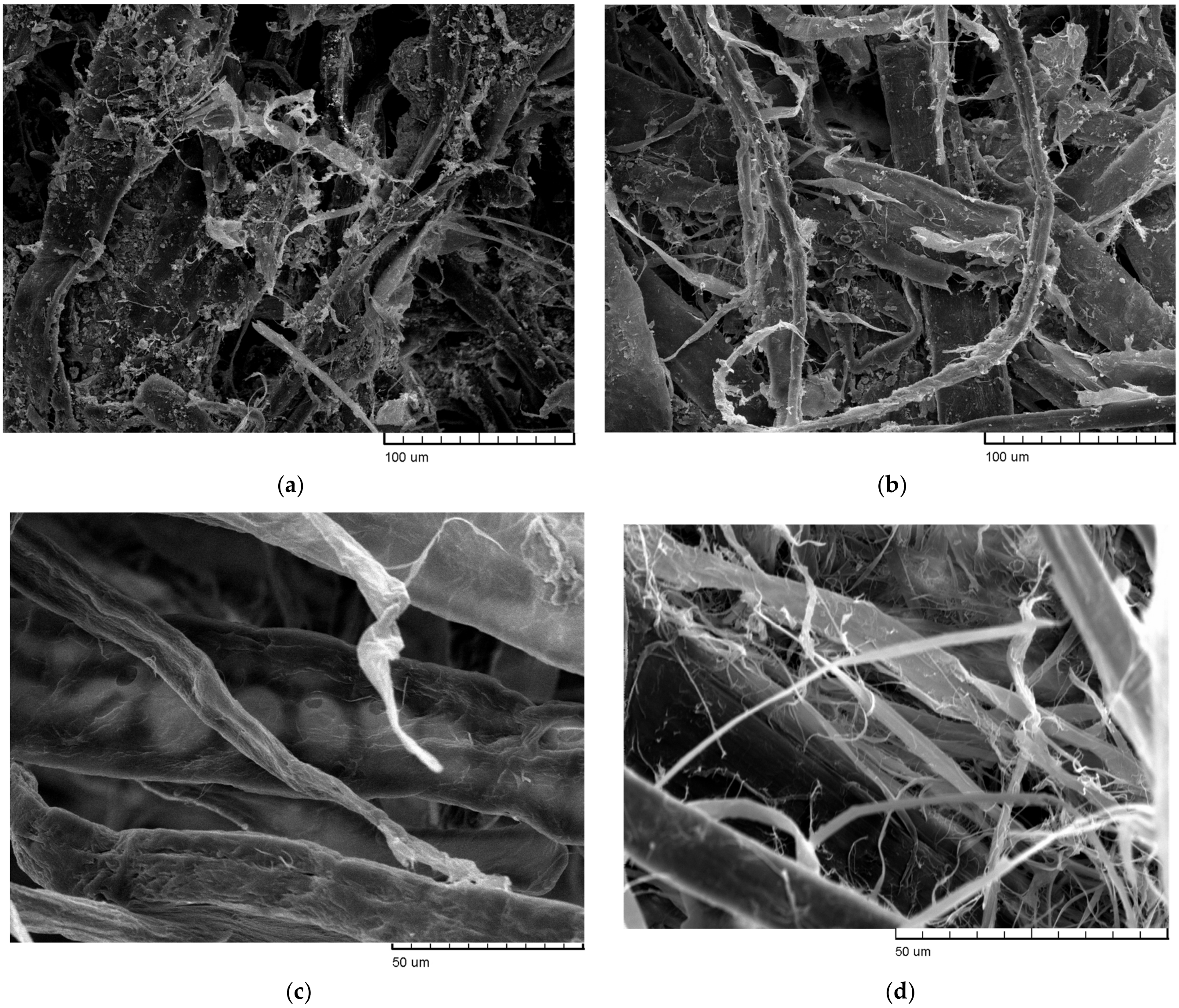

Figure 2. Scanning electron microscopy of fibers used for paper compositions: (**a**) Waste fibers WF, 1000×; (**b**) Waste fibers WFc, 1000×; (**c**) Kraft fibers KF, 2000×; (**d**) Hemp fibers HF, 3000×.

Despite the average size, hemp fibers have great potential for improving paper properties because they fibrillate well, resulting in a larger surface area than wood pulp fibers.

3.2. Characterisation of Fiber Blends' Mechanical Properties

Created fiber combinations were tested for tensile index Nm/g, stretch %, and breaking length km both in the dry and wet state; however, as shown in Figure 4, the differences among samples in the wet state were rather insignificant. Due to the above-mentioned fact, as well as the fact that the values were extremely low, the wet mechanical test results were not further used to investigate and compare the effect of fiber additives and CNF additives on paper properties. It is known that paper strength is based on the bonding strength of fibers; however, natural cellulose fibers without any wet strength agent are easily swelled by water, and the bonds are damaged, which causes poor wet strength [52]. To improve the mechanical performance of wet paper, wet strength agents are added, recently tending more toward natural ones, such as chitosan [53,54], polyelectrolyte complex of polysaccharides [55], cationized birch xylan [56], and other biobased substances.

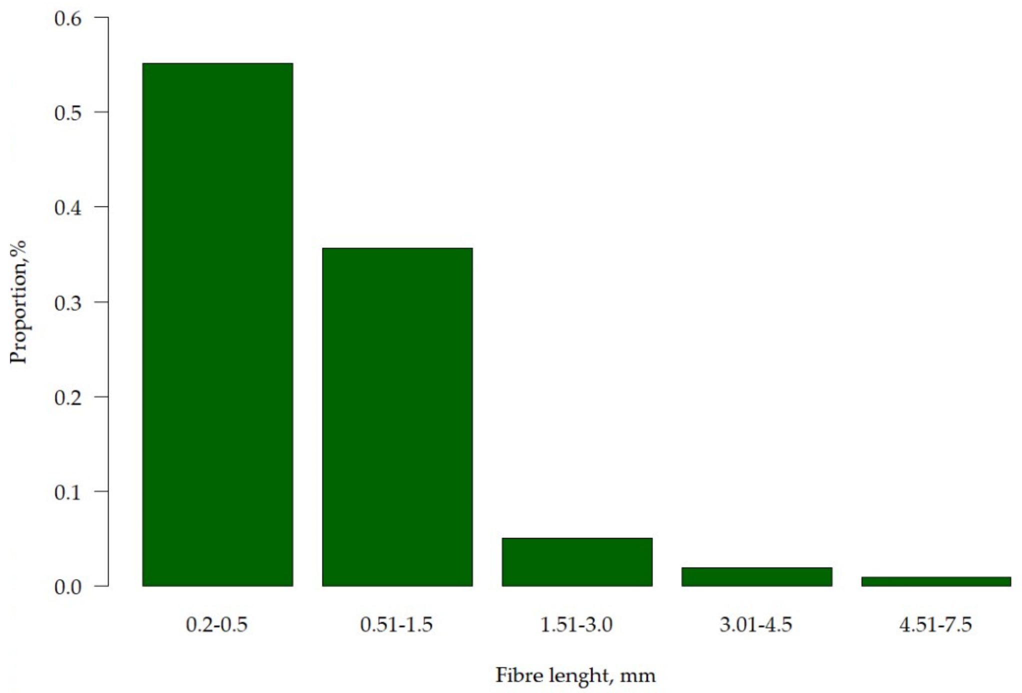

Figure 3. The proportional distribution of hemp fibers HF according to their length.

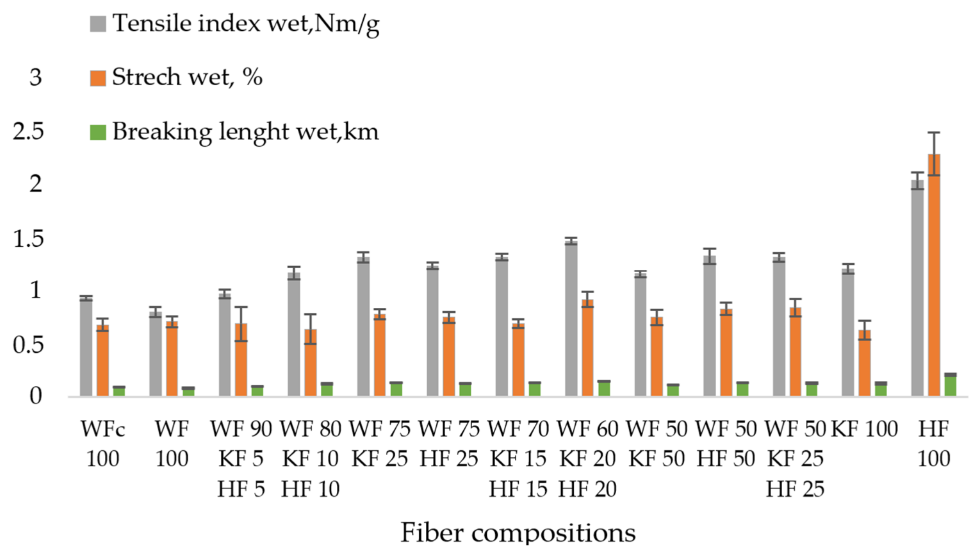

Figure 4. Mechanical properties of different fiber blends in wet state.

When comparing dry state mechanical properties such as burst index and tensile index, there were significant differences among the samples (Figure 5). The addition of 10% mixed virgin fibers (5% KF + 5% HF) increased the burst index by 34% and tensile index by 43%, if compared to paper consisting only of waste fibers. This result was comparable with an addition of 50% of KF to the waste fiber paper, creating the space for discussion on whether it is reasonable to add more single-blend virgin fibers or less mixed virgin fibers from different sources when creating new paper compositions. When 20% mixed virgin fibers (10% KF + 10% HF) were added, there was a 45% increase in burst and a 28% increase in tensile index, and the result was comparable with the addition of 25% KF. However, in the case of adding 25% virgin fibers to the recycled fibers, a very significant increase was demonstrated by HF presence, resulting in a 79% increase in burst index and a 78% increase in tensile index compared to the initial waste fiber material. The addition of an equal amount of KF increased the tensile index by 33% and the burst index by 49%, which is a significantly lower improvement if compared with HF. The identified excellence of HF over KF was also visible in the case of the replacement of 50% of waste fibers. The use of 50% HF improves mechanical performance almost twice (92% for burst index and 90% for tensile index), while 50% KF provided improvements of 27% and 22%, respectively. The addition of mixed fiber (25% KF + 25% HF) provided an 82% increase in burst and a 54% increase in tensile strength performance. The results allow us to conclude that HF or mixed fibers are better options for the improvement of recycled papers than KF. The superior properties of HF fibers are also shown as high results of HF100 papers (Figures 4 and 5), which reached a 2.29 kPa burst index and a 49.3 Nm/g tensile index. The performance of HF can be attributed to its size and structure, which differs from wood fiber by being thinner and shorter (Table 1). Different authors reflect on the diverse properties of hemp fibers. Highly fibrillated HF paper in a dry state reached a tensile index of 60.4 Nm/g with a fiber freeness of 91.5 °SR [57], while in other research, hemp hurd fibers were assessed as suitable for fiber blends and expected to have lower strength compared with typical wood pulp fibers [32]. Gumuskaya et al. [51] claim hemp fibers to be difficult to delignificate, resulting in poor papermaking and mechanical pulp properties, whereas Pal and Lucia [32] report that industrial hemp has favorable features as a pulp resource. The thinness characteristics of hemp hurd fibers enhance the penetration of pulping liquids; furthermore, weaker chemicals can be used for pulping and fibrillation. The excellent mechanical performance of HF in the current research can be attributed to the mechanical posttreatment to 18 °SR freeness, which apparently, has developed a sufficient fiber surface to create strong bonds with recycled and kraft fibers, resulting in papers with outstanding mechanical properties.

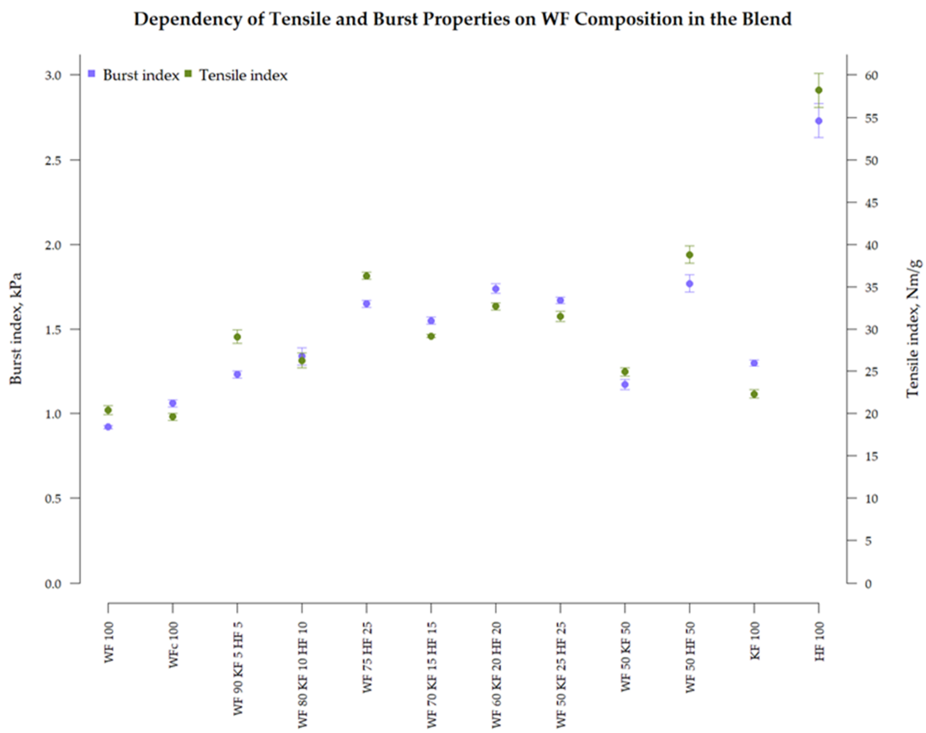

Figure 5. Mechanical properties of different fiber blends.

The minimal variation in stretch until rupture (0.8–1.2%) among the samples suggests a uniform performance within the materials under study (Figure 6). When sorted in ascending order of mechanical strength, the advantage of HF is even more convincing;

nevertheless, the stretch remains the same level as for other samples, containing both fiber combinations and single blend fibers.

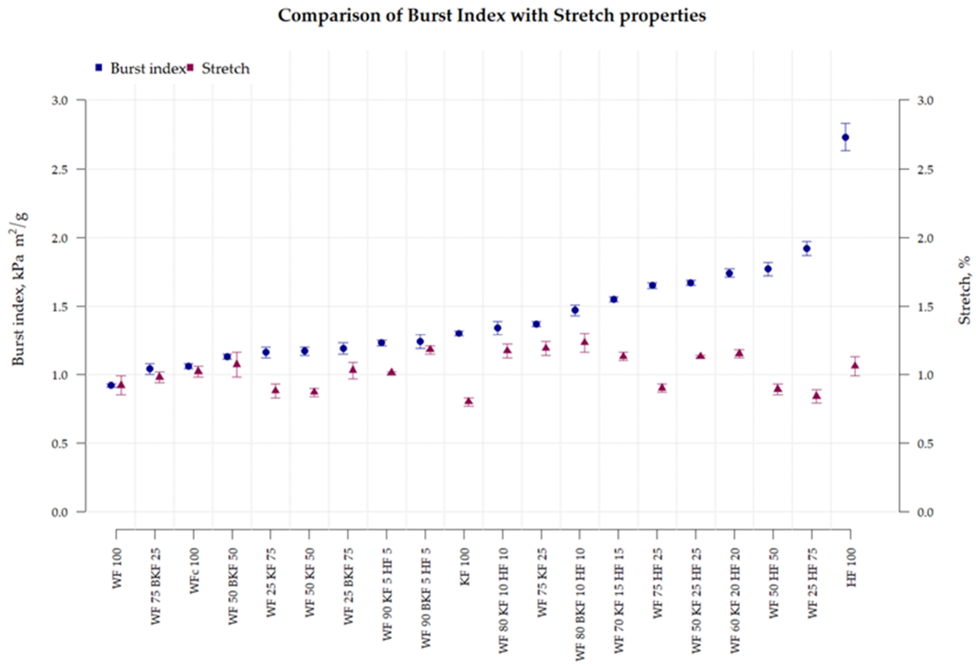

Figure 6. Comparison of burst index and stretch characteristics of fiber blends; samples are sorted in ascending order according to burst index.

3.3. *Impact of CNF Additive on Mechanical Properties of Fiber Materials*

Ammonium persulfate oxidated cellulose nanofibrils (CNF) were added to fiber blends. Figure 7 depicts fiber materials made of a single blend. The graph illustrates the tested tensile index Nm/g for the same fiber samples, with the addition of 1%, 3%, and 5% CNF.

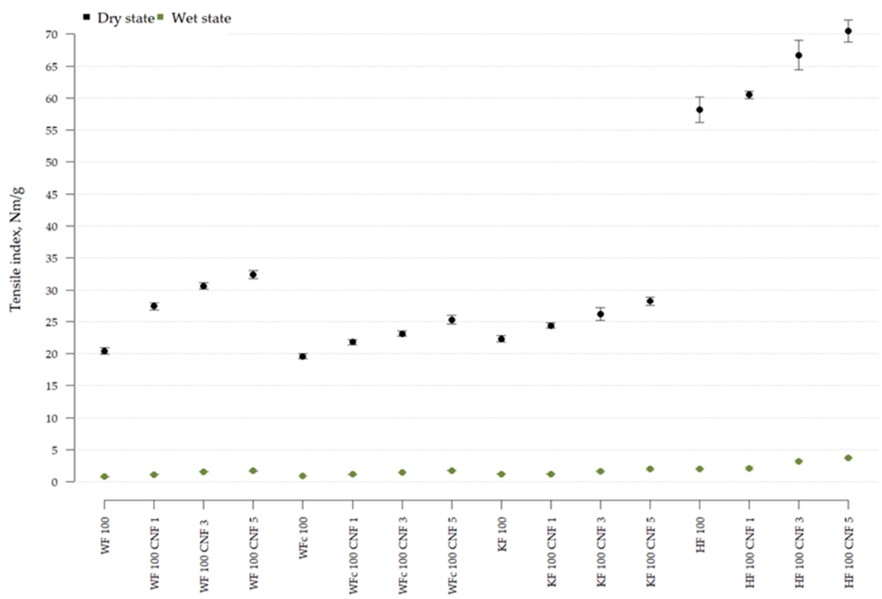

Figure 7. Tensile index of single blend fiber materials with CNF additive.

It can be seen that HF fiber paper was the most robust, and the application of CNF shows a linear increase in strength with a final increase of 21% compared to the starting value. Papers demonstrated various impacts of CNF additive on mechanical strength, starting from a 21% tensile increase for HF100 and reaching a 59% increase in tensile of WF100 after a 5% CNF addition. It can be concluded that fiber properties played a significant role in determining the impact of CNF. A more noticeable effect was observed in the increase in mechanical properties of HF-containing materials. It can be related to the increased surface of fibrillated hemp fibers, thus giving the place for more intense bonding with nanofibrils and other fibers in the blend. The impact of adding CNF to waste fiber materials varied depending on the batch. When it comes to WFc, the improvement in mechanical properties was relatively lower (30% increase after adding 5% CNF) than in the case of WF (59% increase after adding 5% CNF). This could be attributed to the composition of the recycled fibers and potential admixtures because the fundamental properties of the fibers themselves do not show significant differences (Table 1).

However, for a comprehensive understanding, the impact of CNF growth was also analyzed for fiber blend papers (Figure 8 and Table 2). In Figure 8, the samples are arranged according to the impact derived from the addition of CNF at a rate of 5%. The impact intensity of CNF addition is compared to the samples without CNF. The greatest increase in the tensile index is observed in samples in which the content of waste fibers comprises 50% of the material composition. However, it is evident that compositions containing 70% and 80% of waste fibers also yield commendable results.

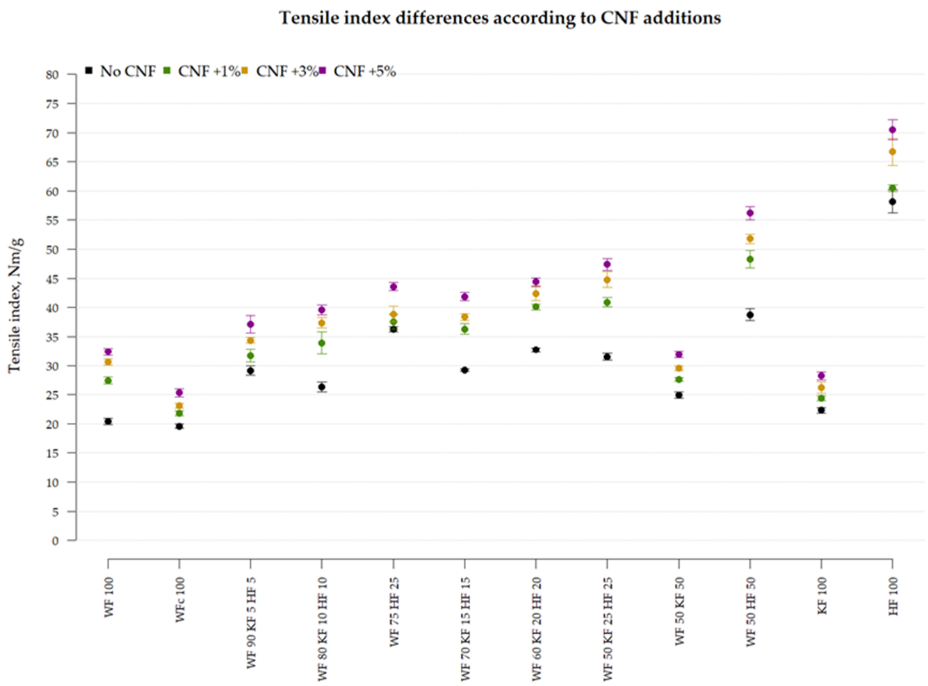

Figure 8. Tensile index of fiber blends with CNF additive.

Table 2 contains results regarding the influence of CNF addition on the tensile strength of fiber materials in their dry state. The analysis is conducted based on the average of five measurements, aiming to determine the relationship between the presence of CNF and the enhanced tensile strength in various materials. By examining the growth patterns of tensile strength concerning different concentrations of CNF additives, Pearson's product–moment correlation coefficient (PCC) between two continuous variables was estimated.

Table 2. Tensile index (TI) growth of fiber materials according to the CNF addition percentage growth.

Trial	1	2	3	4	PCC	Formula
Additive → Tensile Index, Nm/g ↘	No CNF	1% CNF	3% CNF	5% CNF		
WF 100	20.4	27.4	30.6	32.4	0.9571	TI = 17.9 + 3.92 × trial
WFc 100	19.6	21.8	23.1	25.3	0.9952	TI = 17.85 + 1.84 × trial
WF 90 KF 5 HF 5	29.1	31.7	34.4	37.1	0.9999	TI = 26.4 + 2.67 × trial
WF 80 KF 10 HF 10	26.3	33.9	37.4	39.6	0.9615	TI = 23.45 + 4.34 × trial
WF 75 HF 25	36.3	37.6	38.9	43.6	0.9412	TI = 33.3 + 2.32 × trial
WF 70 KF 15 HF 15	29.2	36.3	38.4	41.9	0.9688	TI = 26.4 + 4.02 × trial
WF 60 KF 20 HF 20	32.7	40.1	42.4	44.4	0.9446	TI = 30.55 + 3.74 × trial
WF 50 KF 50	24.9	27.6	29.5	31.9	0.9979	TI = 22.75 + 2.29 × trial
WF 50 KF 25 HF 25	31.5	40.9	44.8	47.4	0.9563	TI = 28.25 + 5.16 × trial
WF 50 HF 50	38.8	48.3	51.8	56.2	0.9725	TI = 34.85 + 5.57 × trial
KF 100	22.3	24.4	26.2	28.2	0.9996	TI = 20.4 + 1.95 × trial
HF 100	58.2	60.5	66.7	70.5	0.9865	TI = 53.2 + 4.31 × trial

According to the given formula, it is possible to calculate the subsequent CNF concentration's effect on the tensile index of the sample. This allows for the planning of fiber material blends according to the required material strength. Since the coefficients of the regression formula vary significantly (due to the differing intensity of CNF impact on various fiber compositions), they can be grouped to create a combined formula, simplifying the calculation process. The addition of CNF exhibits a linear impact on the tensile index. Such a trend can be formulated into a linear equation for predicting the tensile index. However, to ensure the sustainability of this trend, further tests with varying percentages of CNF additives should be conducted in subsequent studies, in order to understand whether the model maintains its linear nature at higher CNF proportions.

It is known that CNF contributes to the enhancement of paper properties through several mechanisms because of their high aspect ratio and large surface area, which cause increased bonding between fibers in the paper matrix. Furthermore, CNF acts like a filler, filling the gaps between bigger fibers [34]. This leads to improved mechanical strength, which has been demonstrated by the obtained results.

The obtained results align very well in the context of articles published by other authors. Hu et al. [10] improved the tensile index and burst index of recycled paper by 35.5% and 49.4% at a 5.0 wt% addition of CNF produced using different mechanical methods. Balea et al. [26] added 0.5% CNF to recycled paper and obtained a 20% increase in the tensile index of recycled paper. A significant increase in the tensile index by 95% was reached by adding 9% CNF to unrefined hardwood pulp handsheets [36]. Increasing the amount of CNF additive can be considered for further research on fiber blends; however, it is important to consider the economic aspects, as well, because CNF is still relatively expensive from an industrial perspective [34].

3.4. Characterisation of Fiber Blends' Air Permeability

Assessing fiber compositions without CNF, it is visible that fiber composition had a significant impact on the air permeability of paper (Figure 9). The highest results showed KF paper and compositions containing KF. Most likely, this effect was caused by the lack of refining resulting in a non-developed fiber surface after the kraft pulping process and the lowest content of fines (Table 1); as a result, a loose fiber net was formed. The lowest air permeability results had HF-containing paper. As concluded in Section 3.2, the increased hemp fiber surface was the main factor responsible for HF-containing paper properties, as well as for low air permeability as a result of intensive bonding between fibers. A higher content of fines could be the reason for lower results in the case of WFc compared with WF.

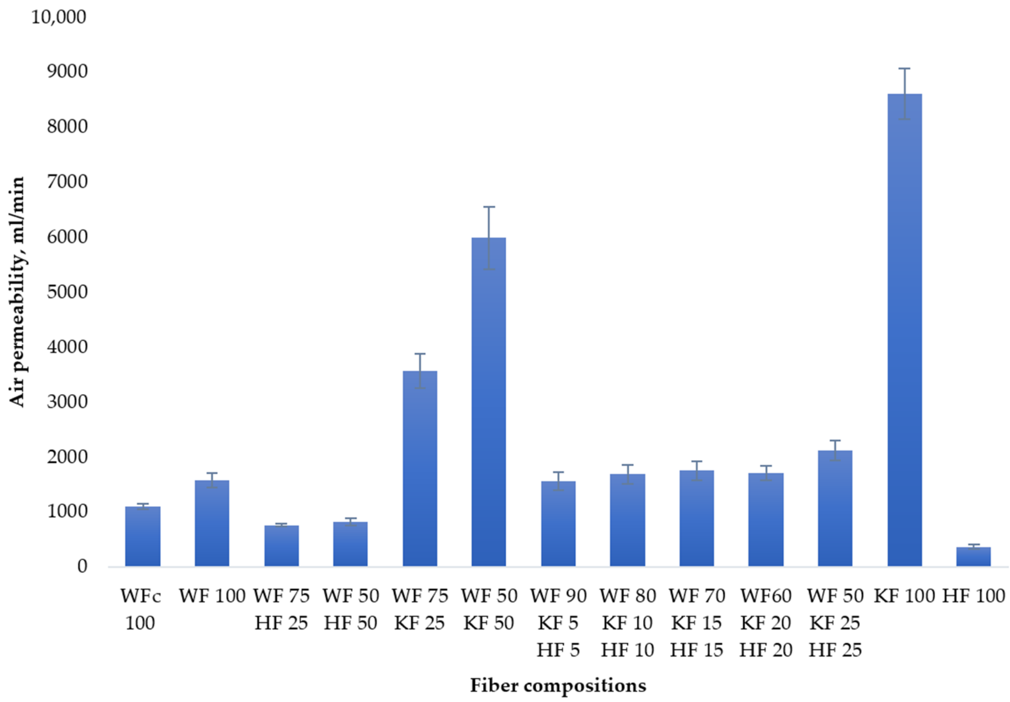

Figure 9. Air permeability of different fiber compositions.

When searching for the correlation between mechanical properties and air permeability among all investigated compositions, including with added CNF, it is evident (Figure 10) that at higher tensile indexes, there is a tendency for air permeability to decrease. Such behavior is typical for porous fiber materials and reflects the fiber bonding ability and efficiency, which is more intense when CNF is added. The data trend exhibits a logarithmic behavior, allowing for the prediction of material property alterations based on composition changes and the proportion of added CNF.

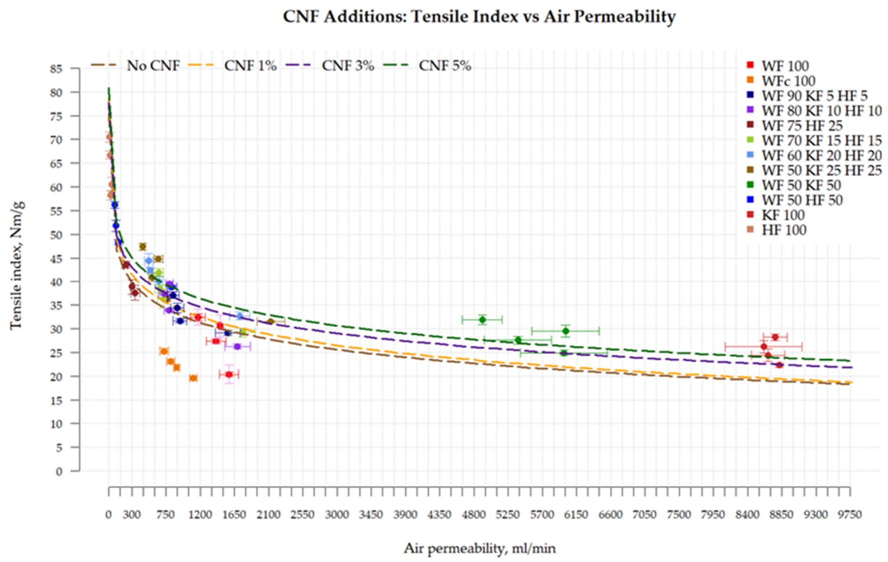

Figure 10. Tensile index vs. air permeability.

The impact of CNF addition on the air permeability differed among fiber compositions. At the highest CNF rate of 5%, air permeability decreased by 46–79% if HF were present in the material. For paper consisting of WF and WFc fibers, air permeability decreased, reaching 35% and 26%, respectively, while it only decreased by 9–18% if the composition consisted of WF and KF; furthermore, the increase in KF proportion decreased the impact of CNF. The interaction of CNF depends on the fiber properties. In the case of a more developed surface that HF had or the presence of the significant amount of fines that the WF and HF had, greater changes were caused in the structure of the paper when CNF was added through the filling of the gaps between fibers and the formation of more interfibrillar bonds that reduce the porosity of the material. However, KF apparently did not have a significant available area for bonding with other fibers and CFN. The ability of nanofibers to reduce the porosity of paper is well-known [58,59], and the performance depends on its source and production method [34]. Other authors report the decreasing porosity or air permeability after CFN addition: Balea et al. [26] reached a reduction of approximately 80%, while Hu et al. [10] reduced the porosity by 81–85% with the addition of 5% CNF.

3.5. Biodegradability

Because developed fiber materials were intended for packaging applications, the biodegradation test was organized as a trial of the disintegration of packaging material in a pilot-scale aerobic condition, according to adapted EN 14045:2003 [46]. The test included a 12-week composting procedure with weekly inspections of the sample and weighting residual nondegraded particles of material at the end of the test. As could be expected, all the papers were degraded during the test, in week 4 and week 5. Figure 11 shows the degradation progress of sample WF 50 KF 50, which was randomly selected for the representation of all inspected fiber samples in the manuscript because all fiber combinations showed similar biodegradation results. According to the results (no residual sample was detected after a 12-week test), all materials were recognized as fully biodegradable and home-compostable. The test can be considered equivalent to home composting because the temperature varied from 22 °C to 24 °C and moisture varied from 70 to 77% in the compost media during the test.

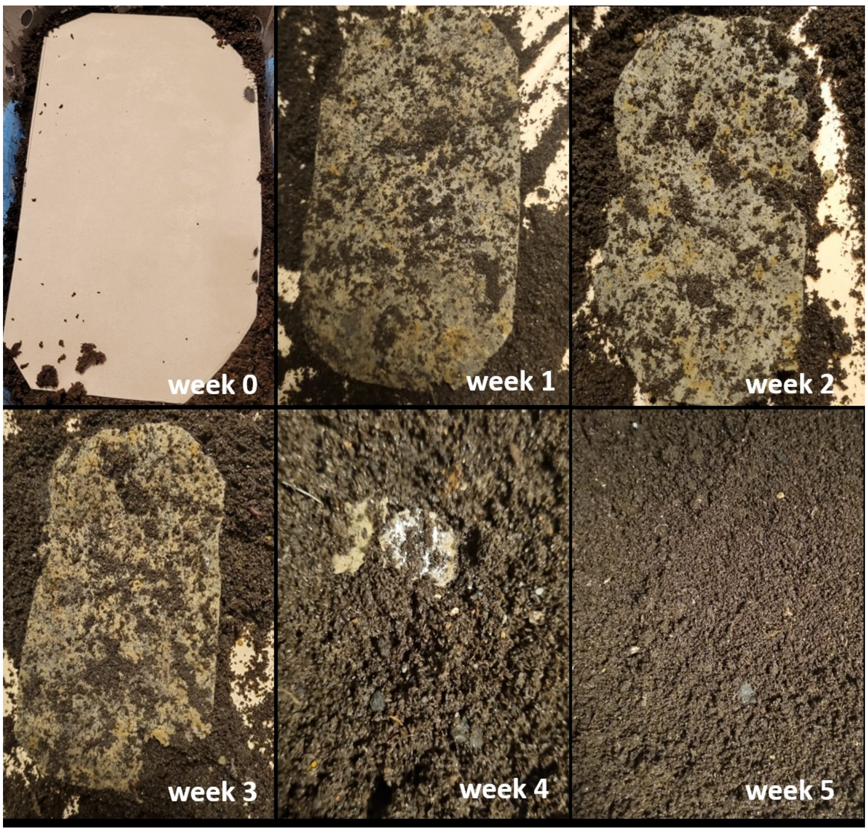

Figure 11. Weekly biodegradation progress of paper sample WF 50 KF 50.

The observations and results of tests carried out in the presented study were in accordance with the conclusions of Venelampi et al. [60], who determined the full biodegradation of paper samples consisting of recycled fiber, mechanical pulp, and bleached chemical pulp. The degradation of paper depends on the origin of fibers—those with lower lignin content (e.g., chemical pulp) degrade faster [60]. Overall, the biodegradation test results were predictable because the investigated materials consisted of natural fibers and biopolymers. The only precaution was the partly minor composition of waste fibers because unknown paper additives can cause slower disintegration or affect the quality of the compost [60]. Fast disintegration was also observed for materials containing waste fibers; however, harmful effects, e.g., toxicity, have not been investigated so far, leaving room for further research, especially considering the potential use of developed materials for food packaging. The recycled pulp can contain the remnants of pigments, fillers, dyes, and other chemicals of the paper industry, which could be the source of toxic metals [61], so the relevant investigation is a prerequisite before the development of the product. Another important safety indicator that should be investigated prior to approving the product as safe for the consumer is microbiology-related properties. Cellulose-based fibers are natural and welcoming substrates for microbial colonization, especially in appropriate humidity and temperature conditions, which was proved using a biodegradation test. However, to enhance antimicrobial activity against pathogens during the period of product use, antimicrobial agents should be added. Chitosan or chitosan derivatives [62] can be used from natural sources or synthetic biocides to ensure the antimicrobial properties of packaging produced from recycled fibers. However, testing and ensuring antibacterial properties is another prerequisite and should be involved as an important step in product development.

4. Conclusions

The hypothesis of the research about the possible significant improvement of recycled paper by using virgin wood and hemp fibers in different combinations was confirmed. Several combinations of waste fibers, hemp soda pulp, and softwood kraft pulp improved by 1%, 3%, and 5% CNF were investigated. Research has shown that combining virgin fibers from different sources improved the properties of recycled paper more than when a single type of fiber additive was used. A 10% mixed virgin fiber (hemp + wood) additive allows for the replacement of 50% wood kraft fiber additive, achieving equivalent mechanical strength indicators. A 50% hemp fiber addition to recycled fibers showed the best results, both for increasing mechanical properties up to a tensile index of 56 Nm/g and reducing the air permeability of recycled paper; furthermore, the addition of CNF increases the performance to an even greater extent. Research also highlighted the challenge of the recycled fiber industry in terms of raw material heterogeneity. Obtained fiber material samples showed suitability for home composting, thus, contributing to the goals of the European Green Deal regarding reducing landfill waste and the development of cleaner products.

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