Herein, cellulose nanofibers (PO-CNFS) and cellulose nanocrystals (PO-CNCs) are prepared directly from native softwood spruce and hardwood beech via the same alkaline periodate oxidation under equal conditions. PO-CNFS and PO-CNCs are obtained by simply regulating the reaction time between 2 and 7 d. Of particular note, the preparation of CNFs is achieved using the alkaline periodate oxidation. PO-CNFS obtained from spruce and beech have average diameters of 7.2 ± 3.1 and 6.8 ± 3.0 nm as well as average lengths of 487.1 ± 69.0 and 452.1 ± 80.0 nm, respectively. The PO-CNCs obtained from spruce and beech are 4.6 ± 2.0 and 4.8 ± 2.5 nm in diameter as well as 181.6 ± 49.4 and 168.4 ± 59.0 nm in length, respectively. These nanocelluloses contain carboxy groups that enable good colloidal stability of nanocelluloses in water. The nanocellulose content isolated from spruce and beech increase with the reaction time and reach 1.35 ± 0.20 and 1.24 ± 0.10 mmol g⁻¹ after oxidation for 7 d, respectively. Both PO-CNFS and PO-CNCs can be used for the formation of highly transparent films with good mechanical properties.

1. Introduction

Biobased nanomaterials derived from renewable biomass, for example, nanocelluloses, have drawn growing attention over the last three decades. Nanocelluloses can be extracted from plants, several marine animals, and so forth. Due to the different aspect ratios (length to diameter ratio, L/D), nanocelluloses include generally two main categories, cellulose nanocrystals (CNCs) with native crystalline cellulose and cellulose nanofibrils (CNF) containing both crystalline and amorphous regions. CNCs usually have average diameters of 3–10 nm and aspect ratios of 5–50, while CNFs generally have diameters of 5–30 nm and aspect ratio over 50. As biobased nanomaterials, CNCs and CNFs combine the properties of cellulose with the advantages of nanoparticles as reported in recent investigations, such as the ideal biocompatibility and nontoxicity, large availability due to the inexhaustible origin of raw materials, excellent mechanical properties with very high elastic modulus, high aspect ratio showing specific anisotropic properties, abundant surface groups, the feasibility of forming specific chiral structures in aqueous suspensions, tunable surface chemistry, and amphiphilicity. Therefore, they are emerging as sustainable and important candidates in many application scenarios.

Previous works on the method for preparation of nanocelluloses have made extraordinary progresses, including mechanical refining, chemical acid hydrolysis, oxidation method, and so on. In these methods, mechanical refining generally consumes more energy to separate the cellulose-contained resource into nanocelluloses. On the other hand, the isolation via chemical acid hydrolysis, or oxidation which include the 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO)-mediated oxidation and ammonium persulfate (APS) oxidation can reduce the energy consumption by 20 to 30 times for the preparation of the same amount of nanocelluloses. Generally, CNFs with dimensions of 3–7 nm in diameter and 500–2000 nm in length can be prepared via TEMPO-mediated oxidation (TEMPO/NaClO/NaClO₂ with pH 4–7 and TEMPO/NaBr/NaClO with pH 10). CNCs with the dimensions of 3–5 nm in diameter and 50–300 nm in length can be obtained via the methods of acid hydrolysis, APS oxidation, and TEMPO-mediated oxidation (TEMPO/NaClO/NaClO₂ with pH 4.8). Among these, CNFs and CNCs can be prepared by TEMPO-mediated oxidation method, however, it has to be assisted a homogenizer with high energy consumption (such as 300 W for 5 min). It is also worth noting that the intense mechanical treatment is generally required after aforementioned acid hydrolysis and oxidation. As well, the starting materials for the isolation of nanocelluloses must be pretreated to separate the cellulose from the native matrix, e.g., softwood spruce and hardwood beech. Currently, it is highly desired to develop a facile method to prepare CNCs and CNFs via a one single method with low energy consumption and without big change of the method, when dealing with the possible complex market needs of CNFs and CNCs in the future.

Here, we report a facile method to produce nanocelluloses by oxidizing native softwood spruce (SW) and hardwood beech (HW) via alkaline periodate oxidation (PO-nanocelluloses refers to both PO-CNFS and PO-CNCs from the alkaline periodate oxidation). The modification on the chemical structure of the...
native wood by alkaline periodate oxidation was investigated. The morphology was characterized with transmission electron micrograph (TEM) measurements. Moreover, the films of PO-CNCs and PO-CNFs were prepared by filtration and were subjected to characterizations regarding optical and mechanical properties.

2. Results and Discussion

2.1. Preparation of PO-CNFs and PO-CNCs by the Alkaline Periodate Oxidation

As described in Figure 1a, alkaline periodate oxidation at pH 10 was performed for the isolation of CNFs and CNCs. In general, 1.0 g of dried native wood specimens (SW and HW) with the dimensions of 35.0 mm × 10.0 mm × 1.0 mm (L × W × H) were soaked in 3.0 wt% KOH for 24 h as pretreatment and then were oxidized by an excess of 10.0 g of orthoperiodic acid (H₅IO₆). The pH value of reaction solution was adjusted to 10.0 with an aqueous KOH solution. Specifically, the reaction mixture was stirred for a certain time with the exclusion of light to prevent the formation of radicals. After separation and purification, the oxidized native wood suspension was treated by ultrasonication shortly at 96 W for 5 min. After the oxidation, 10.8 ± 0.7 wt% and 15.5 ± 0.5 wt% of PO-CNFs after 3 d, 15.3 ± 0.5 wt% and 18.5 ± 0.6 wt% of PO-CNCs after 7 d were obtained from spruce and beech, respectively.

To elucidate the isolation process of PO-CNCs and PO-CNFs, the native wood specimens in the reaction solutions were monitored for up to 7 d (Figure 1b). A dark brown color of wood was faded to lightly brownish products (the stable supernatant of suspension after standing still for 24 h) after extended reaction durations to 4–5 d. The color change indicates gradual decreasing contents of lignin in wood specimens and their dissolution in the solutions during the proceeding alkaline periodate oxidation (Figure 1b). After 5 d oxidation for spruce and 4 d for beech wood, these wood specimens were totally torn in pieces and suspended in the solutions. As well, the products became gradually colorless with prolonged reaction time of 7 d.

For both wood species, the alkaline periodate oxidation of less than 2 d could not generate sufficient amounts of nanocellulose as the stable suspensions after 24 h standing-still, while abundant big particles as solid fraction were still present (Figure 1b). With the reaction time of more than 2 d, the yields of recovered total solid after the oxidation as well as the yields of the solid fraction in the stable suspensions increased significantly. Their development was also monitored for up to 7 d, where almost all recovered solid represent the nanocellulose (Figure 1c). It should be noted that the solid total refers to the solid fraction from the centrifugation of the reaction solution at 14 000 rpm and thus containing both nanocellulose as well as bigger fragments.

With the reaction time extending from 2 to 7 d, the yield of total solid from the spruce decreased from 54 ± 1 wt% after 2 d to 19 ± 2 wt% after 7 d. At the same time, the yield of obtained PO–nanocelluloses rose from 10 ± 1 wt% after 2 d to 15 ± 0.5 wt% after 7 d. When beech specimens were used for the isolation of PO–nanocelluloses with alkaline periodate oxidation, the yield of total solids was 40 ± 2 wt% after 2 d, 19 ± 6 wt% after 6 d, and 19 ± 7 wt% after 7 d. In parallel, the yield of obtained PO–nanocelluloses was 15 ± 1 wt% after 2 d, 17 ± 0.3 wt% after 6 d, and 19 ± 0.6 wt% after 7 d. It is obvious that the most rapid reduction of solid content happened within the first 2 d. Nearly 30 wt% of spruce or 60 wt% of beech were converted into soluble products. In contrast, the cellulose part containing PO–nanocelluloses mainly remained within the first 2 d. Prolonged reactions on both wood specimens from 3 to 7 d slightly increased the generation of PO–nanocelluloses. The strong decrease of the total solid at the beginning can be attributed to the rapid removal of major parts of lignin, hemicellulose, and the amorphous regions of cellulose due to the improved accessibility. In comparison, further oxidation of the remaining solid fraction after 2 d isolation was strongly hampered due to the low reaction probability of alkaline periodate.
oxidation on highly ordered cellulose. Thus, the yield of PO–nanocelluloses increased only slightly after 2 d.

2.2. Morphology and Dimensions of Nanocelluloses after Various Oxidation Times

The solid fraction in the supernatant of suspension after standing still for 24 h was characterized by TEM measurements. The morphology of products from the alkaline periodate oxidation of spruce and beech were nanofiber or rod-like nanoparticles (Figure 2). The representative morphologies of spruce specimens after 3, 5, and 7 d reaction, as well as those of beech specimens after 3, 4, and 7 d reaction are displayed. The solid fraction from 5 d (spruce) and 4 d (beech) reaction were selected due to complete dissolution of the wood specimens (Figure 1b).

According to the general definition, the aspect ratios \((L/D)\) of CNCs and CNFs are 5–50 and over 50, respectively. As shown in Figure 2a,d, CNFs were obtained from spruce and beech after 3 d oxidation. The lengths of these PO-CNFs from spruce and beech specimens are 487.1 ± 69.0 and 452.1 ± 80.0 nm, respectively. Their average diameters are about 7 nm and aspect ratios \((L/D)\) are over 50. After 7 d oxidation on spruce (Figure 2c) and beech (Figure 2f), rod-like CNCs were generated. The average diameters of PO-CNCs from spruce and beech are 181.6 ± 49.4 and 168.4 ± 59.0 nm, respectively. Moreover, their diameters lie between 3 and 10 nm and aspect ratios \((L/D)\) are between 5 and 50. Therefore, CNFs were obtained after 3 d alkaline periodate oxidation on spruce and beech, while CNCs after 7 d. As the intermediate state of the alkaline periodate oxidation, obtained solid fractions from spruce 5d (Figure 2b) and beech 4d (Figure 2e) contained both PO-CNCs and PO-CNPs. Hence, the production of these two kinds of nanocelluloses with distinct aspect ratios via one single method was achieved. It is rarely reported that nanocelluloses with tunable lengths can be prepared only by varying the reaction time under otherwise equal conditions. Moreover, a novel approach for the preparation of CNFs was presented, which demonstrates at the same time the other feature of alkaline periodate oxidation.

2.3. Chemical Composition and Surface Chemistry of Nanocelluloses

The main chemical composition of the nanocelluloses from the spruce and beech were verified by ATR-FTIR spectroscopy (Figure 3a,b). The typical characteristic signals of cellulose were found in the FTIR spectra of the PO–nanocelluloses. For instance, the absorbance peaks at around 3340 cm\(^{-1}\) (O–H stretching vibration), 2900 cm\(^{-1}\) (C–H symmetrical stretching), 1420 cm\(^{-1}\) (C–H bending vibration), 1161 cm\(^{-1}\) (C–O–C stretching vibration) and 1057 cm\(^{-1}\) (C–O–C ring stretching vibration) were the main characteristic peaks of nanocelluloses.

Moreover, low-intensity signals of lignin appeared in FTIR spectra, such as the peaks at 1595–1598 and 1504–1511 cm\(^{-1}\) (C–C stretching of the aromatic ring), and around 1270 cm\(^{-1}\) (C–O aryl group of lignin). Moreover, the presence of carboxyl groups was also confirmed by the absorption band at about 1600 cm\(^{-1}\) (asymmetric carboxylate stretching vibration). The amount of carboxyl groups in nanocelluloses were determined by the conductivity titration. After 3 d alkaline periodate oxidation, 1.12 ± 0.12 and 1.09 ± 0.09 mmol g\(^{-1}\) of the carboxyl groups were detected in the PO-CNFs isolated from spruce and beech, respectively. With the prolonged oxidation time, the content of the carboxyl groups increased slightly to 1.25 ± 0.11 mmol g\(^{-1}\) after 5 d reaction on spruce and 1.16 ± 0.07 mmol g\(^{-1}\) after 4 d reaction on beech. After 7 d reaction, 1.35 ± 0.20 mmol g\(^{-1}\) (spruce) and 1.24 ± 0.10 mmol g\(^{-1}\) (beech) of the carboxyl groups were detected in the PO-CNCs. This is consistent with previous findings that alkaline periodate oxidation can introduce carboxyl groups on nanocellulose surface and improve its dispersibility. In contrast, slightly higher amounts of carboxyl groups were introduced in the present
work, and they are similar to the content of carboxyl groups in CNCs (1.4 mmol g\(^{-1}\))\(^{[23]}\) and CNFs (1.29 mmol g\(^{-1}\))\(^{[24]}\) prepared from the TEMPO-mediated oxidation method.

These carboxyl groups are expected to facilitate the isolation and stabilization of PO–nanocelluloses by breaking the interfibrillar hydrogen bonding and introducing sufficient electrostatic repulsive forces. As shown in Figure 3d, zeta-potential values of PO–nanocelluloses isolated from spruce and beech were raised from \(-27.6 \pm 1.3\) and \(-28.1 \pm 1.7\) (mV) for PO-CNFs to \(-31.9 \pm 1.6\) (mV) and \(-38.4 \pm 1.4\) (mV) for PO-CNCs, respectively. The apparent higher zeta potentials of PO-CNCs than those of PO-CNFs are consistent with the fact that more carboxyl groups were found on PO-CNCs.

2.4. Films of Various PO–Nanocelluloses

Highly transparent films were obtained with PO–nanocelluloses via the vacuum-filtration method (Figure 4a). The transparency of the films which is an important characteristic for the potential applications of green electronics or packaging was detected with UV–Vis spectrophotometry. As observed in Figure 4a, transmittances of approximately 45% and 80% in the wavelength range of 600 to 800 nm are quite similar for the films of the neat PO-CNF (spruce and beech 3d) and PO-CNC (spruce and beech 7d). Globally, the optical transmittance of the PO-CNCs films was higher than the PO-CNFs films. Especially, the transmittance of PO-CNCs films was over 75% at the wavelengths of more than 600 nm and the transmittance of PO-CNFs films less than 50.0%. This can be attributed to the smaller and more uniform size distribution of PO-CNCs. According to the Rayleigh's scattering theory,\(^{[25]}\) the scattering cross-section is directly proportional to the fiber diameter. With the larger diameter of nanocelluloses in the films, the amount of scattered light will increase significantly, which result in lower optical transmittance.\(^{[26]}\) In addition, a much lower content of adsorbed lignin and hemicelluloses in PO-CNCs can be another reason. With the pretreatment by soaking in 3.0%
Elongation at break [%] 7.6
Young's modulus [GPa] 2.2

aqueous KOH solution and the alkaline periodate oxidation for further 7 d, most of lignin was removed from the solid fraction as the soluble compounds.[27] The resulting suspensions after 7 d of reaction are almost colorless compared to those after 3 d of reaction (Figure 1b), which also leads to only very low amount of adsorbed color compounds in PO-CNCs and thus lower absorption in UV–Vis wavelength range.

As shown in Figure 4b and Table 1, the mechanical properties of PO-CNFs and PO-CNCs films were investigated by the static tensile strain–stress measurements. The tensile strength of PO-CNFs (spruce and beech 3d) and PO-CNCs (spruce and beech 7d) films were averagely 84–108 and 131–147 MPa, respectively. In comparison with some of previously reported nanocellulose films from CNF and CNC (61–275 MPa) the resultant PO–nanocelluloses films showed comparable mechanical properties.[28] The tensile strength and Young's modulus of PO–nanocelluloses films increased with the size reduction of nanocellulose. For instance, the tensile strength of the films of PO–nanocelluloses spruce 3d and spruce 7d were 84.0 ± 3.2 and 146.9 ± 2.1 MPa, respectively (Figure 4b and Table 1). In parallel, the elongation at break of PO–nanocelluloses films exhibited an opposite behavior. While the size of nanocelluloses became shorter during the oxidation process, the PO-CNCs films (spruce 7d and beech 7d) showed smaller values of elongation at break. For instance, the elongation at break of films of PO-CNCs (beech 7d) was 5.5 ± 0.1% compared to 7.6 ± 2.3% for films of PO-CNFs (beech 3d). The reasons for these different mechanical properties between PO-CNFs and PO-CNCs films should be their different microstructures. PO-CNFs contained both amorphous and crystalline regions, whereas most amorphous regions of cellulose were removed in PO-CNCs. The crystalline region has the highly ordered alignment and correspondingly higher Young's modulus than the amorphous regions. On the contrary, the amorphous regions in the PO-CNFs allow their films to sustain more elongation during the tensile testing. During the elongation process, these loose cellulose chains in the amorphous regions could be extended and rearranged, leading to the regeneration of intramolecular H-bonds and therefore higher elongation at break.[29]

### Table 1. Mechanical properties of the nanocellulose films.

| PO–nanocelluloses films prepared from native wood | Beech 3d | Beech 7d | Spruce 3d | Spruce 7d |
|-----------------------------------------------|---------|---------|----------|----------|
| Tensile strength [MPa]                        | 108.0 ± 2.6 | 131.0 ± 2.5 | 84.0 ± 3.2 | 146.9 ± 2.1 |
| Young's modulus [GPa]                         | 2.2 ± 0.3 | 4.8 ± 0.5 | 2.7 ± 0.2 | 3.8 ± 0.8 |
| Elongation at break [%]                       | 7.6 ± 2.3 | 5.5 ± 0.1 | 8.3 ± 1.9 | 4.6 ± 1.1 |

#### 3. Conclusion

In summary, PO-CNFs and PO-CNCs were directly isolated from the native hardwood and softwood via alkaline periodate oxidation. By varying the reaction time only, the production of nanocellulose as CNFs and CNCs was achieved by one single method without changing the reaction system. The individual PO-CNCs and PO-CNFs have the diameters of 5–7 nm and lengths of 200–500 nm, respectively. More than 1.0 mmol g⁻¹ of carboxyl groups was generated on both PO-CNCs and PO-CNFs. In addition, PO–nanocelluloses films are mechanically strong with high optical transparency.

#### 4. Experimental Section

**Materials and Chemicals:** Dried native wood samples (softwood spruce and hardwood beech) with a dimension of 35 mm × 10 mm × 1 mm were prepared in own workshop. Periodic acid (H₅IO₆), and potassium hydroxide (KOH) were bought from Sigma-Aldrich. All chemicals were of analytical grade or higher. Deionized water (DI water) was used throughout the experiments.

**Isolation of Nanocelluloses:** H₅IO₆ (10 g) was dissolved in DI water to give a 6% (w/w) solution and the pH of the solution was adjusted to 10 using aqueous KOH solution. Dried wood samples (1.0 g) were soaked in 3 wt% aqueous KOH solution for 24 h, and then dipped into the alkaline periodate solution. The reactions were performed at 40 °C with gentle stirring in dark. The reaction times were set as 2, 3, 4, 5, 6, and 7 d and the obtained samples are referred as 2d, 3d, 4d, 5d, 6d, and 7d. Immediately after the reaction, the solid parts (total solid) were collected via centrifugation on Thermo Scientific Multifuge X3 FR for 20 min at 4 °C with the rotation speed of 14 000 rpm. Then, the solid was redispersed in DI water and purified via dialysis in water using the dialysis membrane with a molecular weight cutoff of 3500 Da (Thermo Fisher Scientific). Thereafter, the oxidized native wood suspension was treated by ultrasonication (SONOPLUS ultrasonic homogenizer HD 4200) with the energy input of 96 W for 5 min. Remaining stable PO–nanocelluloses in suspensions after standing still for 24 h were used for yield calculation and further characterization.

**Preparation of PO–Nanocelluloses Films:** Typically, 20 mL of 0.1 wt% nanocellulose suspension was vacuum filtered on a polypivalidene fluoride filter membrane with the diameter of 47 mm and pore size of 0.22 µm (GVWP, Millipore). When a wet gel-cake was formed, no flow of free water was observed, and the apparatus was tilted slightly, the vacuum-assisted filtration was finished which was typically completed within 30 min. Then, the gel cakes were carefully transferred to a glass Petri dish and allowed to dry in the air at room temperature further.

**Measurement of Carboxyl Group Content:** The quantity of carboxyl groups was measured by conductometric titration of the PO–nanocelluloses suspensions as reported previously.[19] Zeta-Potential Measurement: Zeta-potential was measured using a Zetasizer Nano ZS (Malvern Instruments Ltd., UK) with 1 mL of PO–nanocelluloses suspension of 0.1% (w/w) in disposable folded capillary cell.

**FTIR Measurement:** FTIR spectra of samples were obtained on an Alpha FTIR Spectrometer (Bruker, Germany) with a versatile high throughput ZnSe ATR crystal. All samples were measured within the wave number range of 800 to 4000 cm⁻¹ with a resolution of 4 cm⁻¹ and accumulated 24 scans.

**Transmission Electron Microscopy:** A droplet (7 µL) of 0.01 wt% PO-nanocellulose suspension was deposited onto glow-discharged carbon-coated TEM grid. The excess liquid was removed by blotting with a filter paper to leave a thin suspension layer. After ambient drying, another droplet (7 µL) of 2 wt% phosphotungstic acid solution (the pH value was adjusted to 7.0 using 1 M aqueous NaOH solution) was deposited on the TEM grid to negatively stain the sample. The TEM observation was performed on a CM 12 Transmission Electron Microscope (Philips, Netherlands). The diameters and lengths of PO–nanocelluloses were measured and calculated using Nano Measure software.

**Ultraviolet–Visible Spectrophotometry (UV–Vis):** The transmission of the films was measured on a UV–Vis spectrophotometer of SPECORD S 600 at wavelengths of 400 nm to 800 nm.

**Mechanical Properties of PO–Nanocelluloses Films:** The tensile testing of PO–nanocelluloses films was performed using a Z3 microtensile test machine equipped with a 50.0 N load cell from Grip-Engineering Thümler GmbH (Germany). The crosshead speed is 2.5 mm min⁻¹. The
nominal tensile stress and strain were recorded for the calculation of the ultimate tensile strength and elongation at break.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
Research data are not shared.

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