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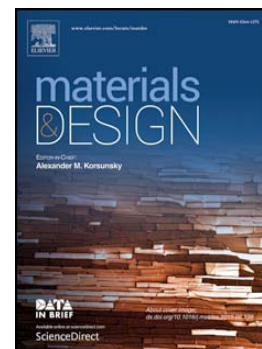
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Cellulose binders for electric double-layer capacitor electrodes: The influence of cellulose quality on electrical properties

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Abstract

Cellulose derivatives are widely used as binders and dispersing agents in different applications. Binders composed of cellulose are an environmentally friendly alternative to oil-based polymer binding agents. Previously, we reported the use of cellulose nanofibers (CNFs) as binders in electrodes for electric double-layer capacitors (EDLCs). In addition to good mechanical stability, we demonstrated that CNFs enhanced the electrical performance of the electrodes. However, cellulose fibers can cover a broad range of length scales, and the quality requirements from an electrode perspective have not been thoroughly investigated. To evaluate the influence of fiber quality on electrode properties, we tested seven samples with different fiber dimensions that are based on the same kraft pulp. To capture the length scale from fibers to nanofibrils, we evaluated the performance of the untreated kraft pulp, refined fibers, microfibrillated cellulose (MFC) and CNFs.

Electrodes with kraft pulp or refined fibers showed the lowest electrical resistivity. The specific capacitances of all EDLCs were surprisingly similar, but slightly lower for the EDLC with CNFs. The same electrode sample with CNFs also showed a slightly higher equivalent series resistance (ESR), compared to those of the other EDLCs. Graphite dispersions with MFC showed the best dispersion stability.

Keywords: nanocomposite, nanocellulose, cellulose, graphite, supercapacitor, electric double-layer capacitor

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1. Introduction

To support the commercialization of sustainable energy applications, the production of inexpensive, environmentally friendly energy storage devices, such as electric double-layer capacitors (EDLCs), is required [1]. Currently, EDLCs are available in various sizes, capacitances and operating voltages [2]. Unfortunately, commercial EDLCs usually contain expensive and toxic components [3, 4]; however, these materials can be replaced with inexpensive and environmentally friendly materials. Previously, we suggested the use of aqueous electrolytes and electrodes with graphite as the active material and cellulose nanofibers (CNFs) as the binding agent [5, 6]. A binder must be added to the active material to form mechanically stable electrodes. Commercial EDLCs often contain polytetrafluoroethylene (PTFE), polyvinyl alcohol (PVA) or carboxymethyl cellulose as the binder [7]. However, these binders are electrically nonconductive and degrade the electrical performance of the electrodes. We have demonstrated that CNFs enhance both the mechanical and electrical properties of EDLC electrodes [5]. CNFs can be derived from wood fibers and are an environmentally friendly alternative to oil-based polymer binders [1, 8, 9]. We showed that CNFs form a network that holds the graphite particles together without covering and clogging the porous electrode structure [10]. The use of CNFs in energy devices is gaining popularity because they are renewable materials with many outstanding properties [1, 11, 12, 13, 14, 15]. CNFs are expensive because they are produced by a chemical pretreatment and extensive mechanical processing; thus, a low-cost alternative is desired. Another drawback of CNFs is their high water retention ability, which complicates the dewatering and the processability of dispersions containing them.

In this study, we determine the most suitable binder for EDLC electrodes among kraft pulp fibers, refined fibers, microfibrillated cellulose (MFC) or CNFs. Kraft pulp fibers, refined fibers and MFC are more cost-effective than CNFs because they are less mechanically processed. Kraft pulp is not further refined or chemically treated and is therefore the most cost-effective alternative. In contrast, CNFs are the most expensive because they are the most refined and chemically treated fibers in this study. To evaluate the electrical properties of electrodes that contain cellulose with these qualities, we measured the electrical resistivity of the electrodes and the capacitance and equivalent series resistance (ESR) of the EDLCs. Furthermore, we performed a sedimentation study to evaluate the dispersion stability of the different graphite-cellulose dispersions.

2. Materials and Methods

2.1. Treatment of cellulose fibers

Cellulose with different qualities was used as the binder in the electrodes. Table 1 lists the cellulose samples and the corresponding preparation processes. All cellulose qualities are based on the same kraft pulp, which is flash-dried pulp from pine and spruce supplied by SCA Forest Products AB, Sweden. One sample, called the kraft pulp sample (also called the untreated sample in this

study), was used and analyzed without any further pretreatment. The untreated sample was already slightly refined in the mill to SR 60, which was compared to completely unrefined pulp that is typically approximately SR 15, where SR (Schopper-Riegler) refers to an index value indicating the amount of mechanical treatment to which the pulp has been subjected. The samples PFI 5000 and PFI 10000 were mechanically beaten using a PFI mill (Papirindustriens forskningsinstitut mill) and were refined for 5000 and 10000 revolutions, respectively, according to ISO 5264-2. The samples marked as TEMPO (2,2,6,6-tetramethylpiperidine) and TEMPO Super-fine were subjected to a chemical pretreatment (TEMPO-mediated oxidation) [16] followed by a mechanical delamination treatment. The chemical and mechanical treatments were performed as follows. The pulp was washed and dewatered. Pulp corresponding to a dry weight of 100 g was mixed with 4 l deionized water to a concentration of 2.5 %. The dispersion was stirred continuously. Then, 20.6 g NaBr (2 mmol g^{-1} dry pulp) and 3.2 g TEMPO (0.2 mmol g^{-1} dry pulp) were added under constant stirring. The dispersion was stirred for 1 hour and left undisturbed for approximately 16 hours. A volume of 792 ml NaClO with a concentration of 14 % (15 mmol g^{-1} fibers) was adjusted with HCl to pH 9.5. The pH-adjusted NaClO was slowly added to the pulp suspension. The pH of the suspension was adjusted to 9.5 by adding 1 M NaOH. When all the NaClO was added and the pH was stabilized, the pulp suspension was dewatered, and then, it was washed with 15 l of deionized water. A suspension with a dry content of 2 % was prepared and dispersed using an Ultra Turrax T50 and a dispersing element S50N-G45F (both from IKA-Werke GmbH & CO. KG) for 150 minutes at 8100 rpm. Samples were obtained after 30, 90 and 150 minutes, corresponding to the samples named TEMPO 30, TEMPO 90 and TEMPO 150. To obtain the sample TEMPO Super-fine, 500 ml of sample TEMPO 150 was diluted with deionized water to a concentration of 0.1 %. The suspension was dispersed for an additional 15 minutes at 14 000 rpm using the Ultra Turrax T25 and dispersing element S25N-25F. The dispersion (200 ml) was sonicated for 20 minutes using a probe sonicator (Vibra Cell, High Intensity Ultrasonic Processor, Sonics & Materials INC., 750 W, 20 kHz, amplitude: 50 %, interval: 0.8 s pulse/0.2 s pause). The sample was centrifuged at 6500 rpm (4700 G) for 1 hour to separate the larger fiber fragments from the CNFs. The supernatant was concentrated with a rotary evaporator at 25 °C to obtain a gel-like CNF suspension.

2.2. Fiber characterization

The total charge of the untreated cellulose and the TEMPO-oxidized cellulose was measured by conductometric titration. The TEMPO-oxidized cellulose was analyzed after the washing step but before the fibrillation with the disperser (Ultra Turrax). The fiber dimensions and the crill of the untreated and refined samples were measured using a PulpEye Analyser (PulpEye, SCA R&D Centre AB, Sundsvall, Sweden). The crill measurement is a technique for qualitatively assessing the fibrillation degree of the pulp, including both defibrillated fibrils and fibrils resting on larger fibers. The method is based on the optical response from both UV and IR wavelengths of light, where the lower detection limit is

100 nm [17, 18]. Approximately 2.5 g dry solids were used for the fiber dimension measurement, and 1 g of dry solids was used for the crill measurements, according to ISO 16065-2.

2.3. Electrode preparation and characterization

Electrodes were prepared by mixing 2.67 g graphite (grade 2299 from Cummings-Moore Graphite) with 0.267 g cellulose. Milli-Q water was added to the mixture to obtain a dispersion with a volume of 300 ml. The dispersion was mixed for 20 min at 8400 rpm with an Ultra Turrax T25 and a dispersing element S25N-25F (both from IKA-Werke GmbH & CO. KG). Afterwards, 50 ml of the dispersion was filtrated through Millipore Durapore Membrane Filters (Merck KGaA, filter type: 0.22 GV, diameter: 90 mm) using a vacuum filtration funnel. The electrode films were dried at room temperature. Electrodes with a thickness between 200 μm and 240 μm were obtained.

2.3.1. Sedimentation study.

To test the stability of the dispersions, test tubes were filled with the freshly prepared graphite dispersions. The height of the resulting clear phase of each sample was measured after certain time intervals. A reference sample with the same amount of graphite but without any cellulose was prepared and measured.

2.3.2. Scanning electron microscope images.

High-quality cross-sections were prepared using a Hitachi IM4000 Ion Milling System. Surface and cross-sectional images of the electrodes were obtained using a field emission scanning electron microscope (FESEM; MAIA3, TESCAN). Secondary electron images were generated using a 3 kV accelerating voltage and a 5 mm working distance.

2.3.3. Electrical resistivity.

To evaluate the electrical properties of the electrodes, the electrical resistivity was measured using a source meter (2611A SYSTEM, KEITHLEY). The resistivity was measured at 10 different locations on the electrode, and the arithmetic mean was calculated.

2.4. Electric double layer capacitor design and measurements

EDLCs were assembled in a test cell by stacking electrodes measuring 3 cm \times 3 cm between two current collectors. The current collectors were composed of platinized titanium foil (Goodfellow; thickness: 0.35 mm, purity: 99.6 %). Grease-proof paper was used as the separator, and 1 M Na₂SO₄ served as the electrolyte. A weight (1.66 kg) was placed on top of the test cell to secure the cell stack and to ensure a good contact between the cell components.

2.4.1. Galvanostatic cycling.

Galvanostatic cycling (GC) was performed with a charge and discharge current of 150 mA using a LabVIEW-based PXI system. After 20 hours of continuous cycling, the specific capacitance and the ESR were determined. The specific capacitance was calculated using the discharge curve according to

$$C = I \cdot \frac{dt}{dV}, \quad (1)$$

where I is the discharge current, dt is the discharge time and dV is the cell potential difference. The specific capacitance C_{sp} was calculated by

$$C_{sp} = 4 \cdot \frac{C}{m}, \quad (2)$$

where m is the mass of active material of both electrodes. The ESR was determined from the voltage drop by

$$\text{ESR} = \frac{dV}{dI}. \quad (3)$$

2.4.2. Cyclic voltammetry.

Cyclic voltammetry (CV) was performed using a potentiostat/galvanostat VersaSTAT 4 from Princeton Applied Research between 0 V and 1 V. Scan rates of 10 mV s^{-1} , 50 mV s^{-1} and 100 mV s^{-1} were applied. The CV measurements were performed after the GC tests. The samples were cycled five times at each scan rate.

3. Results and Discussion

3.1. Fiber properties

The total charge of the untreated fibers, as well as the refined fibers (PFI 5000, PFI 10000), was $82 \mu\text{eq g}^{-1}$. The TEMPO-oxidized fibers (TEMPO 30, TEMPO 90, TEMPO 150, TEMPO Super-fine) obtained a total charge of $1273 \mu\text{eq g}^{-1}$. A high degree of total charge due to TEMPO-oxidation is known to facilitate the degree of fibrillation in the mechanical treatment [19]. After the chemical treatment, the mechanical treatment was applied for different durations to obtain a range of qualities of cellulose fibrils, from a broad particle size distribution of the TEMPO 30 sample to the most narrow distribution and lowest size for the super-fine TEMPO sample. Figure 1 reveals that the super-fine TEMPO sample is well fibrillated, containing only thin nanofibrils. The TEMPO 30 sample retains many large-scale fibers and is the coarsest of the TEMPO-oxidized samples.

Table 2 shows that the fibers were shortened from $1.43 \mu\text{m}$ to $0.74 \mu\text{m}$ during the milling process, while the fiber thickness was maintained. The PFI mill is the most commonly used laboratory refiner, providing high-energy and low-intensity refining, which results in internal fibrillation [20]. Large amounts of

fine material and fibrils were produced during the milling process according to the significant increase in the crill value. The fiber dimensions and crill values reveal that there is a significant difference in the degree of fibrillation between the coarser samples.

3.2. Dispersion stability

The dispersion stability was evaluated by performing a sedimentation study. Figure 2 shows the samples 31 days after their preparation. The image reveals that the samples TEMPO 30, TEMPO 90 and TEMPO 150 provided a significantly better stabilization of the dispersion than the other samples. Only a slight shift in color indicated a sedimentation of the graphite particles. The sample TEMPO Super-fine also sedimented over time. The slight gray color of the clear phase of this sample indicated that it retained small graphite particles. However, the sediment of the TEMPO-oxidized samples contained only small particles and no agglomerates or large clusters. The graphite dispersions with untreated or refined fibers clearly separated. The water phase on top of the sediment was clear and contained only a few large particles, which adhered to the walls of the test tubes. The sediments of these samples contained a large amount of graphite agglomerates and clusters.

Figure 3 presents the sedimentation progress. The height of the clear phase is plotted against the time elapsed since the mixing of the samples. A reference sample containing the same amount of graphite but no cellulose was also analyzed.

The TEMPO 30, TEMPO 90 and TEMPO 150 samples achieved the best dispersion stability. This finding can be explained by the highly charged anionic cellulose fibrils that stabilized the dispersion. Hajian et al. suggest that the ability of CNFs to stabilize carbon nanomaterials in water is caused by an association between the materials [21]. The associated materials are further stabilized by electrostatic repulsion due to the charges of the CNFs. In contrast, the untreated and refined samples did not stabilize the dispersions. The graphite particles in these samples sedimented directly after the mixing process, indicating that samples containing untreated or refined fibers cannot be stored and must be processed directly after mixing to obtain a uniform composite. Although the fast sedimentation speed limits the processability, dispersions with untreated or refined fibers can be dewatered rapidly. Fast dewatering is favorable because the processing time is reduced and the energy consumption of the electrode production process decreases. Dispersions containing MFC or CNFs dewater slowly; thus they require energy-intensive dewatering and drying steps. Because the TEMPO samples have a high specific surface area, a substantial amount of water binds to the surface. The water retention was shown to increase with fibrillation [22].

The TEMPO Super-fine sample showed a good performance during the first 4 days after the mixing process. Afterwards, the graphite particles in the sample sedimented, and a light gray liquid phase remained on top of the sediment, as shown in figure 2. This result indicates that the light gray phase contained small, well dispersed and separated particles. The samples TEMPO 90 and TEMPO

150 showed a similar behaviors. They appeared to be stable throughout the first two weeks, but started to sediment afterwards. From these data, we hypothesize that smaller nanofibrils stabilize the graphite particles, but that the distance between the particles decreases. This phenomenon might explain the smaller volume of the graphite dispersion and the formation of a sediment. This theory would also explain the observations described above in which the sediment of the TEMPO-oxidized samples does not contain any large graphite agglomerates or clusters. The results for the CNF dispersion further indicate that smaller fibrils might fail to stabilize all graphite particles. In a previous test, we observed that a larger amount of CNFs than MFC was required to stabilize the same mass of graphite particles.

3.3. Electrode properties

3.3.1. Electrical resistivity.

Figure 4 shows the electrical resistivity of the electrodes. Electrodes with TEMPO-oxidized CNFs showed a higher electrical resistivity than electrodes with non-treated or mechanically beaten fibers. Electrodes with refined fibers and the electrode with untreated fibers exhibited only minor differences in resistivity. The sample PFI 10000 showed the lowest electrical resistivity of all the tested electrodes. Only small changes in resistivity could be observed for the TEMPO samples that were dispersed for 30, 90 and 150 minutes. A significantly larger resistivity was observed for the electrode with CNFs (TEMPO Super-fine), which showed the highest electrical resistivity of all the electrodes. The scanning electron microscope (SEM) images show that the CNFs created a film-like structure around the graphite particles, and they most likely decreased the contact points between the conductive particles, thus affecting the resistivity (see figure 5). The non-treated fibers, the refined fibers and the MFC formed a web-like structure that only partly covered the graphite. Thus these fibers did not degrade the resistivity as much as the CNFs. From this point of view, rather coarse fibers than fine fibrils should be used to achieve a low electrode resistivity.

3.3.2. Electrode structure.

Figure 6 shows the SEM images of the electrode cross-sections for the samples TEMPO 30 (right) and PFI 10000 (left). The TEMPO-oxidized cellulose produced a uniform structure in the electrode, while the PFI-milled cellulose produced cracks and non-uniformity due to large-scale fibers. The cracks are detrimental to the EDLC performance and can increase the ESR and resistivity. The same structure can be seen in the SEM surface images shown in figure 7. The left image corresponds to the untreated fibers, and the right image is the TEMPO 30 sample.

Furthermore, we observed that the electrodes with untreated or refined fibers are more brittle than electrodes containing MFC or CNFs. This brittleness might cause a disruption in these electrodes during sample preparation and during assembly of EDLCs. Thus, we suggest using MFC or CNFs as the binder in EDLC electrodes.

3.4. Electric double-layer capacitor properties

3.4.1. Galvanostatic cycling.

The specific capacitance of each sample was calculated from the GC measurements. The results are plotted in figure 8. All EDLCs, except for that containing super-fine CNFs, exhibited similar specific capacitances in the range of 40.7 F g^{-1} to 43.7 F g^{-1} . The sample TEMPO Super-fine showed the lowest specific capacitance (35.75 F g^{-1}). The highest specific capacitance of 43.66 F g^{-1} was measured for TEMPO 30. The lower capacitance of the TEMPO Super-fine can be explained by the CNF film that partly covered the electrode surface, as shown in figure 5. The CNF film hindered the electrolyte transport to the electrode surface and into the pores of the electrode. Furthermore, the CNF film on the electrode surface resulted in a poor electrical contact between the electrodes and the current collectors, which led to a higher contact resistance. The CNF film also inhibited a good electrical contact between adjacent graphite particles, which explains the higher electrical resistivity reported in figure 4.

Furthermore, the ESR of the EDLCs was measured, and the results are displayed in figure 9. The sample TEMPO 30 exhibited the lowest ESR of 0.53Ω , and the sample TEMPO Super-fine showed the highest ESR of 0.80Ω . The untreated and refined samples resulted in ESR values of approximately 0.6Ω , and TEMPO 90 and TEMPO 150 showed values of approximately 0.75Ω . The high ESR of the TEMPO Super-fine EDLC is consistent with our previous findings from the SEM study and the capacitance measurements. These results confirm our theory that the CNFs created a film-like structure that hindered the ion transport to the electrode surface and deteriorated the electrical contact between the graphite particles as well as between the electrodes and the current collectors. Thus, the formation of the CNF film resulted in a higher electrode, electrolyte and contact resistance, thus resulting in a higher ESR.

3.4.2. Cyclic voltammetry.

Figure 10 shows the I-V curves from the CV measurements of the EDLCs. The devices were cycled at three different scan rates: 10 mV s^{-1} , 50 mV s^{-1} and 100 mV s^{-1} . The differences among the EDLCs were the shapes of the curves and the current plateaus, showing slightly higher capacitances (plot areas) for the TEMPO-oxidized samples than for the untreated and refined samples. This result was most evident at a scan rate of 10 mV s^{-1} . At higher scan rates, the curvature shifted substantially for all units.

The shapes of the curves of the CV measurements showed that chemical reactions may have occurred in addition to electrostatic charging and discharging; this was observed at all scan rates, but it was most pronounced at 10 mV s^{-1} . At this scan rate, higher currents were observed between 0 V and 0.3 V compared to the relatively flat current plateaus between 0.3 V and 0.9 V for both the charge and discharge regions. This behavior was most prominent for the TEMPO-oxidized samples at low scan rates and might have been caused by redox reactions of functional groups on the cellulose surface. Faradaic capacitance has been observed due to redox reactions of oxygenated groups on electrode materials [23]. These reactions are slow and can only be observed at low scan rates.

At 10 mV s^{-1} , an exponential increase was observed in the charge current between 0.9 V and 1 V, which could indicate electrolyte electrolysis.

Furthermore, the different curve shapes also indicated that the cell voltage required to obtain a certain charge current density was different among the samples (the bending distance differed), thus indicating a higher resistance to charge transfer (charge propagation) in the TEMPO-oxidized samples than in the untreated and refined samples. This trend was most evident at the scan rates of 50 mV s^{-1} and 100 mV s^{-1} and might be a result of the higher electrode resistance due to the CNF film that covered the electrode surface. The CNF film hinders the ions from accessing the electrode surface, which leads to longer ion diffusion times.

4. Conclusion

In summary, the electrical properties of graphite electrodes for EDLCs are only slightly influenced by the degree of fibrillation of the cellulose fibers. The finest cellulose quality in this study, the CNFs, was expected to achieve the best performance as a binder and dispersing agent. Our results, however, show that CNFs are not the best choice for electrodes, because they exhibit the highest electrical resistivity due to film formation of the CNFs on the electrode surface. Thus, EDLCs with CNFs also show the lowest specific capacitance and the highest ESR of all the tested samples. Therefore, the use of CNFs as a binder is not recommended. A better and more economical alternative is the use of electrodes with MFC, because of the superior electrical properties and because the graphite dispersions with MFC exhibited the longest shelf-life in this study. Although the ability to stabilize dispersions is favorable, MFC and CNFs have a high water retention ability, which requires energy-intensive dewatering to form electrodes. The most cost-efficient binders are kraft pulp or refined fibers; they show good electrical properties, and their dispersions can be easily dewatered. However, kraft pulp and refined fibers cannot stabilize water-based graphite dispersions. Therefore, these dispersions must be processed and dewatered directly after their preparation. In conclusion, the use of MFC as a binder in electrodes for EDLCs is recommended because it possesses the best electrical properties and dispersion stability and is relatively cost-effective.

Conflict of interest

There are no conflicts to declare.

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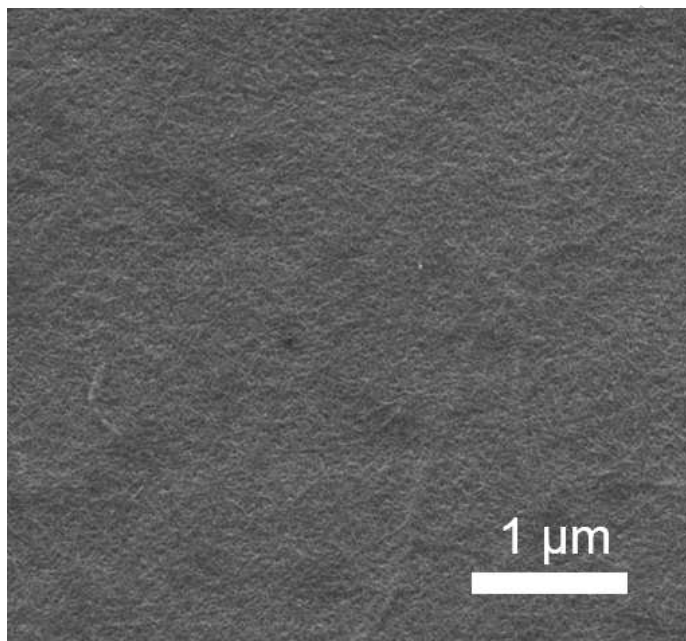


Figure 1: Scanning electron microscope image of the surface of the cellulose sample TEMPO Super-fine. Only thin nanofibrils and no large fibers can be observed, which confirms that the cellulose is well fibrillated.

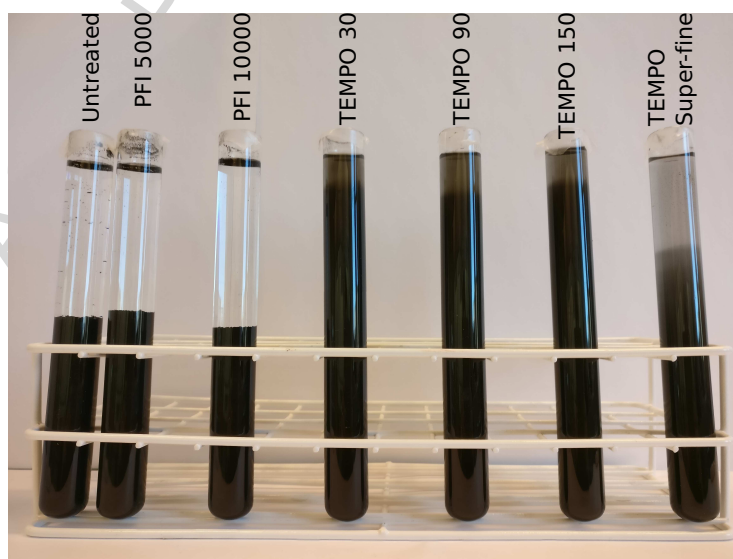


Figure 2: Sedimentation samples containing cellulose with different qualities 31 days after preparation. The TEMPO-oxidized samples provided a significantly better stabilization of the graphite dispersions than the samples with coarser qualities.

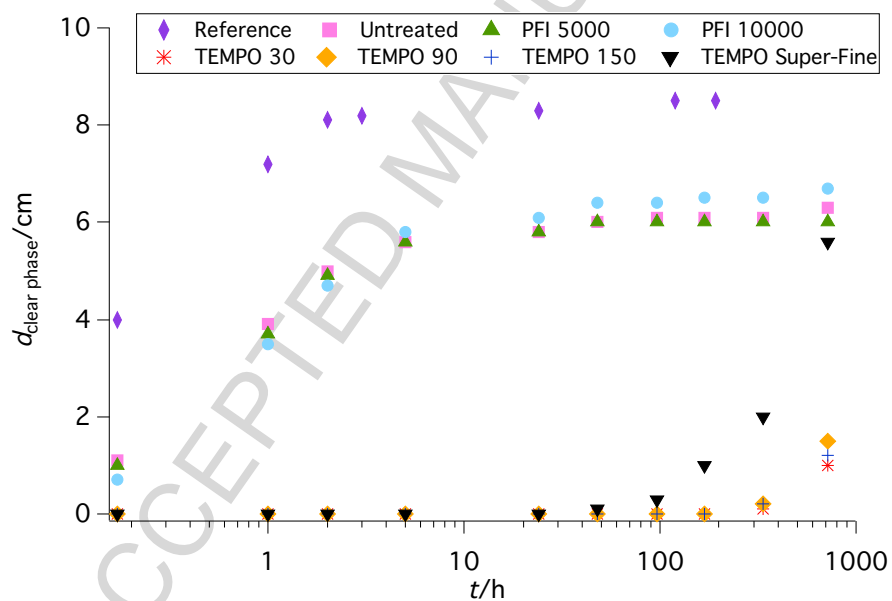


Figure 3: Sedimentation study of graphite dispersions containing cellulose with different qualities. A clear trend in stabilization ability was observed in the following order of good to poor stability: microfibrillated cellulose; cellulose nanofibers; refined/untreated cellulose.

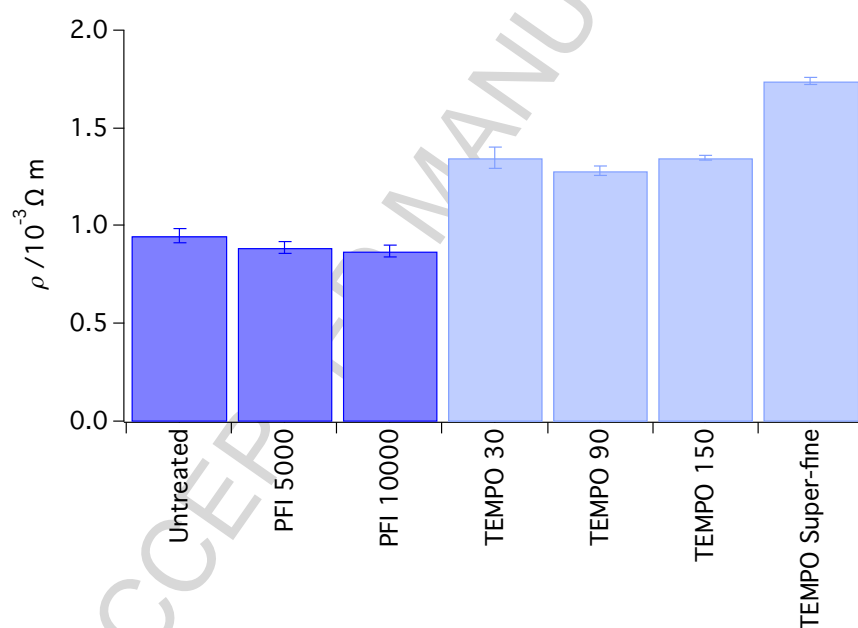


Figure 4: Electrical resistivity of electrode films containing varying grades of cellulose. A higher resistivity was observed for TEMPO-oxidized samples than for electrodes with untreated or refined fibers.

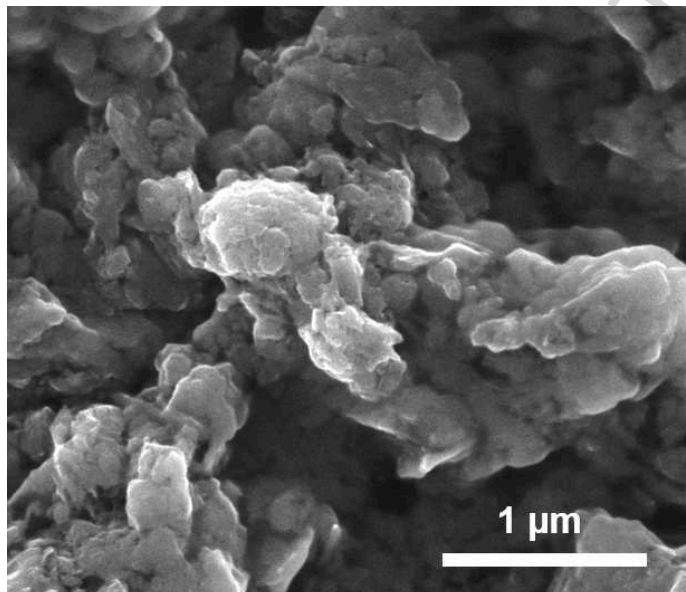


Figure 5: Scanning electron microscope image of the electrode containing the TEMPO Superfine cellulose. The cellulose nanofibers create a film-like structure around the graphite particles.

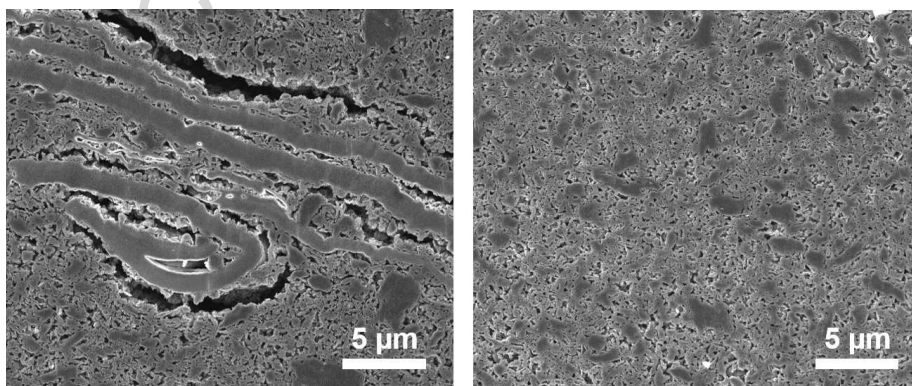


Figure 6: Cross-section scanning electron microscope images of the electrode containing PFI 10000 fibers (left) and TEMPO 30 cellulose (right). The PFI-milled cellulose induced cracks along the large fibers. A more uniform electrode structure was obtained with TEMPO-oxidized cellulose.

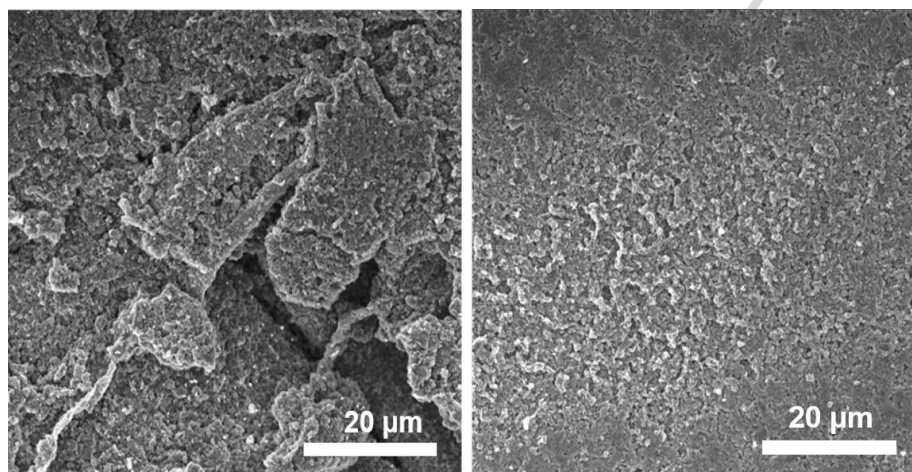


Figure 7: Scanning electron microscope images of the electrode surfaces of the untreated sample (left) and sample TEMPO 30 (right). The untreated sample had an uneven surface with many cracks. TEMPO 30 showed a uniform surface.

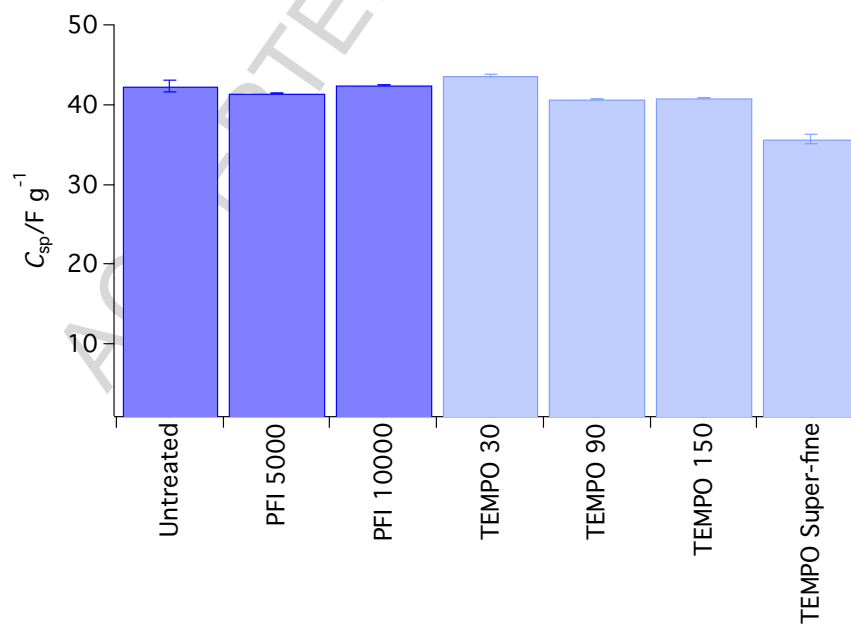


Figure 8: Specific capacitance of electric double-layer capacitors measured using galvanostatic cycling. All capacitors showed similar capacitances, except for sample TEMPO Super-fine, which had a slightly lower specific capacitance.

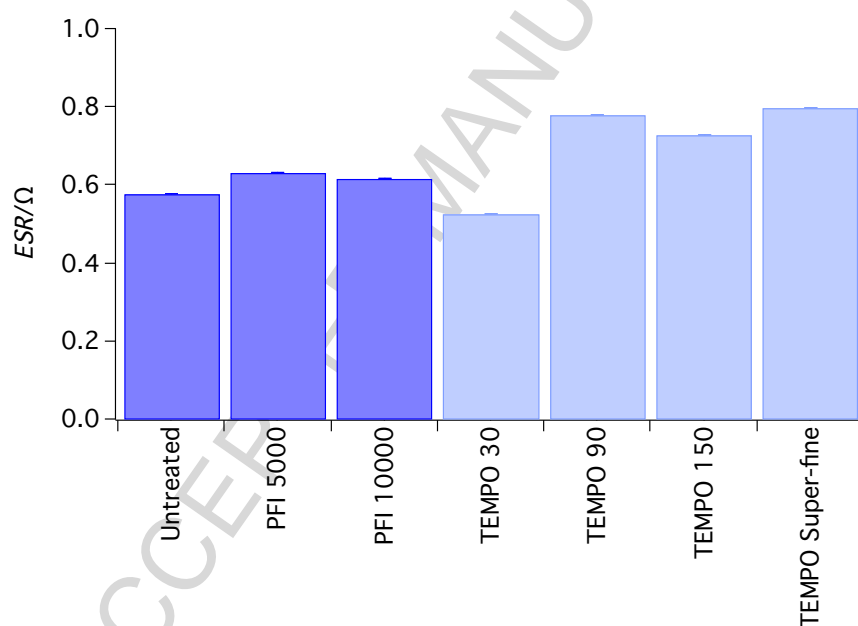


Figure 9: Equivalent series resistance of electric double-layer capacitors measured by galvanostatic cycling. The lowest resistance was obtained for TEMPO 30 and the highest was measured for TEMPO Super-fine.

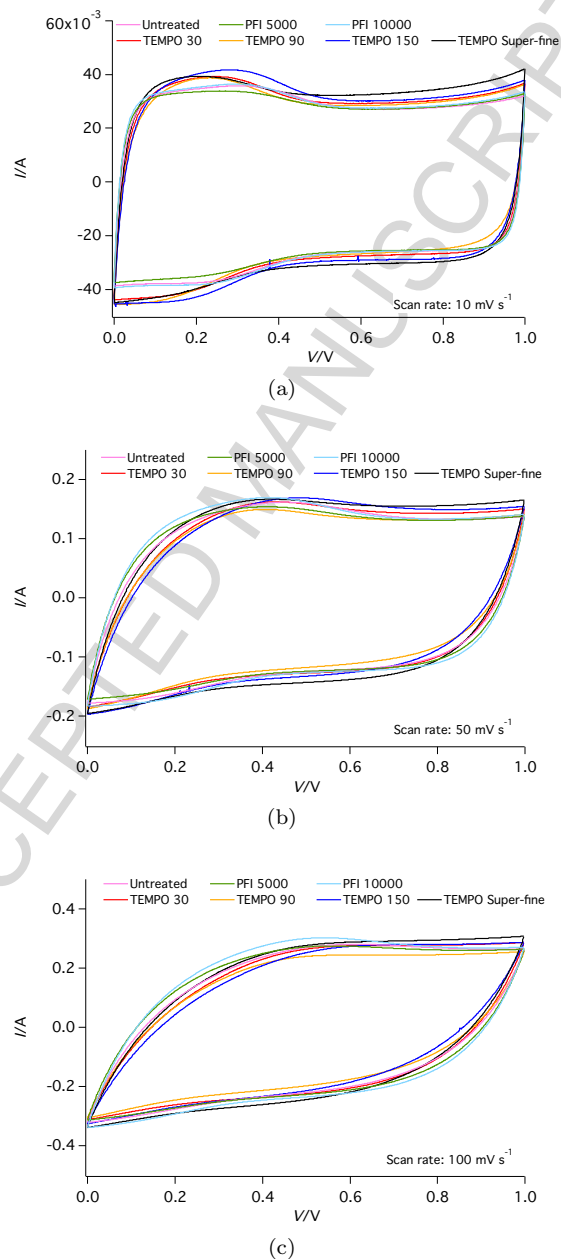


Figure 10: Cyclic voltammetry curves of electric double-layer capacitors with electrodes containing cellulose of different qualities. The cyclic voltammetry measurements were performed at scan rates of: a) 10 mV s⁻¹, b) 50 mV s⁻¹, and c) 100 mV s⁻¹.

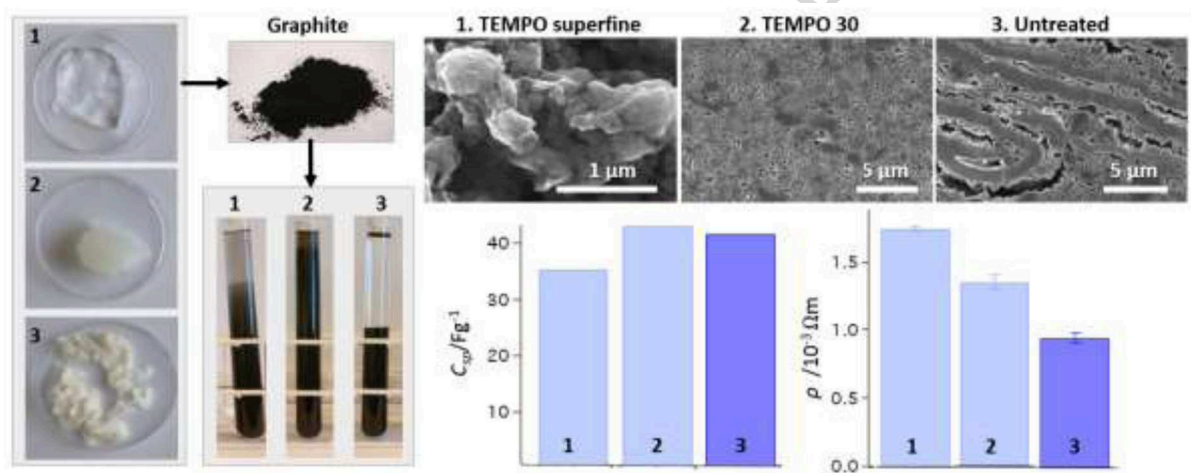
Table 1: Mechanical and chemical treatments of cellulose fibers and the corresponding sample names.

Sample	Refining (PFI milling)	Chemical pretreatment (TEMPO oxidation)	Dispersing (Ultra Turrax)	Purification (sonication+centrifugation)
Untreated	—	—	—	—
PFI 5000	✓(5000 ×)	—	—	—
PFI 10000	✓(10000 ×)	—	—	—
TEMPO 30	—	✓	✓(30 min)	—
TEMPO 90	—	✓	✓(90 min)	—
TEMPO 150	—	✓	✓(150 min)	—
TEMPO Super-fine	—	✓	✓(165 min)	✓

Table 2: Fiber dimensions and crill values of the untreated and refined cellulose samples. The length, width and crill values confirm the internal fibrillation due to refining.

Sample	Length/mm	Width/ μm	Crill
Untreated	1.43	30.0	209.9
PFI 5000	0.97	30.2	225.3
PFI 10000	0.74	30.8	236.1

Graphical abstract



Highlights

- The electrical properties of graphite electrodes with cellulose binders for electric double-layer capacitors are only slightly influenced by the degree of fibrillation of the cellulose fibers.
- Microfibrillated cellulose provides the best electrical properties and is a cost-efficient alternative to highly processed cellulose nanofibers.
- Microfibrillated cellulose further shows an excellent performance as dispersing agent in aqueous graphite dispersions.
- Untreated and refined fibers exhibit a good electrical performance as well, but fail to stabilize water-graphite dispersions.