Effect of Nanofibrillated Cellulose on Alginate and Chitosan Film Properties as Potential Barrier Coatings for Paper Food Packaging

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This study aimed to test the utility of ammonium persulfate (APS) oxidised nanofibrillated cellulose (NFC) as an additive for chitosan- and alginate-based biopolymer films that could eventually be used as paper coatings for food packaging applications. Sodium alginate and chitosan were used as the base for the films. Various concentrations of APS oxidised NFC ranging from 0% to 10% were used as a reinforcing agent, resulting in six combinations of either alginate-NFC or chitosan-NFC composite films. Biofilms were tested for their mechanical properties (tensile strength and strain), grease barrier properties, air permeability, water vapour permeability, and degradation in the soil. Overall, when using the ammonium persulfate oxidation pretreatment method, the best performance of the films was estimated with the addition of 2.5% NFC.

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INTRODUCTION

In 2021, every EU citizen generated an average of 35.85 kg of plastic packaging waste (Eurostat 2023), while only close to 40% of that packaging waste was recycled (41% in 2019 and 38% in 2020) (Eurostat 2021). Packaging accounts for roughly 1/3 of the global plastic use, making it the most significant plastics-consuming segment (Statista 2023). The pollution from plastic packaging that is not correctly disposed of nor properly recycled creates a substantial burden on the environment, and, in the case of plastic packaging for food, it may threaten human health (Jadhav et al. 2021). Therefore, there is an urgent need to replace fossil fuel-derived plastics with other, more sustainable alternatives. Packaging made of bio-based renewable polymers with non-toxic and biodegradable properties coming from aquatic environments or made of food and agricultural waste, and thus not requiring additional land use for their growth, could represent a good alternative to the current single-use plastic packaging.

Two types of polysaccharides – chitosan and sodium alginate – were chosen for this study. Due to their well-organized hydrogen bond network, films made of these polysaccharides have good oxygen barrier properties (Hassan et al. 2018) that are important for food packaging.
Chitosan is most often commercially produced from shellfish processing wastes; it is non-toxic and biodegradable and has good mechanical and oxygen barrier properties (Leceta et al. 2013; Sirviö et al. 2014; Wang et al. 2018). Sodium alginate is a polysaccharide derived from brown seaweed and, like chitosan, is biodegradable, non-toxic, and renewable (Abdel Aziz and Salama 2021; Carina et al. 2021). Its unique colloidal properties allow for the creation of transparent and uniform films (Abdel Aziz and Salama 2021; Carina et al. 2021; Zinina et al. 2023). Importantly, sodium alginate (E401) has been approved by the European Commission as a food additive (Food (ANS) 2017). However, films made solely of sodium alginate do not possess good mechanical properties (Mao et al. 2023; Zinina et al. 2023). Like chitosan, sodium alginate is hydrophilic, which implies weak moisture barrier properties of the films (Hassan et al. 2018; Mao et al. 2023). It has been shown that incorporating cellulose nanomaterials can improve the properties of the films (Yu et al. 2017; Shanmugam et al. 2019). Cellulose is one of the most abundant polysaccharides currently available, and nanofibrillated cellulose (NFC, also called cellulose nanofibril) can be isolated from wood and other plant sources using chemical and/or mechanical treatments to partially disrupt their natural structures (Lavrič et al. 2021). Ammonium persulfate (APS) has been used to produce carboxylated cellulose nanocrystals (CNC) since 2011, when it was first done by Leung et al. (2011). However, high concentrations of APS have been mainly used to release crystalline parts and to fully destroy amorphous parts of cellulose. The ability to gently oxidise cellulose using lower concentrations of APS for facilitated mechanical fibrillation has also been observed (Filipova et al. 2018). APS-oxidized NFC was compared with traditional TEMPO-oxidized NFC regarding improvement of paper properties, and it was found to give an equivalent outcome at the same degree of carboxylation (Filipova et al. 2020). As shown previously, cellulose nanoparticles, when combined with other biopolymers due to their size, form a denser microstructure, thus acting as a reinforcing agent for the films. In addition, they create a physical barrier reducing the gas molecule movements through the film (Lavrič et al. 2021). While NFC has been used before as a reinforcing agent, it has not been shown how adding different concentrations of APS-oxidised NFC would affect alginate and chitosan biofilm mechanical and degradation in the soil properties, since different functional groups may result in different compatibility with other biopolymers and composite film properties.

Therefore, this study’s novelty was to show how creating composite films using different concentrations of NFC acquired through the APS oxidation pretreatment method as a reinforcing agent would improve their barrier properties, making them suitable as potential coatings for paper food packaging.

**EXPERIMENTAL**

**Materials**

Sodium alginate, glycerol, and high molecular weight chitosan ($M_w = 310,000$ to $375,000$ Da) with at least 75% of deacetylation were purchased from Sigma Aldrich (Steinheim, Germany). Nanocellulose was obtained from the bleached hardwood kraft pulp (kindly provided by Metsä Fibre, Äänekoski, Finland).
Nanofibrillated Cellulose Production

The NFC was produced as described previously (Filipova et al. 2018; Lavrič et al. 2021). Briefly, bleached hardwood Kraft pulp was treated chemically by oxidising fibres at 70 °C for 4 h under continuous stirring in ammonium persulfate (APS) solution with the APS to fibres ratio of 5:1. The oxidation process was stopped by placing the mixture in the ice bath until it reached 15 °C. Thereafter, oxidised fibres were washed until neutral pH was reached and then kept at 4 °C. Afterwards, oxidised cellulose fibres were treated mechanically. Fibres were suspended in water (1.5% w/w), sonicated for 15 min (90% power, 9 s on, 1 s off) using an ultrasonic homogeniser (Sonic-650W, MRC Ltd., Holon, Israel), and processed in a microfluidiser (LM20, Microfluidics, Quadro Engineering, Waterloo, Canada). The processing was done for the first three times, through 200 μm ceramic chamber H30Z, followed by 100 μm diamond chamber H10Z at 300–600–900–1500 bar with three passes at each pressure, and then - with 6 passes at 2000 bar. During this procedure, samples were kept in an ice bath. A semi-transparent viscous 1.5% w/w solution was obtained and kept at 4 °C until used. The yield of NFC was ~ 80% from the initial amount of pulp.

Fabrication of Films

Films were prepared using different ratios of alginate: glycerol: NFC and chitosan: glycerol: NFC. Biopolymer concentration for alginate and chitosan was chosen at 2% w/w by dissolving alginate in deionised water, glycerol, and chitosan in a 1.0 wt% aqueous solution of lactic acid and glycerol. Glycerol was used as a plasticiser with a ratio of 1:2 to the biopolymer. Alginate and chitosan were dissolved for 15 min at 40 °C under constant stirring using a Heidolph RZR 2050 (Heidolph Instruments, Schwabach, Germany) stirrer. For both biopolymers, NFC additives in amounts of 1%, 2.5%, 5%, 7.5%, and 10% w/w to the main biopolymer were used; NFC was homogenised with UltraTurrax (Ika, Staufen, Germany) at 10,000 rpm for 5 min before being added to the main biopolymer. A vacuum pump removed air bubbles to obtain a homogeneous dispersion. Finally, composite films were prepared using a casting method. The solutions were poured into 10 cm Ø polyurethane petri dishes until reaching 30 g and then left at room temperature (RT) at a relative humidity (RH) of 40% overnight, after which they were dried at 60 °C for 6 h for alginate films and 7 h for chitosan films in the Universal oven U (Memmert GmBH, Schwabach, Germany).

Mechanical Properties

Before any tests, films were conditioned for 72 h in the chamber with 23 °C and RH 50%. Sample thickness was determined by micrometre F16502 (Frank PTI, Birkenau, Germany) at 14 random positions of the 10Ø cm sample before cutting samples into three 1.5-cm-wide and 5-cm-long strips. Then, the thickness of each strip was measured in the middle section, and the samples were weighed using an analytical scale.

The films’ tensile strength (TS, MPa) and elongation at break (EAB, %) were tested in line with the standard method ASTM D882 (2018) using Frank-PTI’s Tensile Tester Vertical F81838 (Frank PTI, Birkenau, Germany). Samples were tested at 10 mm/min speed with the clamping length set to 50 mm. All measurements were done in triplicate.

Scanning Electron Microscopy (SEM)

Film surfaces and side views were observed under SEM Tescan Vega TX (Brno, Czech Republic). Samples were torn to expose the presence of air bubbles or aggregates.
Before imaging, samples were coated with gold plasma using a K550X sputter coater (Emitech, Chelmsford, UK). Films were placed vertically on graphite tape and scanned under 1000x magnification.

**Air Permeability**

Air permeability was tested according to ISO 5636-3 (2013) at 23 °C and 50% RH, using Air Permeability Tester 266 (Lorentzen & Wettre, Sweden). Measurements were done in triplicates.

**Grease Barrier Properties**

Considering that the composite films were investigated as a possible paper coating for food packaging materials, the grease resistance of the films was determined according to the TAPPI T454 om-10 standard (2010). The grease resistance evaluation of the films was performed using olive oil, rapeseed oil, and turpentine as an example of low molecular oil to measure grease penetration through greaseproof material. Films were placed on the Whatman filter papers (Merck, Steinheim, Germany), and five drops of each oil were placed on the samples. Film permeability was verified at 1 h, 4 h, 8 h, 24 h, 48 h, and then every day until 7 days.

**Water Vapour Permeability (WVP)**

Water vapour transmission rate (WVTR) was determined according to ISO 2528 (2017) at 23 °C and 50% RH using dynamic climate chamber MKF 240 (Binder GmbH, Tuttlingen, Germany). Containers with 5 cm Ø holes in the caps were prepared. About 3 g of anhydrous CaCl$_2$ were placed in each container and were covered with biopolymer film, sealed with grease on the sides to ensure tight seal, and screwed with the cap. Vessels with CaCl$_2$ were weighed before being placed into a condition-controlled chamber. The amount of water vapour transferred through the composite films was calculated from the increasing weight of the containers, which were weighed in 1-h intervals for 8 h and then 3× per day until equilibrium was reached. The average increase in water mass per day was calculated against the sample area. The WVTR was calculated based on the Eq. 1,

\[
WVTR = \frac{\Delta m}{A \cdot t} \quad (g \cdot m \cdot day^{-1}) \tag{1}
\]

where \(A\) is the tested area (m$^2$), \(t\) is the time after 24 h of testing, and \(\Delta m\) is the mass difference of the tested sample.

Based on WVTR, water vapour permeability (WVP) was calculated (Eq. 2),

\[
WVP = \frac{WVTR \cdot e}{p_s \cdot RH} \quad (g \cdot m / Pa \cdot s \cdot m^2) \tag{2}
\]

where \(e\) is the average thickness of the sample (m), \(RH\) is the relative humidity (%) of the climate-controlled chamber, and \(p_s\) is the partial pressure (Pa).

**Fourier Transform Infrared Spectroscopy (FTIR)**

The Fourier transform infrared (FTIR) spectroscopy was used to study the chemical structure of the polysaccharide films. The FTIR was performed using KBr (IR grade, Sigma Aldrich, Darmstadt, Germany) pellets with a Thermo Fisher Scientific Nicolet iS50 spectrometer (Waltham, MA, USA). The analysis was performed in a 4000 to 450 cm$^{-1}$ spectral region with a resolution of 4 cm$^{-1}$, and 32 scans. The pellet contained ~2 mg of the
milled films and 200 mg of KBr. Both spectra were normalised to the highest absorption maxima.

**Degradation in Soil**

The rate of decomposition of the films in the soil was determined according to the standard method EN 14045 (2003). The experiment was conducted in a climate-controlled room at 23 °C and 50% RH for up to 3 weeks using compost (NPK 0.5-0.07-0.05, pH 6 to 7, organic matter content 16%, and moisture > 50%) acquired from Zeltabele Ltd. (Jaunauce, Latvia).

Four groups – 10 cm Ø of pristine alginate and chitosan films and films with 10% NFC additive – were each placed in a perforated plastic container filled with compost and then buried in a large compost tray until fully covered. The film degradation rate was monitored every 7 days. Degradation was assessed visually. The content of the compost was placed on a flat surface, and the material was investigated to find out whether any unbroken pieces could be distinguished from the rest of the compost. Once no distinguishable pieces could be found, it was considered that the film had biodegraded.

**Statistical Analysis**

Before statistical analysis, data were verified for normality and homogeneity of variances using Levene’s test. Statistical analysis was done using SPSS software (IBM SPSS Version 20.0, Armonk, NY: IBM Corp). All values were represented as mean ± SEM. For normally distributed data with equal variances, one-way ANOVA and Tukey post-hoc test were used. When data were not normally distributed or had unequal variances, the non-parametric Kruskal–Wallis and Mann–Whitney U-test was used. For comparison between the two groups, Student’s t-test was used.

**RESULTS AND DISCUSSION**

**Mechanical Properties**

Before establishing experimental groups, the best ratio of glycerol to biopolymer was tested. It has been shown that the low molecular weight of plasticisers such as glycerol results in low intermolecular forces between the polymer chains and promotes the mobility of the structure (Brdlík et al. 2022). Glycerol was chosen as the plasticiser to improve the mechanical properties, in particular, the flexibility of the films (Silva et al. 2023). In the literature, 30 wt% of glycerol with respect to the main biopolymer was commonly used (Roy and Rhim 2020; Lavrič et al. 2021; Marangoni Júnior et al. 2021; Priyadarshi et al. 2021). At this ratio, the films were too brittle and would break easily. The reason could be that the RH of the surrounding environment outside of the experimental chambers would reach 40% at best, indicating low moisture content in the air and thus affecting the flexibility of the samples. Similarly, it was found that the order of addition of ingredients was also important. If NFC was added to already dissolved alginate or chitosan mixture and then dispersed, it would result in a solution with NFC clumps, while the dispersion of nanocellulose using Ultra-Turrex before the addition of main biopolymer powder resulted in a more homogeneous solution and afterwards more homogeneous films. All 12 groups of alginate-NFC and chitosan-NFC samples are presented in Fig. 1 and were labelled based on the main polysaccharide used and the percentage of the NFC.
Fig. 1. Alginate-NFC and chitosan-NFC samples based on the different concentrations of NFC

As shown in Fig. 2, chitosan in the pristine biopolymer samples showed lower tensile strength (3.25 ± 0.18 MPa) than in pristine alginate samples (12.71 ± 1.22 MPa). Overall, the addition of NFC significantly improved the mechanical strength of both composite films.

Fig. 2. Tensile strengths of alginate and chitosan films based on the added NFC percentages. *p < 0.05; **p < 0.01; *** p < 0.001; **** p < 0.001 (ANOVA with Tukey’s post hoc test for Alginate group and Kruskal-Wallis, Mann-Whitney U test for Chitosan group). Data are represented as ± SEM.

Adding 1% of NFC improved tensile strength only in the chitosan group. However, 2.5% of NFC significantly increased the strength of alginate and chitosan films by 33% (p
< 0.05) and 50% (p < 0.0001), respectively. Adding 5% of NFC compared to 2.5% NFC improved tensile strength only in the chitosan group (p < 0.001) but did not show additional improvements in the alginate group. Films containing 7.5% NFC exhibited the highest tensile strength and the highest resistance to break, significantly higher than chitosan with the 5% NFC group (p < 0.0001). The mechanical properties compared to the control groups were improved by 66% for alginate and 175% for chitosan samples. As shown previously (Sirviö et al. 2014), these improvements in the mechanical properties of the films can be attributed to the formation of tightly hydrogen-bonded networks between NFC and polysaccharide matrix. The 10% NFC addition failed to increase further mechanical properties of the composite films, which could be due to the formation of nanocellulose agglomerates and the inability to introduce nanocellulose in a sufficiently homogeneous manner between polymer matrices. According to the strength results, 2.5% and 7.5% nanocellulose samples showed the best mechanical resistance.

When analysing stretch at break (Fig. 3), alginate films stretched 30% to 40% until the break, while chitosan films stretched 40% to 50%, thus indicating that chitosan films were more flexible. The addition of NFC reduced film flexibility.

![Fig. 3. Elongation at break of alginate and chitosan films based on the percentage of NFC added. *p < 0.05; **p < 0.01; *** p <0.001; **** p < 0.001; p < 0.0001 (Two-tailed t-test). Data are represented as ± SEM.](image)

For alginate films, the addition of 1% of NFC appeared to reduce film flexibility by 20%; however, this drop was not significant. Starting from the addition of 5% NFC, there were significant changes in the elasticity of the alginate samples (p < 0.01). For chitosan, the addition of 1% NFC improved the flexibility of the composite films (p < 0.05), while the flexibility compared to pristine chitosan films dropped 20% only after the addition of 7.5% NFC (p < 0.0001), indicating that it had greater rigidity, and consequently lower elongation of its structure, than the rest of the films. This is not surprising because incorporating NFC decreases the maximum elongation at break of chitosan (Fernandes et al. 2010) and alginate films (Sirviö et al. 2014). Thus, adding 7.5% NFC for chitosan
samples provided the highest mechanical strength and showed a drop in the stretching ability. From the stretchability point of view, 5% NFC for chitosan and 7.5% NFC for alginate showed the best results.

Air Permeability

Air and, in particular, oxygen permeability are important for food packaging to maintain the quality of the food and extend product shelf life (Brody et al. 2008).

All 12 samples were impervious to air, reaching 0.265 mL/min, a baseline value for an air-non-permeable material (aluminium plate). As confirmed by the scanning electron micrographs of the side view of alginate - NFC and chitosan – NFC composite films (Fig. 4), there were no visible pores in the membranes, and the surface of the membranes appeared smooth. This was expected because polysaccharide-based films, especially with alginate, are known to have high gas barrier properties (Senturk Parreidt et al. 2018; Zinina et al. 2023).

Fig. 4. Scanning electron micrographs of the side views of alginate - NFC and chitosan – NFC composite films

The authors’ recent research has shown that hemp fibre-containing paper had the lowest air permeability among different recycled paper blend options (Filipova et al. 2023), reaching 372 mL/min. Therefore, it is hypothesised that using alginate or chitosan with a NFC mixture as paper packaging coatings could further decrease paper permeability to air.

Grease Barrier Permeability

None of the samples showed any oil permeability during the entire time of the experiments, which was expected based on the other publications (Ham-Pichavant et al. 2005; Sirviö et al. 2014; Zinina et al. 2023). The difference in comparison to the other studies with much shorter experimental time (up to 30 min) was that the grease barrier properties evaluation was carried out until 7 days when the experiments were stopped due to oil evaporation, indicating that the composite materials have excellent grease barrier properties.

Because uncoated paper is not grease-proof, paper packaging coating with either an alginate or chitosan composite biofilm layer could greatly improve paper resistance to greases, including food-grade oils, thus allowing its use for food packaging. Considering that the obtained biopolymer films are air and oil-impermeable, they could be used as a
paper coating for packaging materials, e.g., food boxes and bags. Notably, such materials could be recycled repeatedly using the current recycling system, which is difficult with paper materials coated with synthetic polymers.

**Water Vapour Permeability**

Because biopolymer films have a high affinity for water, it is crucial to reduce the moisture transfer between the food and the films and *vice versa*. As shown in Fig. 4, all samples exhibited moderate moisture-barrier properties; in some cases, CaCl$_2$ was fully dissolved only after 7 days (168 h).

![Water vapour permeability](image)

**Fig. 5.** Water vapour permeability based on the percentage of NFC added.

As shown in Fig. 5, the pristine alginate films had higher WVP than pristine chitosan films. While adding 1% NFC did not contribute much resistance to moisture (the decrease in WVP was only 10%), adding 2.5% NFC increased moisture-barrier properties. Because alginate has hydroxyl groups and a large degree of hydrogen bonding, it is highly hydrophilic and, therefore, has high WVP (Marcos *et al.* 2010; Zinina *et al.* 2023). The addition of NFC fills the alginate matrix, thus decreasing the porosity of the films and, consequently, its permeability to moisture (Sirviö *et al.* 2014). Adding higher concentrations of NFC did not further improve the water vapour barrier properties, possibly showing that no new bonds could be created between NFC and alginate. The WVP of the pristine NFC was measured as $0.55 \times 10^{-9}$ g-m/Pa-s-m$^2$ (Ezati *et al.* 2022). The addition of 2.5% NFC to the alginate reached an average of $0.53 \times 10^{-9}$ g-m/Par-s-m$^2$. Overall, adding 2.5% to 10%, NFC improved the moisture barrier properties by 22% to 28%. However, no changes were significant.
In contrast, while pristine chitosan and CHI + 1% NFC had initially similar WVP, adding NFC at higher concentrations (starting from 5% at 51h) significantly increased WVP \( (p < 0.05) \), indicating that the biofilms were more permeable to water vapour. However, there was no significant difference in the increase of WVP between the addition of 2.5% and 5% NFC. The reason could be that chitosan itself is more hydrophobic than alginate, thus having higher resistance to water vapour and that the addition of 1% NFC was too low a concentration to affect the hydrophobicity of the chitosan. However, because NFC, while not being soluble in the water, is more hydrophilic, the addition of higher concentrations of NFC made the entire chitosan-NFC films more hydrophilic, thus increasing their affinity to water. In addition, since chitosan forms polyelectrolyte complexes with NFC, it may form agglomerates, creating uneven at the nano-scale distribution and subjecting the films to higher water vapour permeation. Therefore, considering the mechanical properties, the best performance for chitosan-NFC composite films appears when 2.5% NFC is added.

**FTIR**

The FTIR spectra were recorded to investigate the chemical structure of the raw materials. As shown in Fig. 6, in the ATR-FTIR spectrum of alginate films, fluctuations of the OH group at 3258 cm\(^{-1}\) are clearly visible; C-H vibrations in the -CH\(_2\)- group appeared at 2930 and 2875 cm\(^{-1}\); asymmetric oscillations of the carboxylate ion COO\(^-\) were evident at 1601 cm\(^{-1}\) and symmetric oscillations were seen at 1407 cm\(^{-1}\), and there were various C-C, C-O, C-O-C symmetric and asymmetric oscillations at 1089, 1025, and 930 cm\(^{-1}\). These features are characteristic of the alginate sugar (glucuronic acid) pyranose skeleton residue.

![Fig. 6. FTIR spectrum of alginate-NFC and chitosan-NFC films](image)

The intensity of oscillations of carboxylate ions at 1601 and 1407 cm\(^{-1}\) depends on the amount of water in the films (Xiao et al. 2014); adding 10% NFC to the alginate film binds water in the film and increases the intensity of absorption peaks of carboxylate ions. The ATR-FTIR spectra of chitosan films clearly show the oscillations of -OH and N-H groups at 3275 cm\(^{-1}\), C-H oscillations of the -CH\(_2\)- group at 2924 and 2875 cm\(^{-1}\), deformation oscillations of the -NH\(_2\) group at 1556 cm\(^{-1}\), and various C-C, C-O, C-O-C
symmetric and asymmetric vibrations at 1151, 1026, and 925 cm\(^{-1}\). These absorbances are characteristic of the chitosan sugar (2-aminoglucose) pyranose skeleton residue. Chitosan contains a small amount of acetate groups, which is indicated by the C=O fluctuations of the amide group at 1652 cm\(^{-1}\). Adding 10% NFC to the chitosan film slightly decreased the absorption intensity of amino and amide groups. The obtained spectra are in accordance with the FTIR spectra of alginate and chitosan published in the literature (Lagaron et al. 2007; Xiao et al. 2014). FTIR data showed that the composite films were free of unwanted non-polysaccharide impurities such as aromatic compounds.

### Degradation in Soil

According to the EN 13432 (2000) standard, a film is considered biodegradable when 90% of its material decomposes due to biological action within six months.

Figure 7 shows the biodegradation of the samples. Four groups of samples were chosen – ALG + 0% NFC, ALG + 10% NFC, CHI + 0% NFC, and CHI + 10% NFC. After one week of biodegradation, no pristine alginate samples could be found in the soil, while in the case of ALG + 10% NFC, only small pieces of ~2 cm could be detected in the soil. Chitosan samples were more robust; on average, pieces of ~3 to 4 cm of CHI + 0% NFC and ~4 to 5 cm of CHI + 10% NFC could be found at one week post-deposition in the soil. No alginate-based samples could be detected in the soil by two weeks, indicating their complete biodegradation. In the case of CHI + 0% NFC, although the size of the samples had not changed much since the first week of the experiment, the samples were too brittle to be cleaned from the soil, indicating that the degradation of the sample was almost complete. For the CHI + 10% NFC samples, only small pieces of less than 2 cm were seen at 2 weeks. No biofilms could be detected in the soil at 3 weeks. Therefore, the experiment showed that all samples had a good level of biodegradation, being fully decomposed within three weeks.

![Fig. 7. Biodegradation of the alginate + 10% NFC, chitosan + 0% NFC, and chitosan + 10% NFC films; Scale bar at 2 cm](image-url)
The data for chitosan and alginate films were consistent with other studies. Studies have shown that alginate films decompose in the soil and sand in about 2 weeks (Santos et al. 2022; Zinina et al. 2023). For chitosan, a study analysing film degradation in commercial compost, garden soil, and vineyard soil established that by 10 days, chitosan films were fully degraded (Oberlittner et al. 2021). Slightly slower biodegradation for the NFC-containing samples can be explained by the higher stability of films due to the addition of the NFC. The high crosslinking ability of alginate and chitosan films with NFC contributed to greater resistance to biodegradation factors presented in the soil. However, even with the addition of 10% NFC, the sample degradation rate was rapid, indicating that while improving the mechanical properties of the films, NFC did not hinder sample biodegradation.

Since in paper biodegradation experiments, samples would fully degrade within 12 weeks (Venelampi et al. 2003; Filipova et al. 2023), the addition of alginate-NFC or chitosan-NFC in the form of coatings should not affect the biodegradation of the paper packaging.

CONCLUSIONS

1. The addition of nanofibrillated cellulose (NFC) to the biopolymers improved material functional characteristics. The addition of nanocellulose reduced film flexibility by around 20%; however, it increased the strengths of alginate and chitosan films by 33 to 55% and 66 to 175%, respectively.

2. The obtained biopolymer films exhibited good barrier properties. Even though the water vapor transmission rate (WVTR) was lower than for the synthetic polymers, alginate-NFC and chitosan-NFC films were impermeable to air and oil.

3. Overall, adding 2.5% NFC in alginate and chitosan samples exhibited the best material properties.

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