

## Research Article

# Acrylate/Nanofibrillated Cellulose Nanocomposites and Their Use for Paper Coating

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Nanofibrillated cellulose (NFC) isolated from TEMPO-oxidized rice straw was used to improve thermal and tensile strength properties of acrylate polymer films. Acrylate/NFC mixture containing 15% NFC was used for paper coating, and properties of paper sheets including tensile strength, water absorption, and microscopic structure were investigated. The results showed that the presence of NFC in the acrylate matrix significantly improved tensile strength properties and thermomechanical properties of the acrylate polymer and caused moderate increase in its moisture sorption. The presence of NFC in acrylate emulsion caused significant increase in its viscosity. Paper sheets coated with different thin layers (from 0.2 to 6 microns) of acrylate/NFC showed improvement in tensile strength and decrease of water absorption.

## 1. Introduction

Acrylate polymers are type of polymers which could be referred to plastics and commonly known as acrylics or polyacrylates. They are characterized by transparency and elasticity. Acrylates are widely used for many applications, as in cosmetics, adhesives, textile, paints, and as cement modifiers. Among their applications also is their use in paper coatings as sizing and waterproof agents to reduce water absorption of paper sheets [1, 2]. However, acrylate polymers suffer from some drawbacks such as low mechanical properties at ambient or high temperatures, especially above their glass transition temperature. This limits their application in coating and printing. To overcome these drawbacks, modifications to acrylates have been investigated before their applications. For example, psyllium-g-poly(acrylic acid-co-sodium acrylate)/cloisite 10A semi-IPN nanocomposite hydrogel was prepared by grafting of poly(acrylic acid-co-sodium acrylate) onto psyllium backbone. The prepared nanocomposite hydrogel exhibited biocompatible, tough, and elastomeric soft biomaterial for controlled release of drug molecules.

[3]. Also, for the same application, grafted polymethacrylic acid onto sodium alginate and the prepared hydrogel possess biodegradability, noncytotoxicity toughness, rubber-like elasticity, and suitability for responsive controlled release vector [4].

In the acrylic bone cement preparations, acrylate copolymers composed of methyl methacrylate and acrylic acid were blended with corn starch/cellulose acetate in the presence of hydroxylapatite as filler. This modification improved mechanical properties and also promoted the ingrowth of new bone, leading to the complete healing around the fixed (cemented) implant when biodegradation process occurs [5].

The use of NFC with polyacrylates is not widely studied. For example, cellulose nanofibers and bacterial cellulose were used with polyacrylic acid (PAA) in nanocomposites for electrodes and supercapacitor applications [6]. Also, polyacrylic acid modified with tetraaminophthalocyanine copper (CuTaPc) was deposited on the surface of nanofibrous cellulose by using layer-by-layer (LBL) technique, producing nanocomposites that can be used as a catalyst or as a sensor

[7]. Aerogel prepared from NFC and modified with PVA and PAA produced new material capable of removing *E. coli* bacteria from water [8]. Recently, cellulose nanofibers were also used with acrylate polymer for making membranes for heavy metal ion removal, especially cadmium, from impaired waters [9] and for removal of pyrene from water [10]. The reinforcing effect of three types of NFC (mechanically prepared NFC, TEMPO-oxidized, and enzymatically pretreated NFC) as a filler material in polysodium acrylate superabsorbents was studied; it was found that NFC and enzymatically isolated NFC were more effective than TEMPO NFC as a filler material [11]. NFC also showed reinforcing effect when mixed with different types of acrylates for wood coating before and after aging [12, 13]. NFC was graft-copolymerized with acrylic monomers (glycidyl methacrylate, ethyl acrylate, methyl methacrylate, butyl acrylate, and 2-hydroxyethyl methacrylate) to impart its hydrophobic character and improve its heat resistance [14]. Besides the previous applications of acrylates with NFC, a biodegradable bone cement was prepared by free radical polymerization of solid and liquid phases; the solid phase was corn starch/cellulose acetate blends and the liquid phase was methyl methacrylate and acrylic acid in the presence of hydroxyapatite to confer a bone-bonding character to the bone cements. The results showed that the prepared composite can be used as self-curing bone cements and exhibited several advantages with respect to other commercially available bone cements [5]. The addition of surface-initiated cellulose nanocrystals to the poly(MMA-co-BA) lead to the increase of tensile strength by about 100% compared to the linear P(MMA-co-BA) [15].

In the current work, NFC was isolated from rice straw and used as reinforcing elements in acrylate matrix to improve its mechanical and thermomechanical properties to be used in paper coating applications. At first, films casted from acrylate/NFC containing different ratios of NFC (2.5 to 20 wt%) were evaluated regarding their tensile strength properties, thermomechanical properties, moisture sorption, water vapor permeability, and microscopic structure. In addition, the viscosity of acrylate emulsion/NFC mixtures was studied.

## 2. Material and Methods

**2.1. Preparation of Nanofibrillated Cellulose (NFC).** Nanofibrillated cellulose was prepared from bleached rice straw pulp according to the previously published methods [16, 17]. Rice straw pulp (3 g) was dispersed in distilled water (400 ml) with TEMPO (0.048 g, 0.3 mmol) and sodium bromide (0.48 g, 4.8 mmol). Then, 30 ml of sodium hypochlorite solution was then added with stirring and the pH was adjusted to 10. At the end of reaction, the pH is adjusted to 7 and the product was centrifuged at 5000 rpm. The product was further purified by repeated water addition, dispersion, and centrifugation. Finally, the product was purified by dialysis against water.

To obtain nanofibers, oxidized pulp was first disintegrated by a high-shear homogenizer (CAT Unidrive 1000) at 5000 rpm using pulp suspensions of 2% consistency.

Carboxylic groups' content of NFC was determined according to TAPPI Test Method T237cm-98 and found to be 0.31 mmol/g.

Commercial acrylate latex was used, Acril 33 (copolymer of acrylic acid, methacrylate), 46% and viscosity 1500–3000 mPa.s.

**2.2. Preparation of Acrylate/NFC Nanocomposites.** Nanofibrillated cellulose (NFC) suspension in water (1% wt %) was added to 48% aqueous emulsion of acrylate resin. A series containing acrylate/NFC (2.5 to 20 wt% based on oven dry weight of acrylate) were prepared and poured into petri dishes and dried in an oven at 40°C for 24 hours.

**2.3. Characterization of Acrylate/NFC Nanocomposites.** The characterization of the prepared acrylate/NFC films regarding tensile strength, dynamic mechanical thermal analysis, scanning electron microscopy, and water vapor permeability was carried out as mentioned. Equilibrium moisture content was determined by keeping the samples in a closed desiccator at 98% relative humidity using sodium sulfate saturated solution for different lengths of time, and the increase in weight was recorded.

Tensile tests were carried out with a Lloyd instrument (Lloyd Instruments, West Sussex, United Kingdom) with a 100 N load cell. The measurements were performed on strips with 1 cm width and 8 cm length at a crosshead speed of 2 mm/min at 25°C. Five replicates of each samples were measured and the results averaged.

Static WVP of films was determined according to the ASTM standard (ASTM Standards, 1995).

**2.4. Coating of Paper Sheets and Their Testing.** The coating of commercial writing and printing paper sheets and their testing regarding tensile strength and water absorption were carried out.

Paper sheets were coated manually with acrylate/NFC emulsion using coating bar having a gap clearance of 30–120 microns.

Tensile strength testing of coated paper sheets (for machine and cross directions) was carried out according to TAPPI T494-06 standard method using a universal testing machine (LR10K; Lloyd Instruments, Fareham, UK) with a 100 N load cell at a constant crosshead speed of 2 mm/min. Strips of 20 cm long and 15 mm width were used in the test and the span was 10 cm.

Water absorption test of paper sheets was carried out according to ISO 535 method (known as Cobb method). The test determines the quantity of water that can be absorbed by the surface of paper sheets in a given time.

## 3. Results and Discussion

NFC with a width of about 4–5 nm and several microns in length was isolated from rice straw [18]. Since the aim of the current work was to evaluate the use of NFC/acrylate for paper coating applications, acrylate nanocomposite films containing different ratios of NFC (2.5 to 20 wt%) were first prepared and their properties were evaluated to choose a suitable sample for paper coating. The properties evaluated

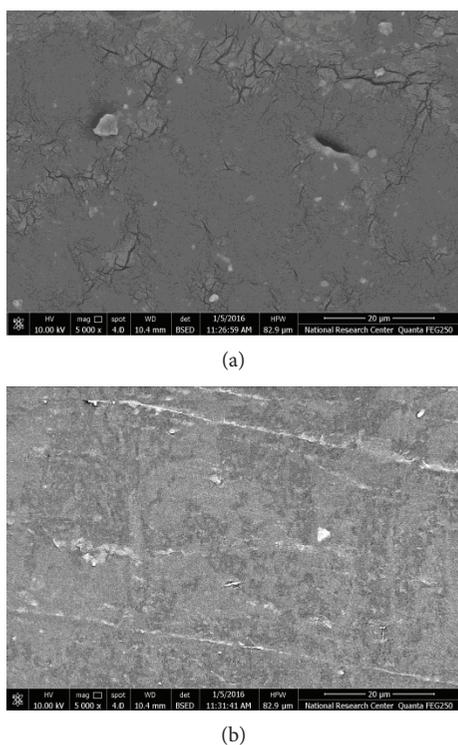


FIGURE 1: SEM of acrylate film (a) and acrylate/NFC containing 15% NFC.

were tensile strength properties, thermomechanical properties, moisture sorption, water vapor permeability, and microscopic structure.

**3.1. Properties of Acrylate/NFC Nanocomposite Films.** SEM images of acrylate and acrylate/NFC nanocomposite films are shown in Figure 1. As shown in the figure, neat acrylate surface appeared cracked; this could be due to the low glass transition temperature of the polymer; during drying at 45°C, it softened, and upon cooling, the surface became not smooth and shrunk. The addition of NFC to acrylate resulted in film with crack-free and smoother surface.

**3.1.1. Mechanical Properties.** Regarding the effect of NFC on tensile strength properties of acrylate films, the addition of NFC to acrylate polymer resulted in the improvement of its tensile strength and tensile modulus up to about 450% and 657%, respectively. This could be due to the strength of the nanofibers in addition to possible covalent crosslinks between NFC and acrylate polymers [19].

On the other hand, there was a decrease in strain at break or at maximum load, especially at NFC loading of >5% (Figure 2). The outstanding reinforcing effect of NFC in polymer matrices is explained mainly by entanglements of the nanofibers in the matrix [20] and formation of continuous network within the polymer matrix above critical threshold. The percentage increase in tensile strength ranged from 38% to 450% on adding 2.5% to 20% of NFC, respectively, while percentage increase in Young's modulus ranged from 10% to 657% on adding 2.5% to 20% of NFC, respectively. On the other hand, percentage decrease in strain at

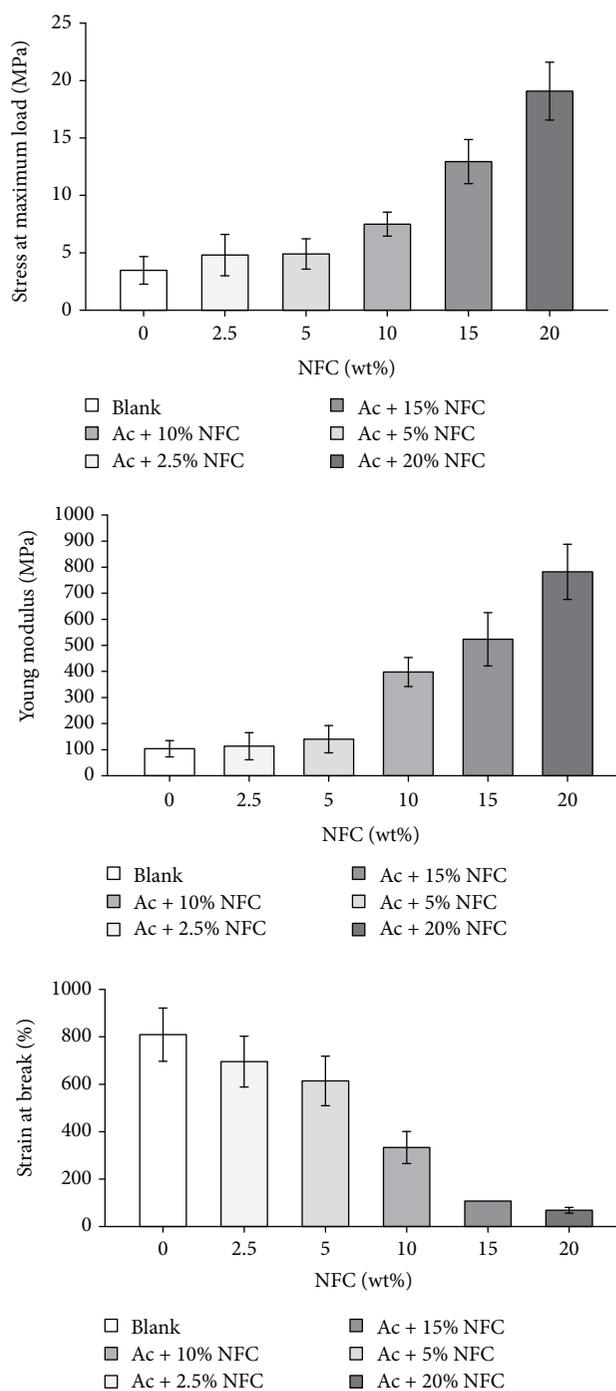


FIGURE 2: Tensile strength properties of acrylate/NFC nanocomposite films.

maximum load was high and ranged from 14% to 91.5% on adding 2.5% to 20% of NFC, respectively.

**3.1.2. Dynamic Mechanical Thermal Analysis (DMTA).** To study the reinforcing effect of NFC below and above glass transition temperatures of the acrylate polymer, DMTA was carried out at temperature range from -100 to 150°C. Figure 3 shows DMTA curves of acrylate/NFC nanocomposite films. As shown from storage modulus curves, the presence of NFC resulted in increasing stiffness of acrylate

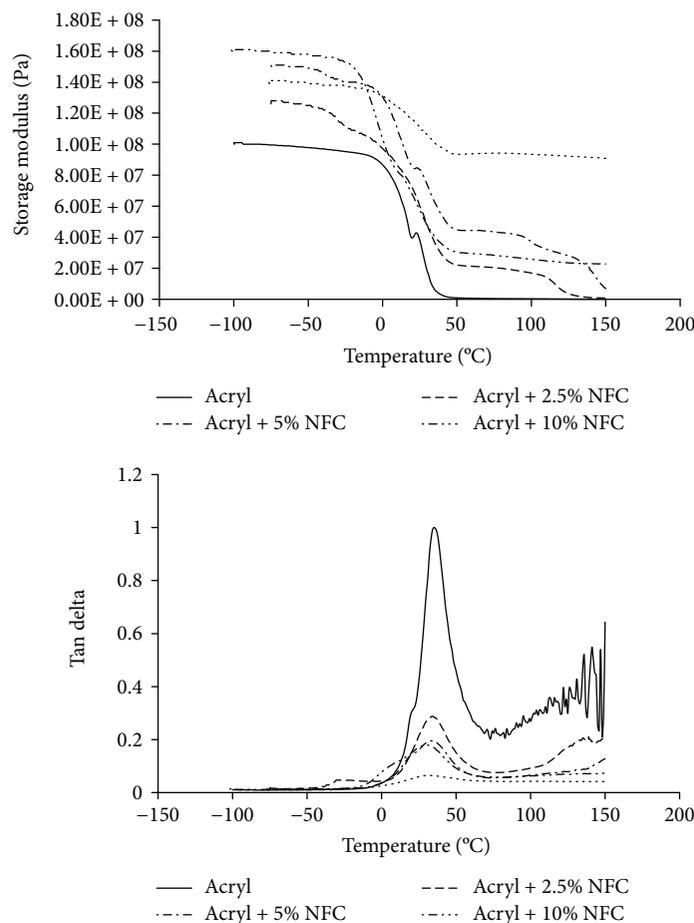


FIGURE 3: DMTA of acrylate/NFC nanocomposite films.

below and above  $T_g$  of the polymer even at the lowest NFC loading used (2.5%). The storage modulus curve of acrylate polymer showed a drop in storage modulus which started at about  $-5^\circ\text{C}$ , as a result of approaching  $\alpha$ -relaxation associated with motion of long chain of acrylate polymer at its glass transition temperature. The presence of NFC clearly reinforces the acrylate polymer at its rubbery stage as it is clear from the significant increase in storage modulus, which increased as the content of NFC increased. The drop in storage modulus of neat acrylate as a result of transformation from glassy state to rubbery state was extremely high (6300%) while that drop was 500%, 259%, 247%, and 46% in the case of acrylate/NFC nanocomposites containing 2.5, 5, 10, and 20% of NFC, respectively. These values clearly indicate the reinforcing effect of NFC on the acrylate matrix and mean that NFC can support tensile strength of the polymer when it is soft by the action of temperature. In accordance with storage modulus curves, Tan delta curves showed peaks corresponding to the  $\alpha$ -relaxation and transformation from the glassy state to the rubbery state of acrylate polymer. Although there was no significant change in the peak value of the tan delta curve of neat acrylate polymer as a result of the addition of NFC (peak temperatures  $34$ – $28^\circ\text{C}$ ), the magnitude of the peak was greatly influenced by the amount of NFC. The decrease in Tan delta values as

a result of increasing NFC content means less energy dissipation at the rubbery stage of the polymer. Due to the nanosize diameter of NFC, their high dispersion and strong interaction with the polymer, the formation of immobilized polymer layer around NFC is expected to occur and thus restricts motion of polymer chains and causes decrease in damping factor [20]. For the neat polymer sample at temperature above  $100^\circ\text{C}$ , the polymer became very soft, and when subjected to stress during the test, the curve became very noisy due to the loss of mechanical strength. The DMTA results obtained in the current work are in close agreement with previous work on the use of NFC with styrene-co-acrylate latex [20].

**3.1.3. Moisture Sorption of Acrylate/NFC Nanocomposite Films.** Moisture sorption is an important property of films used in different applications. It could affect the mechanical properties of films and products packed by these films. Figure 4 shows moisture sorption of acrylate/NFC nanocomposite films conditioned at 98% relative humidity and  $25^\circ\text{C}$  for six weeks. As shown in the figure, neat acrylate film is characterized by relatively moderate moisture sorption. After one week, the moisture content of acrylate film was about 6%. The addition of NFC to acrylate resulted in increasing moisture sorption of the films. However, for films containing up

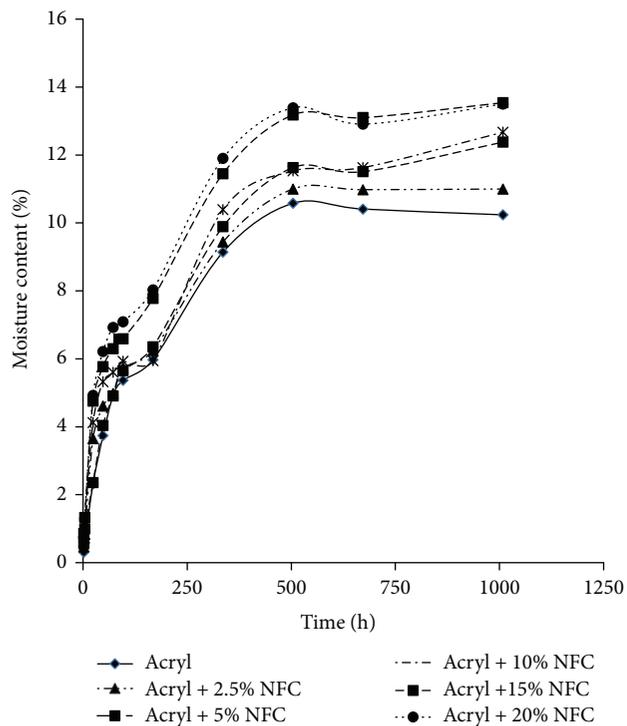


FIGURE 4: Moisture sorption of acrylate/NFC films at 98% relative humidity and 25°C.

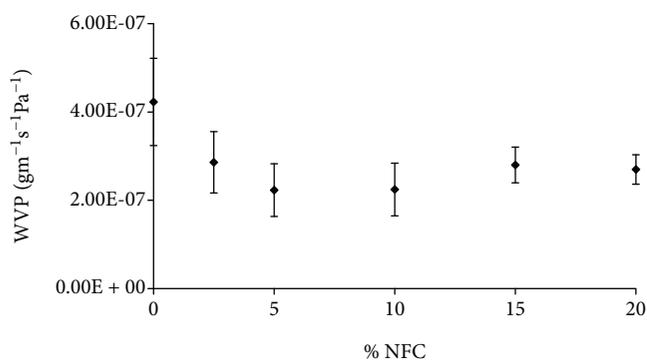


FIGURE 5: Water vapor permeability of acrylate/NFC nanocomposite films.

to 10% NFC, the rate of moisture sorption was higher than that of acrylate film, but after one week, the moisture content of films (~6.1–7.8%) was close to that of neat acrylate film. For acrylate/NFC films containing 15–20% NFC, moisture sorption was higher than other samples. The rate of moisture sorption for film containing 20% NFC was higher than that containing 15% NFC, but after one week, the moisture sorption of both of them was close to each other (~6–8%). The higher moisture content of films containing NFC is due to the higher hydrophilic nature of NFC. After 8 weeks, equilibrium moisture content of neat acrylate film was 10.2% while acryl/NFC composite films had equilibrium moisture content from 11 to 13.5%. The obtained results showed that in spite of the high hydrophilic character of NFC, they were homogeneously distributed within the acrylate polymer

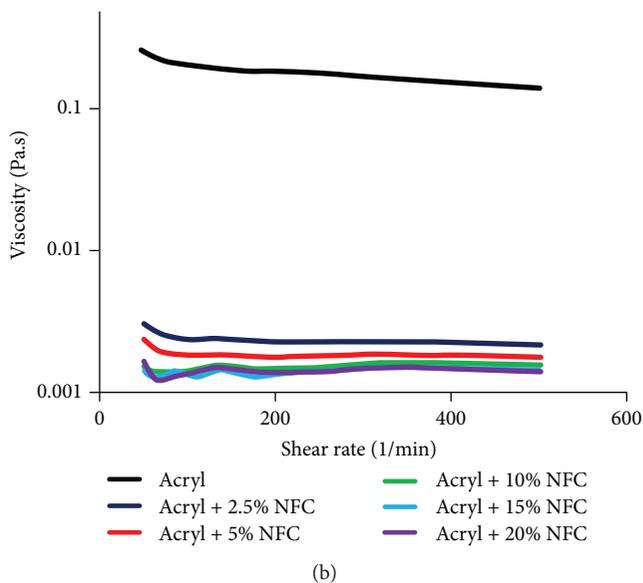
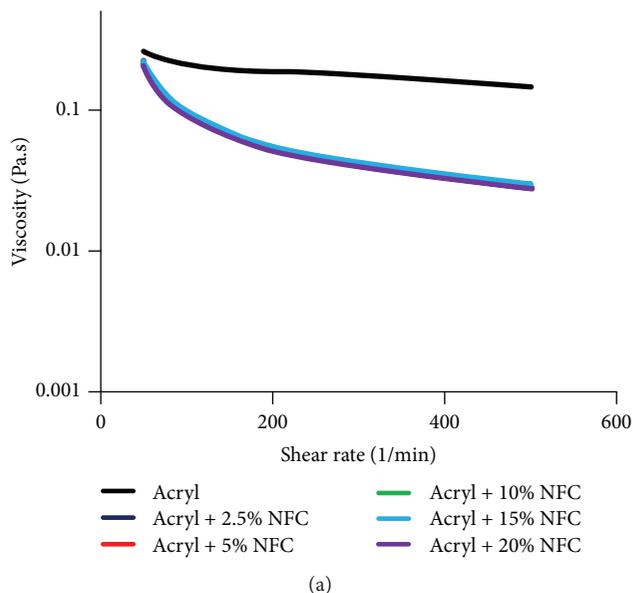


FIGURE 6: Viscosity of acrylate/NFC mixtures (a) and acrylate containing the same water content in the NFC (b).

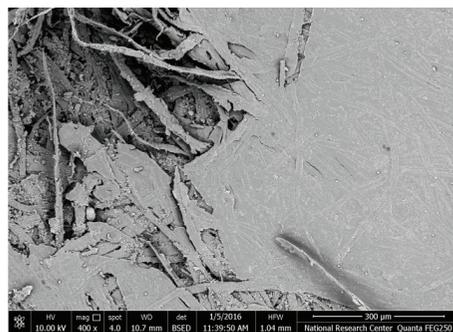


FIGURE 7: SEM image of paper sheet coated with acrylate/15% NFC mixture using 120-micron gap bar.

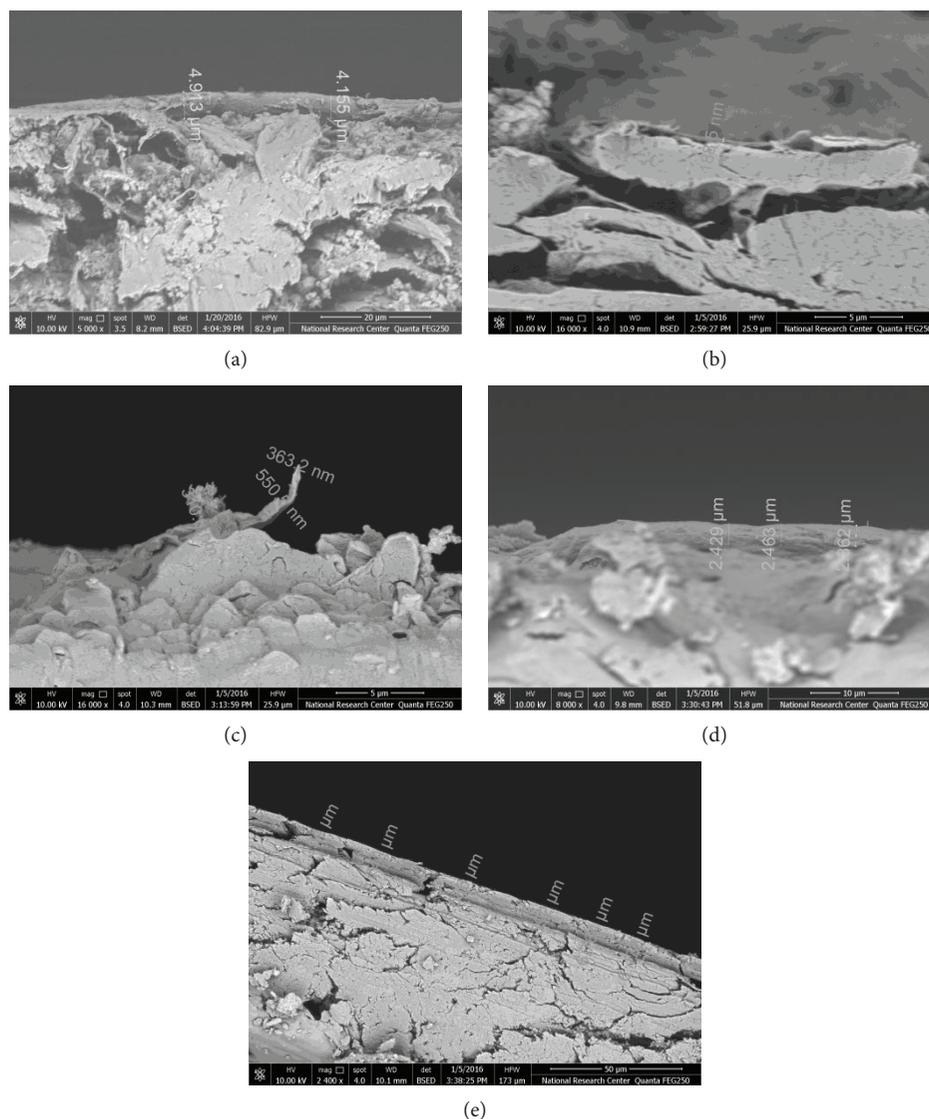


FIGURE 8: SEM images of paper sheets coated with neat acrylate (a) acrylate/15%NFC using 30 (b), 60 (c), 90 (d), and 120-micron (e) gap bar.

matrix and moderate moisture sorption took place at the high relative humidity used in the test.

### 3.1.4. Water Vapor Permeability of Acrylate/NFC Films.

Water vapor permeability is an important property in applications related to packaging. The presence of cellulose nanomaterials (cellulose nanocrystals or cellulose nanofibers) in polymeric matrices can affect the water vapor permeability of these polymers. The expected role of cellulosic nanomaterials in polymer matrices is that they make the movement of water vapor across polymer film more torturous due to their fibrous nature and high crystallinity and thus decrease the rate of permeability. However, this was found to be dependent on the relative hydrophilicity of cellulosic nanomaterials to the polymer matrix. For example, in case of using cellulose nanofibers with chitosan [21], hydroxypropyl and carboxymethyl guar [22], and gelatin [23], a decrease in WVP of the polymer matrices was noticed, while in case of using

cellulose nanofibers with polylactic acid [24], an increase in WVP was recorded.

In the current work, water vapor permeability of acrylate/NFC nanocomposite films was studied and the results are presented in Figure 5. The presence of NFC in the acrylate polymer matrix resulted in significant decrease in its WVP even at low NFC loading. But it was noticed that there was a tendency for increasing WVP at NFC loading > 10% although it remained lower than that of neat acrylate films. The decrease in WVP ranged from about 32% to 48% on increasing NFC from 2.5 to 10% with the lowest values at 5–10%. The tendency of WVP to increase at 15–20% NFC could be due to the high hydrophilic nature of NFC but still far from the WVP of neat acrylate film.

### 3.2. Coating of Paper Sheets with Acrylate/NFC Suspension

**3.2.1. Viscosity of Acrylate/NFC Suspension.** The viscosity of aqueous polymer emulsion is an important factor in paper

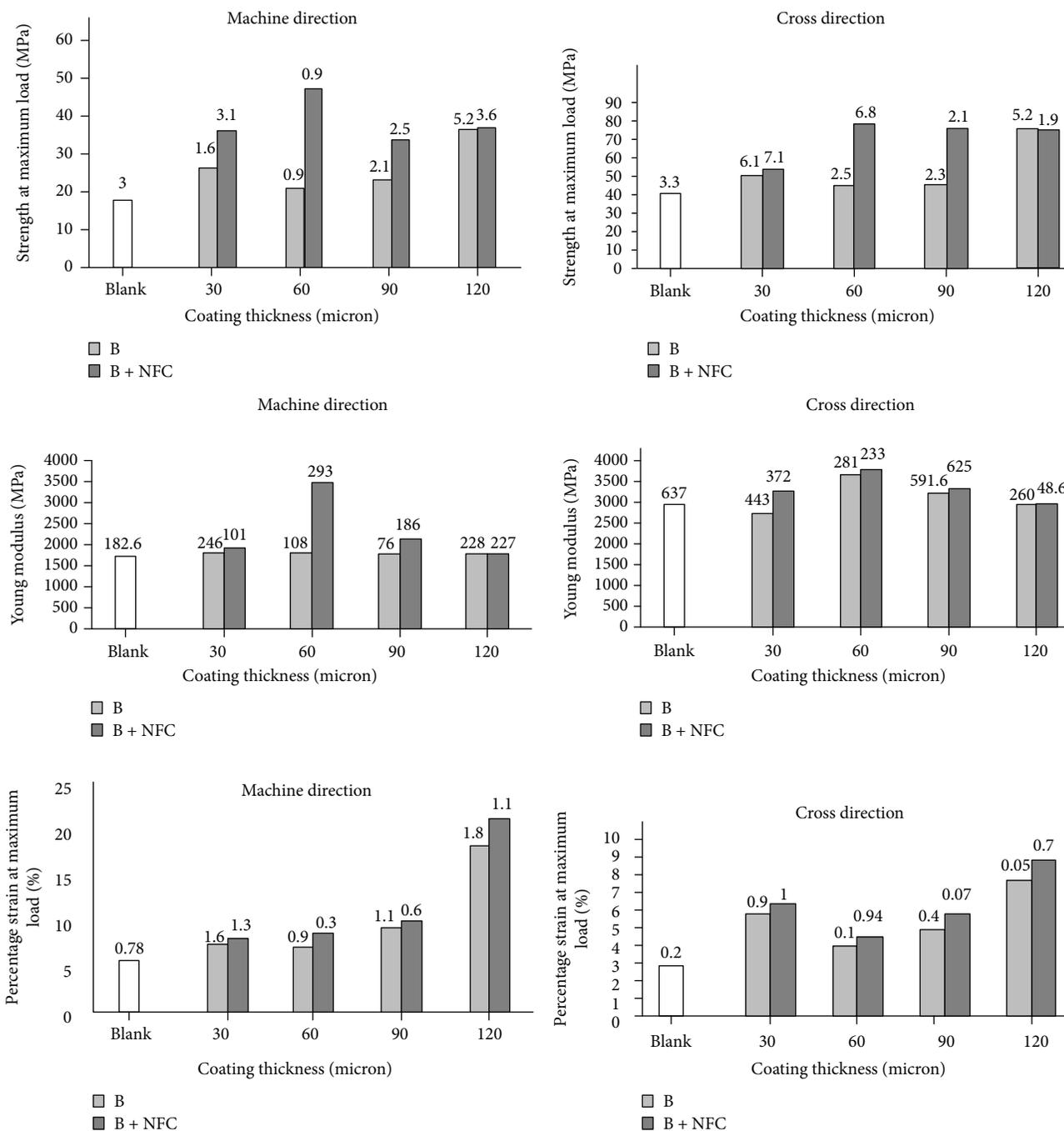


FIGURE 9: Mechanical properties of paper sheets coated with acrylate/NFC mixture. Values above the columns are the standard deviation.

coating applications since suitable viscosity ensures ease and uniform application of a thin layer of polymer as well as the penetration of polymer into the surface of coated layer. If the viscosity of polymer is very low, it cannot be applied as a continuous thin film and high water content affects ease of drying and strength of the paper after drying. On the other hand, highly viscous polymers may reduce the penetration of polymer between fibers of paper and thus do not improve strength properties of paper sheets. In the current work, NFC aqueous suspension in water (2 wt%) was added to

acrylate emulsion in ratios from 2.5 to 20 wt% (based on solid content of acrylate), and the viscosity of the mixtures at different shear rates was measured (Figure 6(a)). It was noted that although water content of the added NFC was high and resulted in dilution of acrylate emulsion, there was no noticeable decrease in the viscosity of the mixture on increasing the amount of NFC suspension added. The figure also shows that at low shear rate ( $50 \text{ min}^{-1}$ ), the viscosity of neat acrylate emulsion was close to the different acrylate/NFC emulsion compositions, while on increasing shear rate the

viscosity of acrylate/NFC mixtures was lower than that of neat acrylate emulsion. The obtained results indicate high dispersion of NFC in the mixture and strong interaction with the acrylate polymer. In addition, high affinity of NFC to water and formation of gel can cause an increase in viscosity of the mixture in water. Furthermore, the formation of NFC network within acrylate chains can also cause an increase in the viscosity of the mixture regarding the viscosity of acrylate solution without NFC. To confirm these different assumptions, the same amount of water present in NFC suspension was added alone to the acrylate emulsion and the viscosity was measured. As it is clear from Figure 6(b), the viscosity of acrylate emulsion is largely decreased as a result of the addition of water even at the lowest amount of water added.

**3.2.2. Properties of Coated Paper Sheets with Acrylate/NFC Films.** In the following part of work, the acrylate/NFC emulsion containing 15% of NFC was used for coating commercial writing and printing paper sheets using coating bar with a gap clearance of 30–120 microns. It was noticed that the coating operation was easy without dripping and with uniform thickness. Figure 7 shows SEM of paper sheets coated with 120-micron clearance gap. As shown in the figure, homogenous film could be coated onto the paper surface although the small thickness applied. It should be pointed out that the uncoated part at the paper surface was deliberately made to show the homogeneity of coating.

In order to know the thickness of coating layer applied and penetration of coating layer into the paper surface, a cross section of coated paper sheets using different clearance gaps (30–120 microns) was investigated using scanning electron microscopy (Figure 8). For comparison, the coating of paper sheet with the as-received acrylic emulsion (~49 wt%) was carried out using a 30-micron clearance gap. Indeed, it was not possible to use acrylate emulsion diluted with the same amount of water present in the 15% NFC suspension since the viscosity became very low and even coating film was difficult to form. As shown in the SEM images, the coating of paper sheet with the as-received acrylic emulsion resulted in film with about 4-micron thickness. It is also noticed that, due to high viscosity of the acrylate emulsion, no penetration of the polymer into the paper surface was noticed. On the other hand, the coating of paper sheets with acrylate 15% NFC mixture resulted in the formation of a thin film with a thickness of about 0.19, 0.5, 2.2, and 6.5 microns in case of using a gap clearance of 30, 60, 90, and 120 microns, respectively. It is also noted from the images that the penetration of acrylate/15%NFC mixture within the cross section of paper sheets could be seen in case of coating using a gap clearance of  $\geq 60$  microns.

**Mechanical Properties of Coated Paper.** Mechanical properties of paper sheets coated with neat acrylate and acrylate/15%NFC using 30, 60, 90, and 120-micron gap bar are shown in Figure 9. As it is clear from the figure, the coating of paper with neat acrylate improves the tensile strength and Young's modulus strength of paper in machine and cross directions. For tensile strength, the improvement was about 105% and 86%, respectively. For strain at maximum load, it was 223%

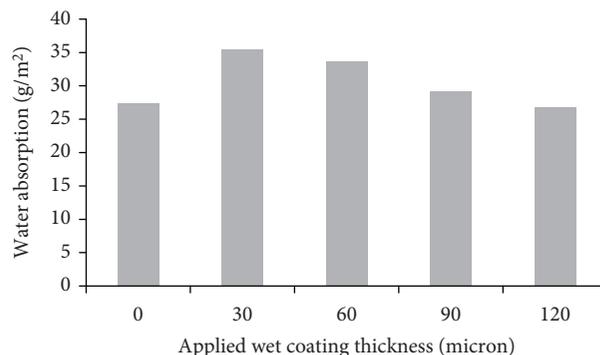


FIGURE 10: Water absorption of paper sheets coated with different thicknesses of acrylate/NFC.

and 168%, respectively, for both directions. While for Young's modulus, the improvement was 4.8% and 8.9%, in the machine and cross directions, respectively. This enhancement in the tensile strength could be due to the filling of the gaps between fibers with the polymer. Furthermore, the coating of paper sheets with a thin layer of the acrylate/NFC mixture leads to further improvement in its tensile strength and flexibility and increased elongation at break of paper sheets in both machine and cross directions. Increasing the clearance gap from 30 to 60 microns resulted in increasing tensile strength in both machine and cross directions; this could be due to increasing the thickness of dry film formed [25]. But increasing the clearance gap to 90–120 microns resulted in a decrease in both tensile strength and Young's modulus. This could be due to the effect of water in the coating mixture which is higher in the case of using higher amounts of the mixture and the known negative effect of wetting and drying of paper sheets [26].

**Water Absorption of Paper Sheets Coated with Acrylate/NFC.** Water absorption test, which measures the amount of water in grams absorbed by one meter square of paper, was carried out on paper sheets coated with different thicknesses. The results are shown in Figure 10. As it is clear from the figure, the coating of paper sheets with acrylate/NFC resulted in higher water absorption than uncoated sheets. The increase in water absorption of paper sheets as a result of coating could be due to the effect of wetting and drying of coated paper sheets. That effect was higher than the effect of the formed layer of acrylate/NFC, which was supposed to decrease water absorption. Water absorption was decreased as the thickness of the coated layer increased from 30 to 120 microns. At 120-micron clearance, water absorption of coated paper sheets was close to that of the uncoated paper sheets. When a thicker layer of acrylate/NFC was formed in the case of using 120-micron clearance, the effect of wetting/drying was counterbalanced by the effect of acrylate/NFC layer formed.

## 4. Conclusions

The addition of TEMPO-oxidized cellulose nanofibers to acrylate polymer can significantly improve the mechanical,

barrier, and thermal properties of acrylate polymer. The enhancement in these properties made the usage of this modified polymer in paper coating desirable. The coating of paper sheets with acrylate emulsion containing 15% oxidized cellulose nanofiber and comparing it with the uncoated sheets cause an efficient improvement in the mechanical properties of the coated paper and a decrease of water absorption as the thickness of the coated layer increase.

### Data Availability

The data used to support the findings of this study are included within the article.

### Conflicts of Interest

The authors declare that they have no conflicts of interest.

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### References

- [1] Q. Wang, D.-Y. Yang, Y.-J. Zhang, G.-H. Li, and B.-Q. Yang, "Preparation of emulsifier-free emulsion of fluorinated acrylate polymer and its application in upgrading paper's resistance properties," *Chung-kuo Tsao Chih/China Pulp and Paper*, vol. 34, no. 9, pp. 17–20, 2015.
- [2] J. Zou, Y.-D. Shen, H.-H. Wang, and G.-Q. Fei, "Synthesize anionic polyurethane/acrylate microemulsion prepared by in-situ surfactant-free emulsion polymerization and its waterproof for paper," *Journal of Functional Materials*, vol. 44, no. 3, pp. 428–431, 2013.
- [3] S. Ganguly, S. Mondal, P. Das et al., "Design of psyllium-g-poly(acrylic acid-co-sodium acrylate)/cloisite 10A semi-IPN nanocomposite hydrogel and its mechanical, rheological and controlled drug release behaviour," *International Journal of Biological Macromolecules*, vol. 111, pp. 983–998, 2018.
- [4] S. Ganguly, P. P. Maity, S. Mondal et al., "Polysaccharide and poly(methacrylic acid) based biodegradable elastomeric biocompatible semi-IPN hydrogel for controlled drug delivery," *Materials Science and Engineering: C*, vol. 92, pp. 34–51, 2018.
- [5] I. Espigares, C. Elvira, J. F. Mano, B. Vázquez, J. San Román, and R. L. Reis, "New partially degradable and bioactive acrylic bone cements based on starch blends and ceramic fillers," *Biomaterials*, vol. 23, no. 8, pp. 1883–1895, 2002.
- [6] H. Fei, N. Saha, N. Kazantseva, G. Wang, H. Bao, and P. Saha, "A strong and sticky hydrogel electrolyte for flexible supercapacitors," *AIP Conference Proceedings*, 2015, Graz, Austria, October 2016, 2015.
- [7] X. Mao, B. Ding, M. Wang, and Y. Yin, "Self-assembly of phthalocyanine and polyacrylic acid composite multilayers on cellulose nanofibers," *Carbohydrate Polymers*, vol. 80, no. 3, pp. 839–844, 2010.
- [8] J. Henschen, J. Illergård, P. A. Larsson, M. Ek, and L. Wågberg, "Contact-active antibacterial aerogels from cellulose nanofibrils," *Colloids and Surfaces B: Biointerfaces*, vol. 146, pp. 415–422, 2016.
- [9] N. Chitpong and S. M. Husson, "High-capacity, nanofiber-based ion-exchange membranes for the selective recovery of heavy metals from impaired waters," *Separation and Purification Technology*, vol. 179, pp. 94–103, 2017.
- [10] S. M. Arteta, R. Vera, and L. D. Pérez, "Hydrophobic cellulose fibers via ATRP and their performance in the removal of pyrene from water," *Journal of Applied Polymer Science*, vol. 134, no. 7, 2017.
- [11] M. Larssona, Q. Zhou, and A. Larssona, "Different types of microfibrillated cellulose as filler materials in polysodium acrylate superabsorbents," *Chinese Journal of Polymer Science*, vol. 29, no. 4, pp. 407–413, 2011.
- [12] F. Grüneberger, T. Künniger, T. Zimmermann, and M. Arnold, "Nanofibrillated cellulose in wood coatings: mechanical properties of free composite films," *Journal of Materials Science*, vol. 49, no. 18, pp. 6437–6448, 2014.
- [13] F. Grüneberger, T. Künniger, T. Zimmermann, and M. Arnold, "Rheology of nanofibrillated cellulose/acrylate systems for coating applications," *Cellulose*, vol. 21, no. 3, pp. 1313–1326, 2014.
- [14] K. Littunen, U. Hippi, L.-S. Johansson et al., "Free radical graft copolymerization of nanofibrillated cellulose with acrylic monomers," *Carbohydrate Polymers*, vol. 84, no. 3, pp. 1039–1047, 2011.
- [15] J. Yu, C. Wang, J. Wang, and F. Chu, "In situ development of self-reinforced cellulose nanocrystals based thermoplastic elastomers by atom transfer radical polymerization," *Carbohydrate Polymers*, vol. 141, pp. 143–150, 2016.
- [16] T. Saito, S. Kimura, Y. Nishiyama, and A. Isogai, "Cellulose nanofibers prepared by TEMPO-mediated oxidation of native cellulose," *Biomacromolecules*, vol. 8, no. 8, pp. 2485–2491, 2007.
- [17] E. A. Hassan, M. L. Hassan, R. E. Abou-zeid, and N. A. El-Wakil, "Novel nanofibrillated cellulose/chitosan nanoparticles nanocomposites films and their use for paper coating," *Industrial Crops and Products*, vol. 93, pp. 219–226, 2016.
- [18] E. A. Hassan and M. L. Hassan, "Rice straw nanofibrillated cellulose films with antimicrobial properties via supramolecular route," *Industrial Crops and Products*, vol. 93, pp. 142–151, 2016.
- [19] P. Lu and Y.-L. Hsieh, "Cellulose nanocrystal-filled poly (acrylic acid) nanocomposite fibrous membranes," *Nanotechnology*, vol. 20, no. 41, article 415604, 2009.
- [20] S. Boufi, H. Kaddami, and A. Dufresne, "Mechanical performance and transparency of nanocellulose reinforced polymer nanocomposites," *Macromolecular Materials and Engineering*, vol. 299, no. 5, pp. 560–568, 2014.
- [21] B. Soni, E.-B. Hassan, M. W. Schilling, and B. Mahmoud, "Transparent bionanocomposite films based on chitosan and TEMPO-oxidized cellulose nanofibers with enhanced mechanical and barrier properties," *Carbohydrate Polymers*, vol. 151, pp. 779–789, 2016.
- [22] L. Dai, Z. Long, Y. Zhao, B. Wang, and J. Chen, "Comparison of hydroxypropyl and carboxymethyl guar for the preparation of nanocellulose composite films," *Cellulose*, vol. 23, no. 5, pp. 2989–2999, 2016.
- [23] R. D. Andrade Pizarro, O. Skurtys, and F. Osorio-Lira, "Effect of cellulose nanofibers concentration on mechanical, optical,

- and barrier properties of gelatin-based edible films,” *Dyna*, vol. 82, no. 191, pp. 219–226, 2015.
- [24] A. Abdulkhani, J. Hosseinzadeh, A. Ashori, S. Dadashi, and Z. Takzare, “Preparation and characterization of modified cellulose nanofibers reinforced polylactic acid nanocomposite,” *Polymer Testing*, vol. 35, pp. 73–79, 2014.
- [25] E. Afra, S. Mohammadnejad, and A. Saraeyan, “Cellulose nanofibils as coating material and its effects on paper properties,” *Progress in Organic Coating*, vol. 101, pp. 455–460, 2016.
- [26] N. Lavoine, J. Bras, and I. Desloges, “Mechanical and barrier properties of cardboard and 3D packaging coated with microfibrillated cellulose,” *Journal of Applied Polymer Science*, vol. 131, no. 8, 2014.



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