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Research Article

Mechanical, Thermal, and Morphological Properties of Nanocomposites Based on Polyvinyl Alcohol and Cellulose Nanofiber from *Aloe vera* Rind

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This work was devoted to reinforcement of polyvinyl alcohol (PVA) using cellulose nanofibers from *Aloe vera* rind. Nanofibers were isolated from *Aloe vera* rind in the form of an aqueous suspension using chemimechanical technique. Mechanical characterizations showed that incorporation of even small amounts of nanofibers (as low as 2% by weight) had significant effects on both the modulus and strength of PVA. Tensile modulus and strength of PVA increased, 32 and 63%, respectively, after adding 2% of cellulose nanofiber from *Aloe vera* rind. Samples with higher concentrations of nanofibers also showed improved mechanical properties due to a high level of interfacial adhesion and also dispersion of fibers. The results showed that inclusion of nanofibers decreased deformability of PVA significantly. Dynamic mechanical analysis revealed that, at elevated temperatures, improvement of mechanical properties due to the presence of nanofibers was even more noticeable. Addition of nanofibers resulted in increased thermal stability of PVA in thermogravimetric analysis due to the reduction in mobility of matrix molecules. Morphological observations showed no signs of agglomeration of fibers even in composites with high cellulose nanofiber contents. Inclusion of nanofibers was shown to increase the density of composites.

1. Introduction

During past decades, a significant number of research works have been devoted to the fabrication of high quality polymeric composites derived from renewable resources. Particularly, production of polymeric composites using cellulosic natural fibers, obtained from plants, has attracted a great deal of attention. Natural fibers benefit from a number of properties including low density, low cost, renewability, good mechanical properties, biodegradability, and availability [1–4]. Such advantages signify their capability as promising replacements for synthetic fibers such as glass [5, 6].

Cellulose, a building unit for fibrous plant cells, is a main constituent of natural fibers. Cellulose is present in plants in the form of fiber bundles including nanosized individual cellulose fibers [7–10]. Cellulose nanofibers can be extracted from plants using different techniques such as (i) mechanical methods, (ii) chemimechanical methods, and (iii) enzymatic methods [8]. Among the aforementioned techniques, chemimechanical and enzymatic techniques are less energy intensive compared to mechanical extraction of nanofibers [11]. Many plants (wood, kenaf, flax, soybean, wheat straw, etc.) have been already used for the extraction of cellulose nanofibers [8, 12–18]. Extracted cellulose nanofibers have a very high aspect ratio and form a web-like network due to (i) creation of hydrogen bonds between fibers and also (ii) mechanical interlocking of the fibers. It is expected that a successful incorporation of cellulose nanofibers (with diameters of lower than 100 nm) in polymeric matrices would substantially contribute to their mechanical properties

due to very high stiffness (as high as 134 GPa for cellulose crystals), strength (up to 10 GPa) [19], and specific surface area of nanofibers. A number of thermoplastics such as starch, polyvinyl alcohol (PVA) [12, 19], polylactic acid (PLA) [14, 20], and even commodity thermoplastics such as polyethylene (PE) and polypropylene (PP) [8] have been tested to fabricate nanocomposites with superior mechanical properties. Thermosetting polymers such as phenolic resins [21] and epoxy [22] with biological or petroleum origins have also been considered for such purposes. Such composites are considered to have numerous applications in the automotive and construction industries as well as in mobile electronics and other products.

However, previous research works have proven that composites reinforced with cellulose nanofibers have only shown moderate mechanical performance. It is believed that the strong hydrophilic behavior of cellulose nanofibers (due to the presence of a high concentration of hydroxyl groups), which causes low interfacial adhesion with hydrophobic polymers, is responsible for the unsatisfactory performance. Furthermore, creation of hydrogen bonds between –OH groups in adjacent cellulose nanofibers results in severe agglomeration in the fibers which limits their specific active surface area significantly [8, 13]. Very high levels of energy are required to overcome such agglomeration and create good dispersion of nanofibers in the matrices. As a result, cellulose nanofibers from chemimechanical extraction technique are usually kept in the form of aqueous suspension. Subsequently, the composites are prepared via solution casting method using water soluble polymers (such as PVA) as the matrix [8, 13].

Aloe vera (*Aloe barbadensis* Miller) is widely cultivated around the world for its gel, while its rind is treated as fertilizer or waste. In a previous study performed in our group [23], cellulose nanofibers (with diameters of generally lower than 20 nm) were extracted from *Aloe vera* rind using chemimechanical techniques and prepared in form of homogenized and stable water suspension. Characterizations of *Aloe vera* rind showed promising properties, especially high proportion of α-cellulose (57.72% ± 2.18%) [23]. In this study, cellulose nanofibers derived from *Aloe vera* rind were utilized as a novel reinforcement for polyvinyl alcohol using solvent casting technique. Composites with different concentrations of cellulose nanofiber (2, 5, 7, and 10% by weight) were prepared and their properties were compared with pure PVA. Mechanical (tensile tests), thermal (dynamic mechanical analysis and thermogravimetric analysis), and morphological (scanning electron microscopy) properties of the composites as well as their densities and transparency were evaluated.

2. Materials and Methods

2.1. Materials. Fresh *Aloe vera* leaves were provided by Southern Fields Aloe Inc., South Texas, and used as raw material for extraction of cellulose nanofibers. Polyvinyl alcohol (Elvanol 71-30 from DuPont) was used as matrix in all composites. PVA is a water soluble and biodegradable polymer and is considered as a good candidate for fabrication of composites containing cellulose nanofiber.

2.2. Isolation of Cellulose Nanofibers. Isolation of cellulose nanofibers from *Aloe vera* rind was performed according to Cheng et al. [23] with slight modifications. First, dried rinds were boiled in water for 2 to 3 hours and their wax cuticle was removed. The rest of *Aloe vera* rind was ground and then treated using 0.01 N HCl solution at 70°C for 2 hours to remove the extractives. Then ammonium hydroxide was used to adjust the PH value of the mixture to 9.5 to remove fat and pectin. The mixture was washed with distilled water to be neutralized and was air dried. The sample was then bleached in a 2% acidified sodium chlorite solution (solid/liquid: 1/20 v/v) for 4.5 hours at 50°C. Ratio of glacial acetic acid/sodium chlorite was 1/1 (v/w). A Masuko commercial grinder was then used to defibrillate the fibers (30 passes) and produce a nanofiber suspension.

2.3. Preparation of PVA/Cellulose Nanofiber Composites. First, PVA powder was dissolved in hot water (80°C) by continuous stirring for one hour using a magnetic stirrer. After dissolution of PVA in water, different amounts of aqueous suspension of cellulose nanofiber (with concentration of 0.6 wt.% of nanofiber) were added to the solution in order to produce composites with 2, 5, 7, and 10 wt.% cellulose nanofiber. Stirring was continued for one hour at room temperature. After stirring, ultrasonication was performed on the solutions for one hour for removal of air bubbles and also further dispersion of the nanofibers. The solutions were cast in Petri dishes and left to dry at room temperature for 120 hours, followed by drying in oven at 50°C for 24 hours. Transparent and flexible nanofiber reinforced films with average thickness of 200 μm were obtained (as shown in Figure 1). The composites containing nanofiber were coded according to their fiber content on a weight basis. For instance, PVA/NC(5) denotes composite film with 5% nanocellulose in PVA matrix.

Slightly yellowish color in the composites with higher concentrations of cellulose nanofiber (as shown in Figure 1) was due to presence of a slight amount of residual organic materials such as lignin [12]. Jonoobi et al. [14] reported the presence of white spots in their composites which they ascribed to aggregation of nanofibers. Figure 1 clearly shows that such spots have not been observed in our composites.

2.4. Tensile Tests. Tensile properties of the nanocomposite films were measured according to ASTM D638, type V. Tension tests were performed at a crosshead speed of 1 mm/min on an Instron model 3367 with a 2 kN load cell at room temperature (23°C). The data reported are Young's modulus, tensile strength, and tensile elongation at break. Each composition was tested with a minimum of five specimens to extract an average and standard deviation for each property.

2.5. Dynamic Mechanical Analysis (DMA). Dynamic mechanical analysis of the nanocomposites (5 mm wide and

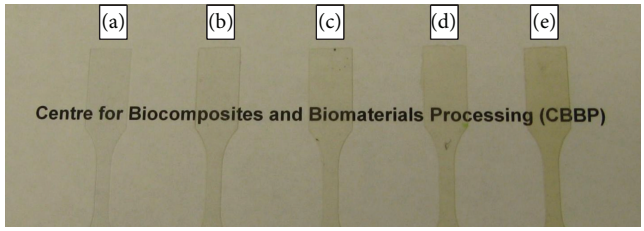

Figure 1: Appearance of fabricated nanocomposite films with (a) 0%, (b) 2%, (c) 5%, (d) 7%, and (e) 10% cellulose nanofiber contents.

24 mm long) was performed using a DMA Q800 (TA Instruments) in tensile mode. Measurements were performed at a constant frequency of 1 Hz and strain amplitude of 0.05% on a minimum of three specimens for each sample. Temperature of the specimens was raised from 30 to 150°C at a constant heating rate of 3°C/min.

2.6. Thermogravimetric Analysis (TGA). Thermal stability of the composites was investigated using a TGA Q500 (TA Instruments) at a heating rate of 10°C/min from 50 to 600°C. The tests were performed in nitrogen atmosphere.

2.7. Scanning Electron Microscopy. Scanning electron microscopy (SEM) was used to study the morphology of the compounds. After tensile tests, fractured surfaces of the composite films with different concentrations of cellulose nanofibers were coated with gold. Then, a Quanta FEG 250 scanning electron microscope was used to take the micrographs at a voltage of 10 kV.

2.8. Density Measurements. In order to measure the density of composites, the volume (cm^3) and mass (g) of each compound were carefully measured and the ratio of mass/volume was reported. For each compound, a minimum of five specimens were used to extract an average and standard deviation for densities of the films.

3. Results and Discussion

3.1. Tensile Tests. Figure 2 presents the stress-strain curves of PVA and its nanocomposites under tensile tests. Tensile characteristics of the samples have also been presented in Figures 3–5.

Tensile moduli of PVA and PVA/cellulose nanofiber composites are presented in Figure 3. It is shown that significant improvement in the tensile modulus of PVA was achieved via inclusion of even small amounts (2%) of nanofibers in sample PVA/NC(2). Tensile moduli of PVA and PVA/NC(2) were 4.26 and 5.63 GPa, respectively (32% increase). This behavior can be explained by creation of strong hydrogen bonds between cellulose nanofibers and PVA which contributes to good dispersion of nanofibers in the matrix and causes superior load transfer from the matrix to reinforcement. Further increase in concentration of the nanofibers up to 10% also increased stiffness of composites substantially. Tensile moduli of PVA/NC(7) and PVA/NC(10) were 7.16 and 7.99 GPa, which show 68% and 88% improvement over pure PVA. Such observations prove that quality of adhesion between the reinforcements and matrix and also level of dispersion of nanofibers in the matrix were good even at high concentrations of fibers.

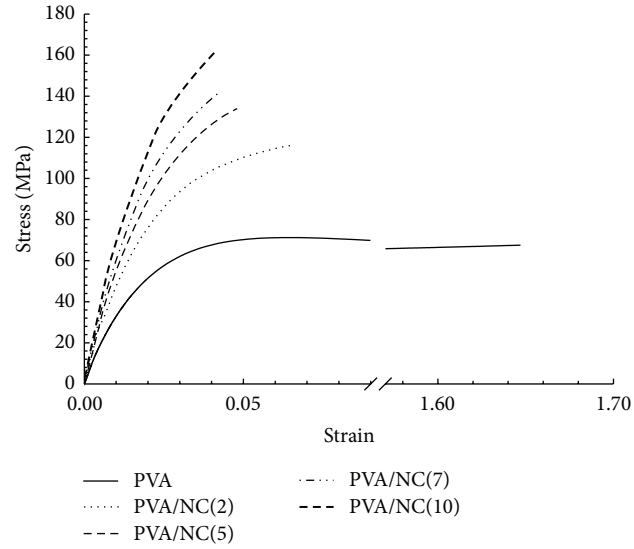

Figure 2: Tensile stress-strain curves for PVA and its nanocomposites.

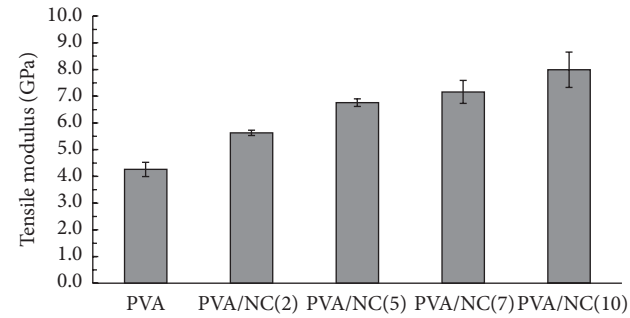

Figure 3: Tensile modulus of PVA and PVA/cellulose nanofiber composites.

Figure 4 presents the effects of inclusion of cellulose nanofibers from *Aloe vera* rind on tensile strength of PVA based composites. Tensile strength of composites is shown to have a similar trend compared to their elastic modulus. Inclusion of 2% nanofiber increased the tensile strength by 63%. Higher concentrations of nanofibers improved the strength even more significantly. Tensile strengths of PVA/NC(7) and PVA/NC(10) were 101% and 125% higher than strength of PVA, respectively.

Qua and coworkers [12] produced PVA/cellulose nanofiber composites using nanofiber from flax fiber and microcrystalline cellulose (MCC). They reported that inclusion of 5% of cellulose nanofiber resulted in only a modest increase in strength of composites (from 141.8 MPa for PVA to 152.17 and 149.71 MPa for composites with MCC and flax nanofibers, resp.). The differences in the reinforcing effects of

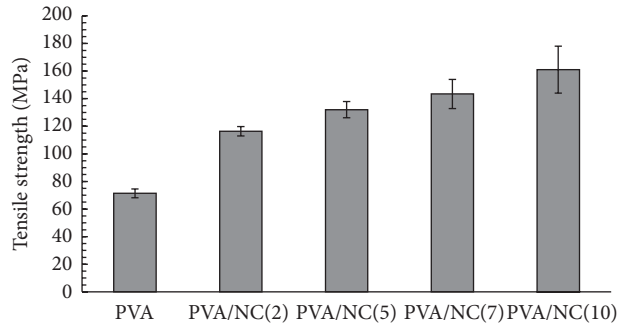

Figure 4: Tensile strength of PVA and PVA/cellulose nanofiber composites.

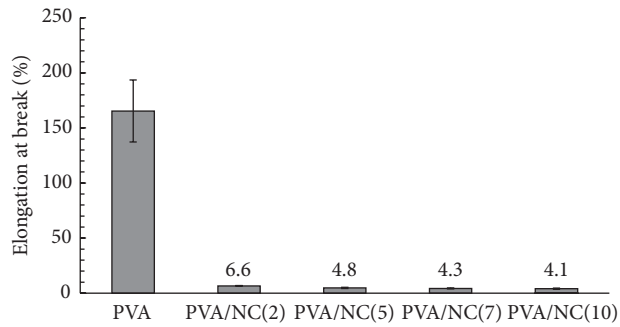

Figure 5: Tensile elongation at break of PVA and PVA/cellulose nanofiber composites.

their nanofibers and *Aloe vera* nanofibers, used in this work, could be ascribed to (i) different processing techniques and conditions for isolation of the fibers and also (ii) the fact that cellulose nanofibers from different plants have different characteristics.

Figure 5 shows that incorporation of the nanofibers resulted in severe reduction in elongation at break of polyvinyl alcohol. Sample PVA/NC(2) had an elongation at break of 6.6% compared to 165% for pure PVA. Further increase in concentration of nanofiber led to even more significant decrease in deformability to as low as 4.1% for composite with 10% nanofiber content. Low deformability of composites under tensile loads can be ascribed to high level of adhesion between the matrix and cellulose nanofibers. Since the nanofibers are not as deformable as the matrix [23], the strong interactions between the fibers and matrix molecules do not allow PVA to elongate.

3.2. Dynamic Mechanical Properties. Dynamic mechanical properties of PVA and PVA/cellulose nanofiber composites are measured to investigate their behavior in a range of temperature. Storage modulus (E') of samples is presented as a function of temperature in Figure 6. The figure shows that, at low temperatures, dynamic mechanical properties of composites are in good agreement with their tensile properties. Increase in concentration of cellulose nanofibers up to 10% increased storage modulus of PVA. At 30°C, storage

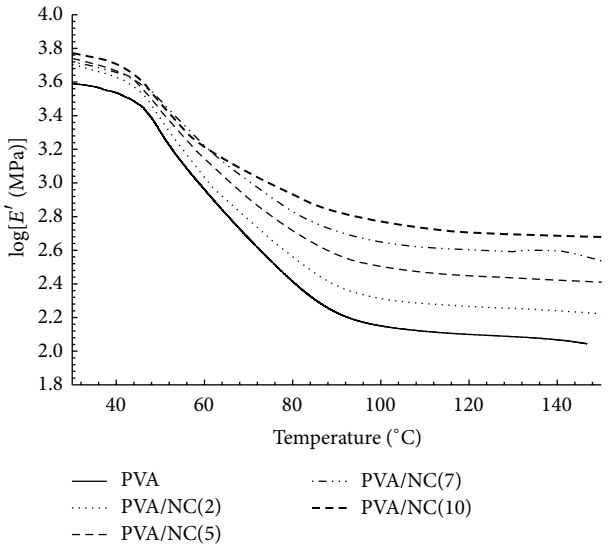

Figure 6: Storage modulus of PVA and PVA/cellulose nanofiber composites as a function of temperature.

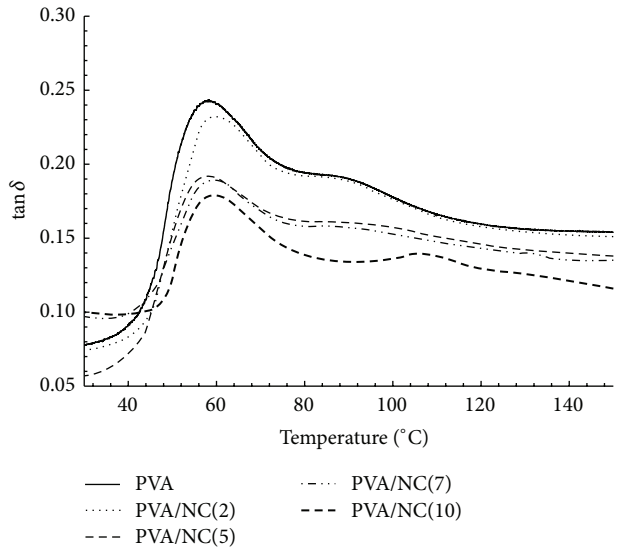

Figure 7: tan δ of PVA and PVA/cellulose nanofiber composites as a function of temperature.

modulus for PVA and composites with 5 and 10% nanofiber were 3899, 5494, and 5907 MPa, respectively.

Study of storage modulus of the samples at elevated temperatures yielded interesting results. It is shown that, at high temperatures (higher than around 70°C), the differences between storage modulus of samples with high and low concentrations of nanofiber were more noticeable. At 100°C, for instance, the storage moduli of PVA, PVA/NC(5), and PVA/NC(10) were 142, 319, and 589 MPa, respectively. In other words, the reduction in modulus of samples, due to increase in temperature, was more significant for samples with low nanofiber contents. This observation proves that at high temperatures (higher than glass transition temperature

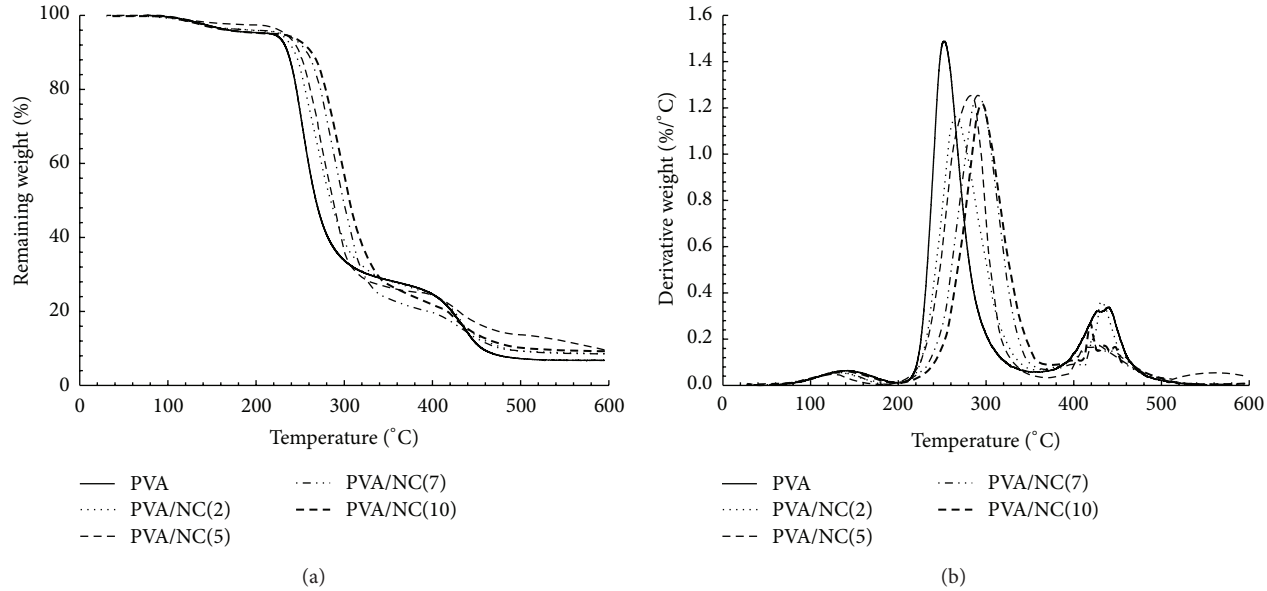

Figure 8: (a) TGA and (b) DTG thermograms for PVA and PVA/cellulose nanocomposites.

of PVA) the physically entangled network of nanofibers in the composites bears a significant amount of load [14].

Figure 7 shows the "tan δ versus temperature" curves for PVA and the nanocomposites. The peak in "tan δ versus temperature" curves represents the glass transition temperature (T_g). It is shown in Figure 7 that addition of 2% of nanofiber resulted in a slight increase in glass transition temperature of PVA (from 58.1°C to 60.4°C). This is ascribed to restrictions in motion of PVA molecules due to the presence of well-dispersed nanofibers. T_g of composites with 5, 7, and 10% nanofiber did not show a noticeable increase over composite with 2%. It is also shown that an increase in concentration of nanofibers resulted in reduction in peak value of tan δ.

3.3. Thermogravimetric Analysis. Table 1 shows the results of thermogravimetric analysis for PVA and PVA/cellulose nanocomposites in terms of $T_{\text{max.dec}}$ (maximum decomposition temperature) and T_{10} (the temperature for 10% mass loss). $T_{\text{max.dec}}$ represents the temperature at which the rate of thermal decomposition of the sample is at its peak.

Inclusion of cellulosic reinforcement is usually known to reduce thermal stability of composites with commodity thermoplastic matrices (polyethylene, polypropylene, etc.). It is shown in Table 1 that inclusion of cellulose nanofibers resulted in an improvement in degradation behavior (increase in degradation temperature) of nanocomposites which is ascribed to (i) higher thermal stability of nanofibers (due to separation of less stable components such as lignin and hemicellulose) and (ii) restriction of the mobility of polymer chains (in PVA) caused by homogenous distribution of cellulose nanofibers in the matrix [12]. For instance, T_{10} of PVA was 235.9°C while it increased to 243.7°C in the nanocomposite with 2% cellulose nanofiber. T_{10} of PVA/NC(10) was 256.1°C.

Table 1: TGA results for PVA and PVA/cellulose nanocomposites.

Sample	$T_{\text{max.dec}}$ (°C)	T_{10} (°C)
PVA	252.2	235.9
PVA/NC(2)	265.7	243.7
PVA/NC(5)	282.5	248.7
PVA/NC(7)	293.2	253.0
PVA/NC(10)	295.6	256.1

Figure 8(a) shows a slight reduction in weight of PVA and nanocomposites around 100°C which is due to evaporation of the absorbed moisture. Thermal degradation of PVA and composites occurred in two stages: (i) 230–330°C and (ii) 390–450°C. It is shown in Figure 8(b) that decomposition temperature of PVA was increased after inclusion of cellulose nanofiber while decomposition rate decreased. It has been suggested that reduction in decomposition rate of composites after inclusion of nanofibers is also due to restriction of the mobility of polymer chains [12].

3.4. Scanning Electron Microscopy. Analyzing the SEM micrographs provides helpful information about distribution of reinforcing phase in the matrix. Morphology of PVA and PVA/cellulose nanofiber composites is presented in Figure 9. It is shown that, in case of composite containing 2% nanofiber, the fractured surface was smooth which signals good dispersion and homogeneity in composite. Even an increase in concentration of the nanofibers up to 10% did not lead to emergence of signs of aggregation of fibers in a micron-scale. In all cases, no signs of fiber pull-out could be spotted.

3.5. Density Measurements. Figure 10 shows the variation of densities for PVA and PVA/cellulose nanofiber composites

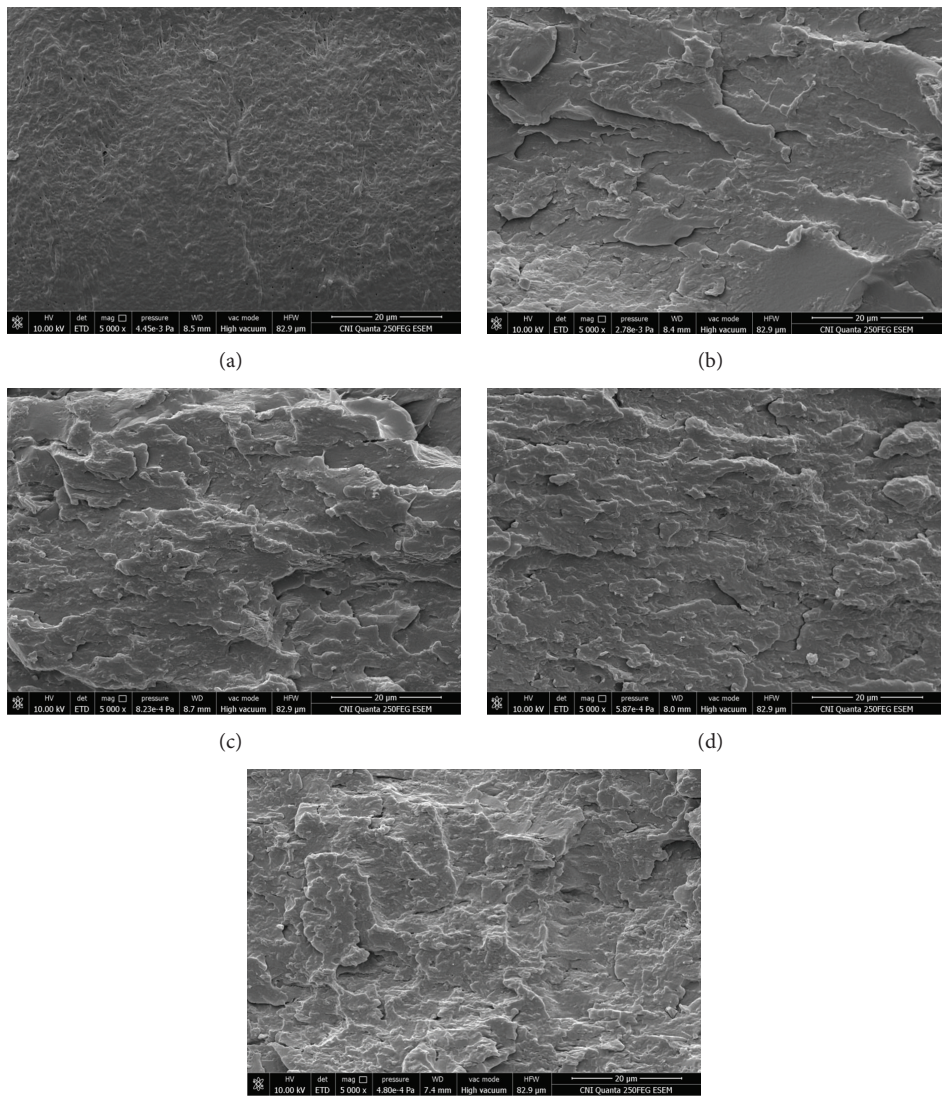

Figure 9: Scanning electron micrographs from (a) PVA and PVA/cellulose nanofiber composites with (b) 2, (c) 5, (d) 7, and (e) 10% nanofiber content with a magnification of 5000x.

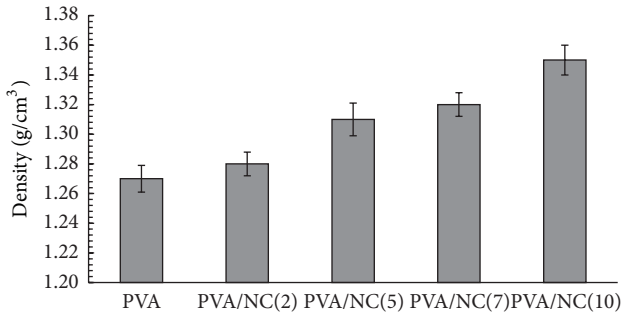

Figure 10: Density of PVA and PVA/cellulose nanofiber composites.

with different fiber contents. It is observed that an increase in content of cellulose fibers resulted in increase in the density of the composites (from 1.27 g/cm^3 for PVA to 1.35 g/cm^3 for composite with 10% nanofiber).

4. Conclusion

In this work, cellulose nanofibers from *Aloe vera* rind were used as novel bio-based reinforcements for polyvinyl alcohol. Cellulose nanofibers were extracted from the rinds using chemimechanical technique. Composites with up to 10% nanofibers were transparent while increase in concentration of the fibers caused a slight yellowish color in samples. Mechanical characterizations proved that even small amount of nanofiber (2% by weight) was significantly effective in improving tensile strength and modulus of PVA. Tensile strength and modulus of PVA increased, 63 and 32%, respectively, after inclusion of 2% of nanofiber. Both the mechanical

and morphological characterizations proved that increase in concentration of nanofiber in the composites (up to 10%) did not lead to any signs of agglomeration. Incorporation of even a small amount of nanofibers resulted in a reduction in deformability of PVA due to low elongation at break of fibers in comparison with PVA. The reduction was more noticeable in case of composites with higher fiber contents. DMA results revealed that the storage modulus of composites showed similar trend to their tensile modulus. It was also shown that the difference between storage modulus of PVA and the composites was more significant at temperatures higher than T_g. TGA results showed that the nanocomposites had higher thermal stability compared to PVA. Maximum decomposition rates for nanocomposites were lower than pure PVA. This behavior was attributed to the role of well-dispersed nanofibers in restricting the motion of the matrix molecules. It was also shown that an increase in content of nanofibers resulted in increase in densities of the composites.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgment

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