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Development and Characterization of Nanocellulose/ Carbonized Waste Rubber Nanocomposites

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Recycling is one of the most popular research topics today. In this study, in addition to the evaluation of waste tires, which are frequently encountered in the industry and difficult to dispose of, a green biomaterial, nanocellulose-based new generation nanocomposite was produced and characterized for the first time. Carbonized waste rubber, obtained by pyrolysis of tire wastes, was reinforced with nanocellulose at levels of 0.10%, 0.25%, 0.5%, and 1% by weight. The prepared nanocellulose-based nanocomposites were investigated by X-ray diffraction (XRD), morphological properties by scanning electron microscopy (SEM), thermal properties by thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), and dynamic mechanical thermal (DMTA). In addition, the percentage of gel contents of the produced nanocomposites were determined. Thermal analyses revealed that the sample containing 1% carbonized waste rubber showed the highest thermal stability and at 750 °C the ash yield increased up to 25% compared to nanocellulose. The fabricated nanocomposites had about 10 times higher storage modulus compared to pure NC. All results show that the green nanocellulose-based nanocomposites can be used for future applications in industry.

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INTRODUCTION

The disposal of non-biodegradable waste rubber tires is among the top issues for a recyclable world. According to the European Union, end-of-life tires are both a critical and valuable source of material for recycling. The alarming increase in the number of rubber tires produced today invites many serious problems for humans and the environment. Collecting waste rubber tires in one place or treating them as garbage is an economical and non-environmental solution. An economical, environmentally friendly, and smart solution is to recycle waste rubber tires as a raw material source in various applications (San Miguel *et al.* 2002; Chan *et al.* 2011; Gupta *et al.* 2013; Katarzyna *et al.* 2020; Lapkovskis *et al.* 2020; Surehali *et al.* 2022). A composite class whose components are at the nanometer level is called a nanocomposite. The reason why nanocomposites show superior properties compared to macro-sized composite materials is that one of their components has nano-sized properties. It is possible to improve the properties of composite structures, especially mechanical, thermomechanical, thermal, and electrical properties using a nanocomposite.

These advantages enable nanocomposites to find a place in many application areas (such as automotive, construction, defense, or medicine).

The nanostructured form of cellulose is called nanocellulose. Because of the sustainable feature of nanocellulose, it will be a type of nanomaterial with many uses in the future. In this context, research and development studies on nanocellulose composites are one of the most current issues of recent years. Nanocellulose can be used in rubber nanocomposites because of their sustainability, low energy consumption, low expansion coefficient, high aspect ratio, low cost, renewability, strength, and lightness.

The application of nanocellulose in rubber is limited in the literature. Most of the studies cover natural rubber material. Additionally, the use of nanocellulose as a matrix material in synthetic rubber nanocomposites has never been encountered before (Njuguna *et al.* 2008; Pasquini *et al.* 2010; Adeosun *et al.* 2012; Abraham *et al.* 2013; Cao *et al.* 2013a, 2013b; Chen *et al.* 2014; Foster *et al.* 2018; Gupta *et al.* 2018; Jardin *et al.* 2020; Dominic *et al.* 2020; Low *et al.* 2021; Yasin *et al.* 2021; Zor *et al.* 2024). Hancharoen *et al.* (2020) used bacterial nanocellulose and modified waste tire rubber for cement composite production. The sound absorption and heat insulation properties of the produced composite material were investigated. As a result, the thermal and sound properties of the composite structure were increased by reinforcing the nanocellulose and modified waste rubber together to the composite body. A triple composite structure containing styrene butadiene rubber and carbon black was designed using modified nanocellulose. Characterization processes confirmed the existence of interfacial interaction of the produced composites and the increase in the mechanical properties of the composite material (Xu *et al.* 2019). In another study, the incorporation of cellulose nanofibers into styrene butadiene rubber instead of carbon black was investigated. The resulting composite was compared with styrene butadiene rubber containing carbon black. The results showed that using cellulose nanofibers to reinforce styrene butadiene rubber was much more effective than carbon black (Sinclair *et al.* 2019). Unlike styrene butadiene rubber, bacterial cellulose fluff was added to the carboxylated acrylonitrile-butadiene rubber. The increase in the thermal and mechanical properties of the produced composite structure was remarkable (Wang *et al.* 2014).

When producing composite materials from cellulose and cellulose derivatives with standard polymer processing technologies, rubber-based components can be easily incorporated into the structure. However, when bringing them together, the interfacial adhesion of the matrix material and the reinforcement is considered. In general, these non-polar components show hydrophobic properties (Mohanty *et al.* 2000; Haydaruzzaman *et al.* 2009; Xu and Li 2012). For this reason, many modification techniques are used to increase the interfacial adhesion of the components. In general, modification strategies fall into two categories: physical treatment and chemical treatment. Lignocellulosic nanomaterials are widely used in electronics, food coatings, and biomedical applications as continuous fibers and additives in paper-based and adhesives, in addition to textiles, batteries, supercapacitors and cosmetics. Many innovations, such as the limited applications of nanocellulose-rubber composites, the use of nano-sized cellulose as a matrix in composite structure, the elimination of the deficiencies in the characterization processes of nanocellulose, and the use of synthetic and waste rubber materials in composites by recycling, reveal the originality of this work.

This study aimed to develop biobased nanocomposite films to be used as coating materials using nanocellulose synthesized from carbonized waste rubber and biopolymer cellulose derivatives, which have found a place in almost all sectors in recent years, and to investigate the sustainability of the product on a commercial scale. The authors try to reveal the production of nanocellulose-carbonized waste rubber (NCCWR) nanocomposites, which are formed by combining nanocellulose and carbonized waste rubber (CWR) material obtained because of carbonization of recycled tire wastes. The crystallinity of nanocellulose-based nanocomposites was determined by X-ray diffraction (XRD). Thermal gravimetric analysis (TGA), differential scanning calorimetry (DSC), and dynamic mechanical thermal analysis (DMTA) were used to determine the thermal and thermomechanical properties of nanocomposites. Scanning electron microscopy (SEM) images confirmed congruences in the morphological structure of NCCWRs. In addition, the percentage of gel content of nanocellulose-based nanocomposites was determined.

EXPERIMENTAL

Materials

The nanocellulose used in this study was obtained from Istanbul University-Cerrahpaşa University Nanotechnology Laboratory. The nanocellulose was spray-dried with a laboratory spray dryer (BUCHI Mini Spray Dryer B-290, Switzerland) according to the procedures described by Peng *et al.* (2012a, 2012b). Nanocellulose was obtained from bleached softwood pulp using 2,2,6,6-tetramethyl-1-piperidin-1-oxyl (TEMPO method), which oxidizes some alcohol groups in the cellulose chain to carboxylic acids. The resulting nanocellulose consists of thread-like particles with a diameter of about 20 nm and a length of 1 micron (Candan *et al.* 2016). In addition, carbonized waste rubber material by pyrolysis method under high pressure was produced and supplied by ZBB GmbH (Germany).

Preparation of Nanocellulose/Carbonized Waste Rubber Nanocomposites

First, 5 g of nanocellulose was dispersed in 50 mL of distilled water with magnetic stirrer until a clear solution was achieved to obtain nanocomposites. The CWR was added to the solution at the rates indicated in Table 1 and kept in an ultrasonic bath for 30 min. The resulting mixture was dried in an oven at 40 °C for 24 h after pouring into a petri dish.

Table 1. Design of Experiments for the Nanocomposites

Formulation	Nanocellulose (g)	Carbonized Waste Rubber (%)
NC	5	-
NCCWR-10	5	0.10
NCCWR-25	5	0.25
NCCWR-50	5	0.50
NCCWR-100	5	1
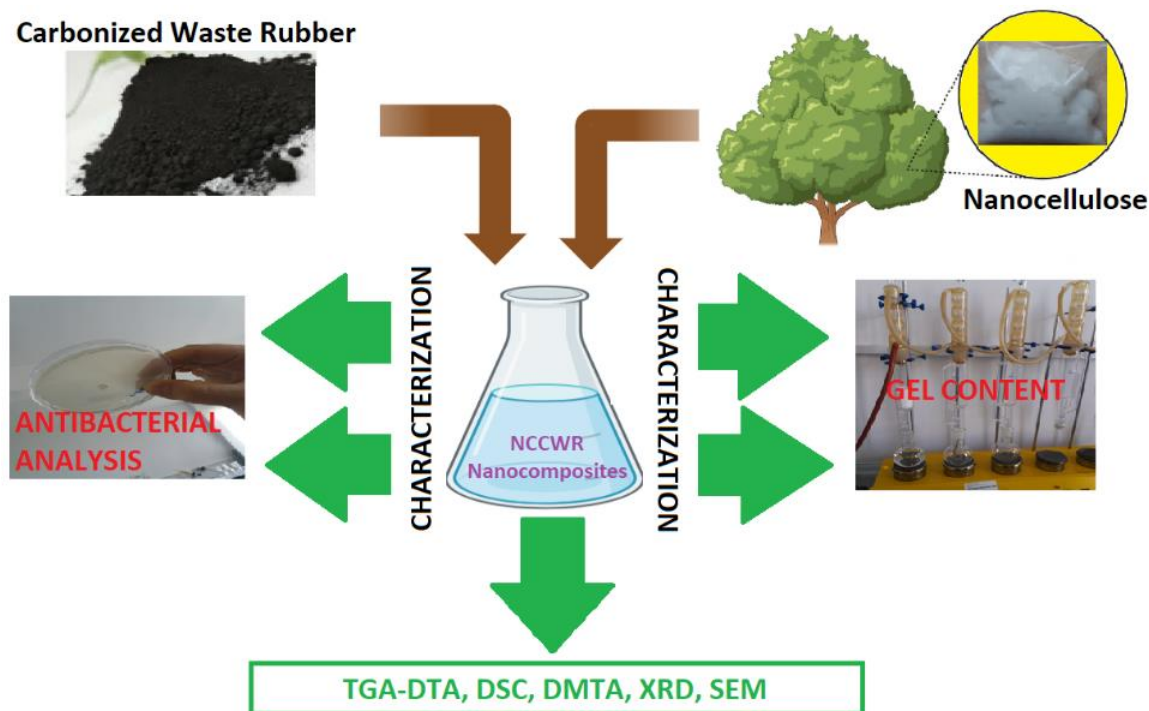
Fig. 1. The path followed to obtain NCCWR and the characterization of the obtained nanocomposites.

Measurements and Characterization

The XRD measurements were performed with a CuKα radiation ($n = 1.54$ Å) diffractometer between $2\theta = 10$ to $90°$ at 45 kV and 40 mA using the Rigaku SmartLab XRD device (Woodlands, TX, USA). The thermal properties of NCCWR nanocomposites were determined by TGA and DSC. The TGA analysis was performed using a Hitachi brand device (Tokyo, Japan). The thermal stability of the prepared NCCWR nanocomposites were determined from 20 to 750 °C in nitrogen environment, at a heating rate of 10 °C/min. The glass transition temperature (T_g) of the nanocomposites was investigated *via* DSC analysis. The DSC analysis was performed using a Hitachi DSC 7020 device. The analysis was completed in nitrogen atmosphere with a heating rate of 10 °C/min from 30 to 350 °C. The DMTA was used to determine viscoelastic properties. Analysis was performed on the HITACHI instrument. Thermomechanical properties were determined by measurements (between 30 °C to 250 °C, 1 Hz frequency, and 5 °C/min heating rate). Soxhlet extraction method was used to determine the gel contents of the nanocomposites. First, the test samples were cut and then samples were weighed. The cut and weighed nanocomposites were extracted with acetone for 6 h. The insoluble part after extraction was dried in an oven at 35 °C for 24 h. The dried nanocomposite samples were weighed again, and their percentage of gel content was calculated. The structure, shape, and physical properties (morphological structure) of the prepared nanocomposites were examined by scanning electron microscopy (SEM). A TESCAN (Brno, Czech Republic) device was used for analysis. The samples were coated with platinum (Sigma Aldrich, Burlington, MA, USA) before the analysis.

RESULTS AND DISCUSSION

XRD Patterns of NCCWR Nanocomposites

X-ray diffraction analyses were performed to reveal the crystallographic structure of NCCWR nanocomposites and the effect of this crystallinity on nanocomposites. The results are shown in Fig. 2. The NCCWR nanocomposites obtained with different waste carbonized rubber formulations had similar XRD patterns. For a film composed of nanocellulose, the recorded diffractions exhibited characteristic diffraction peaks at approximately 16.5°, 22.8°, and 34.3°, corresponding to typical cellulose reflection planes [110], [200], and [004], respectively (Singh *et al.* 2020). As shown in Fig. 2, the effect of carbonized waste rubber reinforced on nanocellulose-based nanocomposites on crystallinity was evident. The increase in the amount of carbonized waste rubber in NCCWR nanocomposites caused an increase in amorphous regions and a decrease in crystallinity. As seen in the SEM images, agglomerates in the nanocomposites support these results.
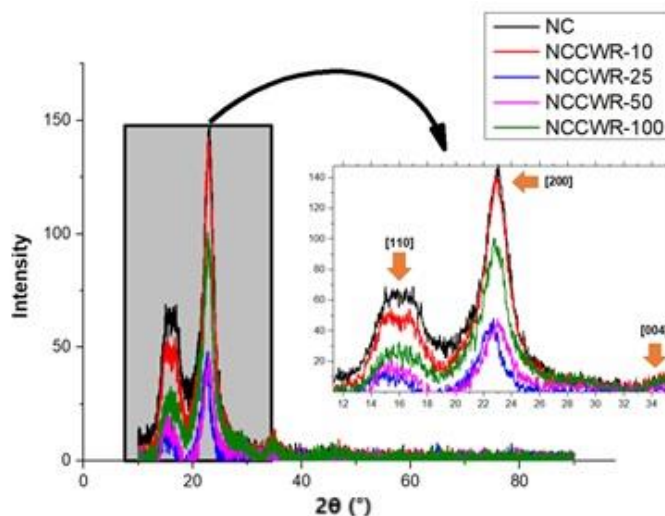

Fig. 2. XRD patterns of NCCWRs

Thermal Properties of the Nanocomposites

Thermogravimetric analysis/differential thermal analysis (TGA/DTA) was used to determine the thermal degradation stability of nanocomposites with nanocellulose matrix. The TGA analysis of cellulose-rubber composites is limited in the literature (Roy *et al.* 2021). TGA/DTA thermograms of nanocomposites are shown in Figs. 3 and 4, and datum from the thermograms are shown in Table 2. When TGA curves were examined, it was observed that all nanocomposite samples started to lose weight between approximately 49 and 71 °C. Losses at these temperatures are known to be due to evaporation of water molecules (Abdulkhani *et al.* 2020). As Fig. 3 shows, the main decomposition temperature generally occurred within the range 321 to 350 °C in a single step. The degradation at these temperatures is attributed to the degradation of the cellulose polymer network (Poyraz *et al.* 2017). When the literature is examined, the degradation temperature of nanocellulose is between 200 and 300 °C, showing that the results are consistent (Gan *et al.* 2020). Because the main degradation takes place in one step reveals that the nanocellulose matrix and fillers used in nanocomposite materials are compatible with each other and decompose

as a single material structure. The addition of CWR to the nanocellulose caused a partial increase in the main degradation temperatures. In contrast, as the amount of CWR increased in nanocomposite materials, a significant increase in ash yield was observed. While the ash yield of nanocellulose was 1% at 750 °C, this value increased to approximately 25% in nanocomposite material with 1% CWR. Ash yield prevents the flame from reaching the inner parts of the material in combustion events and thus contributes to the improvement of the non-flammability properties of the material (Birtane *et al.* 2021). When all the results were examined, the highest thermal stability was observed in the NCCWR-100 sample with 1% CWR additive.
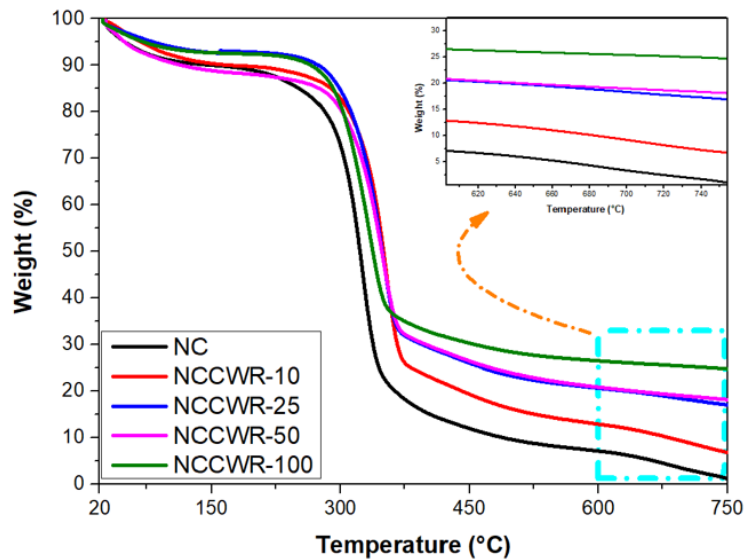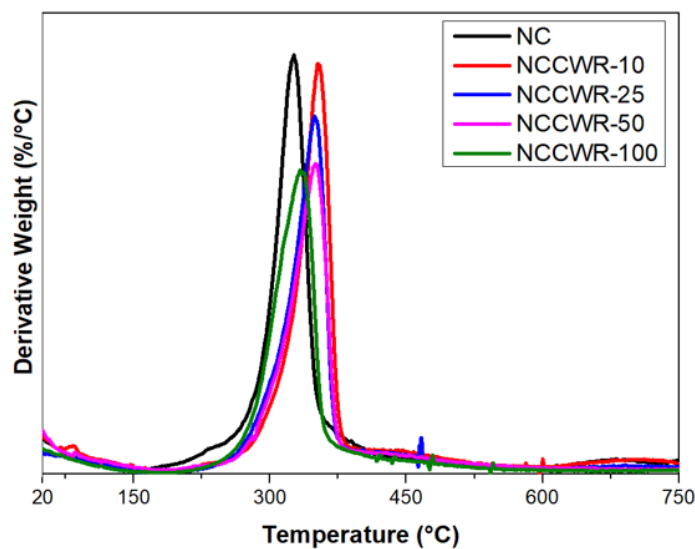
Fig. 3. TGA graph of NCCWRs

Fig. 4. DTA graph of NCCWRs

The DSC curve of nanocellulose-based nanocomposite films is shown in Fig. 5. Because the reinforcement material used was from waste products, it is difficult to distinguish thermal transitions as the amount of additive increases in nanocomposites, unlike unprocessed materials (Rajasekaran *et al.* 2021). Endothermic peaks are observed around 80 °C in nanocomposite film samples. However, the endothermic peak observed in the 1% carbonized waste rubber added F4 sample was around 60 °C. The addition of 1% by weight of carbonized waste rubber into the nanocomposite caused a decrease in the endothermic peak. The reason for this decrease is that the interaction between the hard and soft segments of the matrix and reinforcement materials is broken, leading to microphase separation in the matrix (Gan *et al.* 2020).

Table 2. Thermal Stability Characteristics of the Nanocomposites

Sample	$T_{10\%}$ (°C)	$T_{50\%}$ (°C)	Max. Weight Loss (°C)	Char Yield (%)
NC	106	321	326	1.21
NCCWR-10	130	350	354	6.84
NCCWR-25	274	348	350	17.01
NCCWR-50	116	348	352	18.16
NCCWR-100	265	337	338	24.78
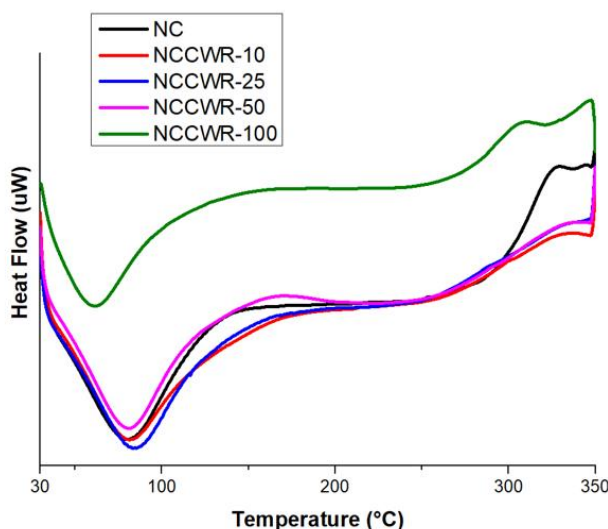
**Fig. 5.** DSC graph of NCCWRs

Viscoelastic Behavior of the Nanocomposites

The mechanical properties of nanocomposite films at different temperatures were determined by DMTA analysis. DMTA analysis of nanocellulose is limited in the literature (Gan *et al.* 2020). Figure 6 shows the DMTA results, and Table 3 shows the data from the results. The 0.50% carbonized waste rubber added NCCWR-50 sample had approximately 10 times higher storage modulus compared to NC. In general, it was observed that there was a large increase in the storage modulus with the increase in the amount of carbonized waste rubber in the composite. The effect of carbonized waste rubber added to the nanocellulose matrix on the storage modulus of nanocomposites was obvious. The same

was true for loss modulus values. When the loss modulus values were examined, it was seen that similar results were obtained with the storage modulus. When the tan delta values were examined, the sharp peaks observed in NC turned into flatter peaks with the addition of carbonized waste rubber. The free volume in the produced nanocomposites decreased with the contribution of the reinforcement element and causes the restriction of chain movements in the polymer matrix. This caused an increase in storage and damping modules (Mohsenzadeh *et al.* 2021). When all thermal results were evaluated, it was seen that the low thermal properties of nanocellulose provide superior performance with the effect of CWR doping.
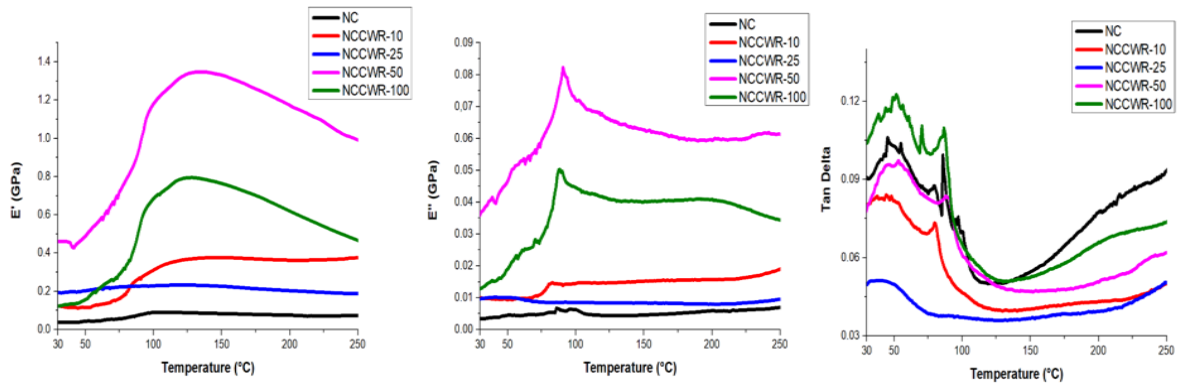
Fig. 6. Viscoelastic properties of NCCWR nanocomposite structures

Table 3. Viscoelastic Properties of the Nanocomposites

| Samples	Initial Storage Modulus 30 °C	E'(GPa) 50 °C	E'(GPa) 100 °C	E'(GPa) 200 °C	E'(GPa) Final Temperature
NC	0.02	0.04	0.08	0.08	0.08
NCCWR-10	0.12	0.11	0.30	0.36	0.37
NCCWR-25	0.20	0.21	0.23	0.21	0.18
NCCWR-50	0.47	0.49	1.18	1.17	0.99
NCCWR-100	0.14	0.15	0.60	0.62	0.47

Gel Content of the NCCWR Nanocomposites

When the results of the gel content test performed with the Soxhlet extraction method were evaluated. It was determined that the gel ratios of all nanocomposites were high. These values were calculated as NC 98.1%, NCCWR-10 84.4%, NCCWR-25 89.3%, NCCWR-50 90.9%, and NCCWR-100 99.2%. From this point of view, high gel ratio indicates that nanocomposite structures with high crosslink density were obtained.

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Morphologies of the Nanocomposites

Figure 7 shows SEM images of broken surfaces of nanocellulose and nanocomposites at different magnifications (x10000, and x20000). The oxidation performed by the TEMPO method had a direct effect on the fiber structure in terms of length and diameter. This effect varied according to the fiber source. This can be attributed to the initial properties, crystallinity, and chemical composition of the fibers (Serra-Parareda *et al.* 2021). TEMPO oxidation was part of a chemical-mechanical hybrid process. TEMPO keeps the partially amorphous regions intact (Onkarappa *et al.* 2020). As can be seen in Fig. 7, nanocellulose derived from TEMPO had a network structure with small nanofibers (Lin *et al.* 2012). A general problem with cellulose-based composites is a mismatch between the hydrophilic and hydrophobic natures of the fiber and the matrix. This often results in poor adhesion at the interface of hydrophobic composite matrix materials (Khalid *et al.* 2021). From this point of view, it has been observed that the addition of CWR to the nanocellulose structure influences the SEM images at different magnifications, and agglomeration occurs in the nanocomposite structure with the addition of CWR. As the amount of CWR in the composite structure increased, the composite structure moved away from the homogeneous structure form and became a heterogeneous structure. Moreover, uneven scattering increased within the structure.
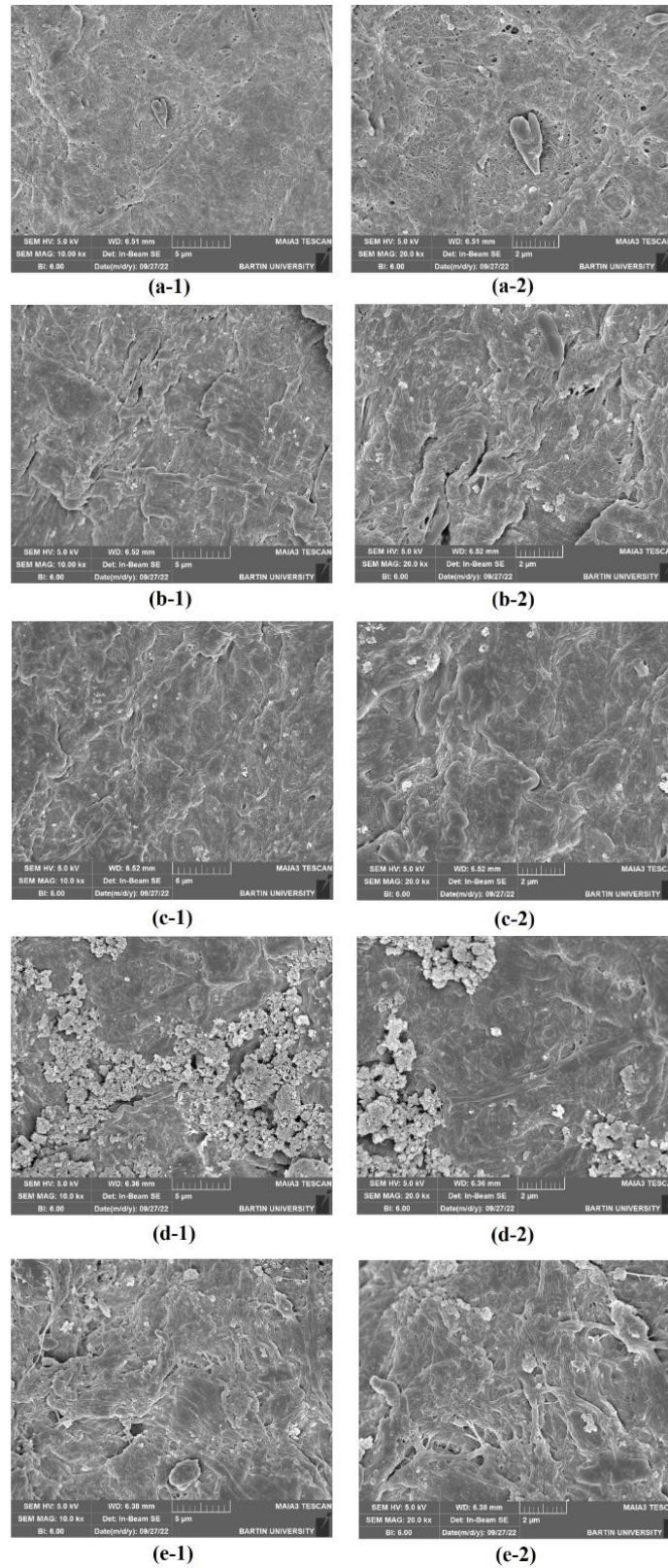
Fig. 7. SEM image of nanocellulose-based nanocomposites (a-1:NC at x10000, a-2:NC at x20000; b-1:NCCWR-10 at x10000, b-2:NCCWR-10 at x20000; c-1:NCCWR-25 at x10000, c-2:NCCWR-25 at x20000; d-1: NCCWR-50 at x10000, d-2:NCCWR-50 at x20000; e-1:NCCWR-100 at x10000, e-2:NCCWR-100 at x20000)

CONCLUSIONS

1. The X-ray diffraction (XRD) results showed that increasing the amount of carbonized waste rubber in nanocellulose-carbonized waste rubber (NCCWR) nanocomposites caused an increase in amorphous regions and a decrease in crystallinity.

2. According to thermogravimetric analysis/differential thermal analysis (TGA/DTA) results, the addition of carbonized waste rubber (CWR) to nanocellulose caused a partial increase in the main decomposition temperatures, and the highest thermal stability was observed in the NCCWR-100 sample with 1% CWR additive.

3. According to the differential scanning calorimetry (DSC) results, the addition of 1% by weight carbonized waste rubber into the nanocomposite caused a decrease in the endothermic peak, as the interaction between the hard and soft parts of the matrix and the reinforcement materials was disrupted, leading to microphase separation in the matrix.

4. According to DMTA results, NCCWR-50 sample with 0.50% carbonized waste rubber additive had approximately 10 times higher storage modulus compared to NC. Similar results were observed for loss modulus values.

5. It was determined that the gel ratios of all nanocomposites were above 85%.

6. According to SEM images, as the amount of CWR in the composite structure increased, the composite structure moved away from the homogeneous structure form and turned into a heterogeneous structure.

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