

## Research Article

# **Comparative Study of Polymer Composites with Cellulose Microfibers from Different Plant Resources**

Jyoti Giri<sup>(1)</sup>,<sup>1,2</sup> Rameshwar Adhikari<sup>(1)</sup>,<sup>2,3</sup> and Janak Sapkota<sup>(1)</sup>

<sup>1</sup>Department of Chemistry, Tri-Chandra Multiple Campus, Tribhuvan University, Ghantaghar, Durbarg Marg, Kathmandu 44605, Nepal

<sup>2</sup>Nepal Polymer Institute (NPI), P. O. Box 24411, Kathmandu, Nepal

<sup>3</sup>Research Centre for Applied Science and Technology (RECAST), and Central Department of Chemistry, Tribhuvan University, Kritipur, Kathmandu 44618, Nepal

<sup>4</sup>Institute of Polymer Processing, Montanuniversität Leoben, Otto Glöckel-Straße 2/III, 8700 Leoben, Austria

Correspondence should be addressed to Jyoti Giri; girijys@yahoo.com

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Wheat stalk (W), Fosro (F), Nigalo with waxy layer (NW), and Nigalo without waxy layer (NWo) were used to extract microcrystalline cellulose (MCC), the xMCC (where x represents origin such as W, F, NW, and NWo) by thermochemical and mechanical treatments. About 10 wt% of xMCC and commercial MCC (C-MCC) were solution casted with ethylene oxide-epichlorohydrin (EO-EPI) to prepare microcomposites. The xMCC and cryo-fractured composites were observed by scanning electron microscopy, and the mechanical properties of the composites were measured by dynamic mechanical analysis to observe the effect of fillers on viscoelastic properties. The results concluded that the xMCCs are homogeneously dispersed in the EO-EPI polymer matrix, which reinforced the viscoelastic and mechanical properties in EO-EPI composites, and reinforcement is dramatically high with NWoMCC compared to NWMCC, WMCC, FMCC, and C-MCC.

## 1. Introduction

The increasing trend of green practices in materials science encompasses the incorporation of low-cost and readily available natural fibers in polymer composites to enhance physical properties such as mechanical strength and stiffness [1–4]. The growing research interest in this area, both from academic and industrial viewpoints, in the development of cellulose-based composite materials is due to the abundance, renewability, and biodegradability of cellulose-based fillers. Several kinds of natural fibers, such as sisal, bamboo, wheat stalk, pineapple leaf, jute, cotton, kenaf, wood, etc., have already been investigated in this context [5–7]. Many researchers explored various fiber isolation processes from these resources, mentioned as controlled acid hydrolysis and mechanical processes [8–10]. The extracted cellulosebased fibers, including micro- and nanofibrillated cellulose and cellulose nanocrystals, have been demonstrated to be useful in the reinforcement of thermoplastics and thermosets for various applications [11–14]. Wood and cotton-based fibers represent the predominant industrial sources for extracting lignocellulosic fibers; however, the competition of using these sources for various other commodity applications may not have good satisfaction, and hence, other possible alternative sources have been explored [15, 16].

Depending on the origin of the raw material and the extraction process, the final morphology and, subsequently, the physicochemical properties of the extracted microcrystalline cellulose (MCC) differ [9, 17, 18]. Extraction of MCC from sugarcane bagasse [19], cotton stalks [19], rice straws and hulls [19, 20], oil palm's empty fruit brunch [21–23], bean hulls [20], fodder grass [24], and jute [25, 26] have resulted in shortening of fibers length forming aggregate structures on observation from scanning electron microscopy (SEM). The aggregated microparticles of cellulose with aspect ratios of ca. 1 are commonly referred to as MCC, whereas further disrupted nanostructures that have higher aspect ratios are referred to as nanocrystalline celluloses (NCCs).

These MCC and NCC from different origins have been in practice to reinforce the polymeric materials [16]. Xian et al. [27] reported that MCC in PLA increased tensile modulus and elongation at break from 206 to 262 MPa and 67.35 to 73.01 MPa, respectively. A similar trend was also mentioned by Mathew et al. [28], where MCC in PLA increased elongation at break and elongation modulus with increasing MCC content by 10%, 15%, 20%, and 25%, respectively. Li et al. [29] introduced NCCs from Avicel MCC to reinforce polyvinyl acetate. Similarly, Samarasekara et al. [30] also demonstrated that NCCs have a more reinforcing effect than the MCC when utilized to fabricate nanocomposites of polypropylene (PP). Ma et al. [31] also reported NCC reinforcement in tensile properties of the ethylene-co-vinyl acetate (EVA)/ NCC nanocomposites [31]. NCCs not only enhance mechanical strength but also induce other properties, such as optical transparency, to some polymeric systems compared to other microscopic fillers [32-34]. Although NCCs seem to have more reinforcing effects than MCC according to the literature [30, 35], NCC preparations are highly time, and energy consuming, at the same time, generally require strong and toxic chemicals too, which are difficult to separate from the fibers in their pure and dry state [36-38]. Therefore, the research needs to focus on overcoming the adversity with developing nanocomposites with NCCs, by selecting some smart low-cost, locally available, comparatively green noble materials that could enhance the viscoelastic mechanical properties of polymer composites to a comparable degree without nanofibrillation [39-41].

In this work, the MCC was extracted from agricultural residue wheat stalk (*Triticum aestivum*) and some other non-food competing fiber sources, such as Nigalo bamboo (*Drepanostachyam species*) and Fosro (*Grewia sclerophylla*), by using common fiber extraction processes, termed as chemical and thermomechanical process [16, 36, 42], which were then used as filler in the matrix of a rubbery polymer matrix, the ethylene oxide-epichlorohydrin (EO-EPI).

#### 2. Experimental Section

2.1. Materials. Sodium hydroxide (NaOH, 5 w/v% in H<sub>2</sub>O), sodium hypochlorite (NaOCl, 4%), sodium bisulfite (NaHSO<sub>3</sub>, 1% in H<sub>2</sub>O), and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, in 20% in H<sub>2</sub>O) were purchased from Qualigen Fisher Scientific, India and were used without further purification. Ethanol and triple-deionized water were purchased from Hangshu Chemical, Kangyuan, China, and Organo Laboratories, India, respectively. Different starting cellulose materials such as Nigalo bamboo shoots (Nigalo) of (*Drepanostachyam* sp.), Wheat straw (*T. aestivum*), refined fibrous wheat straw, and bast fibers of Fosro (*G. sclerophylla*) were collected from the surrounding forest of Kathmandu, to extract cellulose microfibers. For reference, MCC, the Lattice<sup>®</sup> NT-100 (named as commercial MCC (C-MCC) in this work) was purchased from FMC biopolymers. EO-EPI copolymer was used as a matrix and was obtained from Daiso Co. Ltd., Osaka, Japan (Epichlomer<sup>®</sup>, comonomer ratio = 1 : 1, density =  $1.39 \text{ g cm}^{-3}$ , weight-average molecular weight ( $M_w$ ) = ca.  $1 \cdot 106 \text{ g mol}^{-1}$ ).

2.2. Extraction of MCC. Fresh 6-month-old Nigalo shoots were chopped into pieces of ca. 5 cm in length, washed with water, and sun-dried for 10 days at approximately 28°C. From a part of the shoots, outer green and waxy layers (approximately 1 mm thick) were peeled off with the help of a knife. The peeled and unpeeled Nigalo shoots were further chopped to pieces of ca. 1 cm in length. Similarly, wheat straws were also chopped into 1 cm long pieces, washed, and sun-dried in similar conditions. The Fosro bast fibers were obtained by retting the outer bark of the plant [8, 43, 44]. The long white shining bast fibers were chopped into 1 cm long pieces, washed and sun-dried in similar conditions as other starting materials. All these chopped natural fibers were powdered properly with the help of a laboratory grinder. Each powdered fiber flour was sieved through to obtain the particles with a diameter of  $250\,\mu m$  or less.

The powdered fibers were treated with NaOH (5 w/v), solution ratio in 1:50) at  $60^{\circ}$ C with constant stirring for 3 hr to dissolve lignin and hemicellulose [45, 46]. The pulp obtained was neutralized to pH-7 with H2SO4 (5 w/v%) and washed with distilled water. Further, neutral fibers were steam exploded in a laboratory autoclave at 20 lbs pressure for 30 min, followed by bleaching with 4% NaClO solution at constant stirring conditions for 2 hr. Finally, 1% NaHSO<sub>3</sub> solution was added to the bleached fibers to remove excess of NaClO. The fibers were washed properly with distilled water thoroughly until the I<sup>2</sup> test for washing liquid did not produce any color. Then, the fiber flour was treated with 20% H<sub>2</sub>SO<sub>4</sub> solution, stirring in an ice bath for microfibrillation, maintaining the flour: acid (1:1)ratio for 20 min, washed properly with triple deionized water, and finally with ethanol. The cellulose powder thus obtained was dried in a vacuum oven at 80°C and designated as MCC (xMCC, x referring to the initial source from which the MCC was extracted).

2.3. Preparation of EO-EPI Copolymer Composites. The unfilled EO-EPI and EO-EPI composites (filled with 10 w/w% of extracted xMCC each) were prepared by established protocol [47, 48] via solution cast using dimethyl formamide as a common solvent. 5 w/v%) EO-EPI solution in DMF was mixed with 4 hr ultrasonicated (VWR ultrasonic cleaner, 180 W) xMCC solution (0.5 w/v%), mixed initially by ultrasonicating for 30 min and stirring overnight. The mixed solution was further sonicated for 30 min, followed by stirring for 30 mins and subsequently cast into a poly(tetrafluoroethylene) petri dish of a diameter of 120 mm. The solvent was evaporated initially in an oven at 70°C for 3 days and in a vacuum oven maintaining the same temperature at a pressure of 200 mbar. Obtained films were finally compression molded in a hot press at 70°C between Teflon sheets using a pressure of 4 bars for 3 min [49, 50]. Homogenous films of a thickness of ca  $200\,\mu\text{m}$  were finally obtained and were used for further characterization. As a reference material, neat EO-EPI and EO-EPI composites containing 10 w/w% of C-MCC were prepared using a similar procedure.



FIGURE 1: Illustrative photographs of the different plant sources of MCC studied: (a) bundles of wheat stalks, (b) Fosro, (c) Nigalo with outer waxy layer, and (d) Nigalo without outer waxy layer.

TABLE 1: Nomenclature of MCC isolated from different source
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S.no.	Sample code (xMCC code)	MCC source
1	WMCC	Wheat stalk fiber
2	NWMCC	Nigalo with an outer waxy layer
3	NWoMCC	Nigalo without the outer waxy layer
4	FMCC	Fosro fiber
5	C-MCC	Commercial MCC, Lattice <sup>®</sup> NT-100

2.4. SEM. The extracted xMCC, as well as Neat EO-EPI and microfiber-filled EO-EPI composites, were analyzed employing Tescan Vega II (Tescan Brno, Czech Republic) SEM at 5 kV. For the characterization of xMCC samples, a few mg of extracted powder were spread on the carbon tape fixed over the sample holder. Samples of Neat EO-EPI and EO-EPI composites were obtained by cryo-fracture for SEM analysis. All the samples were coated with gold for 100 s at 20 mA to achieve a coating thickness of approximately 3 nm.

2.5. Dynamic Mechanical Analysis (DMA). The unfilled EO-EPI and EO-EPI composites (filled with 10 w/w% of extracted xMCC each) films were characterized by DMA using TA instruments Q800. The samples for DMA analysis were obtained by cutting strips (ca 5.3 mm x 10 mm) from the compression molded films and analyzed over a temperature range between -70 and  $100^{\circ}$ C at a heating rate of  $3^{\circ}$ C min<sup>-1</sup> in tensile mode with a constant frequency of 1 Hz.

### 3. Results and Discussion

In this study, microfibers from new bio-based sources such as wheat stalk, Nigalo with and without outer waxy layer, and Fosro fiber were isolated. Composites were fabricated using these microfibers as well as a C-MCC, with EO-EPI polymer as a matrix by solution casting method. Figure 1 shows the raw resources of xMCC before extraction. Table 1 provides an overview of the nomenclature of xMCC from various starting materials used in this work. SEM images of the xMCC obtained after milling and hydrolysis are provided in Figure 2. The dimensions of the extracted xMCC are vital in determining their physical properties, more specifically,





(b)





(d)

FIGURE 2: SEM images of different forms of the extracted MCC: (a) WMCC, (b) NWMCC, (c) NWoMCC, and (d) FMCC with an expanded view of the indicated areas at the right.

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xMCC	WMCC	NWMCC	NWoMCC	FMCC	C-MCC
Length (ca. µm)	$25\pm5$	$287\pm5$	$300\pm 5$	$30\pm5$	108
Width (ca. $\mu$ m)	$5.6\pm1.5$	$6\pm1.5$	$4.7\pm2.5$	$4.9\pm1.2$	



FIGURE 3: DMA curves showing storage modulus as a function of temperature and for neat EO-EPI and EO-EPI/xMCC composites.

their reinforcing potential in yielding the mechanical properties [48–50]. The dimensional characteristics of the xMCC samples extracted from different sources using acid hydrolysis and retting are shown in Table 2. It can be seen that all extracted xMCC showed rod-like microparticles formed by the elongated fibrils, as reported by Giri et al. [38] for MCC obtained from WS. The average diameters are  $5 \pm 1.5 \,\mu$ m and  $25 \pm 5 \,\mu$ m long for wheat stalk (WMCC),  $6.0 \pm 1.4 \,\mu$ m for Nigalo fiber with outer waxy layer (NWMCC),  $4.7 \pm$  $2.5 \,\mu$ m for Nigalo fiber with outer waxy layer (NWMCC) and  $4.9 \pm 1.2 \,\mu$ m for Fosro fiber (FMCC). It appears that even the diameter of the MCC is similar no matter the origin of the source, most likely due to the application of a similar extraction process.

However, in all cases, it is evident that the xMCC are well-defined fibrils/fiber bundles, even though the degree of fibrillation are dependent on the material. From SEM images, it also appears that the surface roughness of WMCC is much higher compared to other xMCC, and this could just result due to a higher degree of fibrillation and breaking of the fibers during the extraction process.

3.1. Mechanical Properties of the Composites. The mechanical properties of the neat EO-EPI, EO-EPI/xMCC, and EO-EPI/ C-MCC composites prepared by solution casting, followed by subsequent compression molding, were characterized by DMA. The obtained mechanical data, particularly the storage modulus (E') and loss modulus (E'') data, are presented in

TABLE 3:	Glassy and	rubbery	modulus	of the	10w/w%	xMCC	and
EO-EPI	composites	calculate	d from Fi	gures 3	and 4.		

Materials	Glassy modulus E' at –60°C (GPa)	Rubbery modulus <i>E'</i> at 25°C (MPa)	
Neat EO-EPI	3.0	2.7	
EO-EPI/C-MCC	4.9	9.6	
EO-EPI/WMCC	5.8	40	
EO-EPI/FMCC	5.6	43	
EO-EPI/NWoMCC	7.0	227	
EO-EPI/NWMCC	6.5	63	

Figure 3 and Table 3. The values of the parameter are an average of at least five measurements for each sample.

The graphs presented in Figure 3 are plots of storage modulus (E') as a function of temperature at a constant filler loading of 10 wt% in EO-EPI. The increase in reinforcing properties of MCC was well explained by Sun et al. [51], where the addition of MCC up to 20% significantly increased the storage modulus by two folds and kept decreasing with increasing temperature. A similar trend was also observed for the composites prepared in this work.

The neat EO-EPI exhibits an E' of ca. 3.0 GPa at the glassy region (-60°C) and a drastic decrease of modulus around the glass transition region (ca. -25°C), the rubbery modulus for neat EO-EPI is observed around 3 MPa at 25°C. All composites containing constant filler loading of 10 w/w%



FIGURE 4: DMA curves showing the variation of Tan $\delta$  as a function of temperature for neat EO-EPI and EO-EPI/xMCC composites.

show a significant increase in E', both below and above the  $T_{\rm g}$ , when compared to the neat EO-EPI. The relative reinforcement is much more pronounced above the  $T_{\rm g}$ . For instance, the composite containing C-MCC shows a modulus of 4.9 GPa in the glassy region and 9.6 MPa in the rubbery region, which represents a 3.5 folds increase over the stiffness of neat polymer at room temperature.

The variation of Tan $\delta$  values in xMCC with increasing temperature is also clearly observed in Figure 4, following a similar trend as mentioned previously for E' and E''. A similar result is also presented by Selvakumar and Meenakshisundaram [52] in epoxy reinforced with jute and human hair and others [16, 41, 53]. Table 3 clarifies that glassy modulus (at  $-60^{\circ}$ C) is reinforced by all xMCC in almost the same ratio, i.e., ca.1.6  $\pm$  0.5 folds, but the significant reinforcement is observed by NWoMCC 7.0 GPa, i.e., 2.33 folds, and by NWMCC 6.5 GPa, i.e., 2.16 folds than the pure EO-EPI. Similarly, rubbery modulus at room temperature (25°C) is also found to be prominently increased by NWoMCC to 227 MPa, which is 84.07 folds than the pure EO-EPI, whereas NWMCC increased to 63 MPa, which is 23.33 folds than pure EO-EPI. The significant reinforcement by NWoMCC is due to the good compatibility of NWoMCC even in comparison to NWMCC. The removal of the waxy outer layer increases the compatibility of the NWoMCC. Table 3 indicates moderate reinforcement in the viscoelastic property, i. e., glassy modulus and rubbery modulus for WMCC, 5.8 GPa and 40 MPa, respectively. Such reinforcement is observed due to the stress transfer from polymers to natural fibers which are supposed to have good stress-holding capacity [52]. The results observed in this work are in agreement with previous works as well [54].

The dramatic increase in the glassy and rubbery modulus of the composites filled with xMCC suggests that the reinforcement could have resulted due to the multiscale distribution of filler based on nature. In Table 3, the dramatic increase in the rubbery modulus of EPO-EPI/NWoMCC indicates the high reinforcement due to NWoMCC. This directly correlates with the fact that the removal of the waxy layer of Nigalo increases its compatibility with hydrophilic MCC. As the waxy layer increases hydrophobicity and MCC being hydrophilic in nature, it will have less compatibility with Nigalo with waxy layer MCC (NWMCC) in comparison to Nigalo without waxy layer MCC.

As observed with the SEM images, different dimensions of the fibrils are present in the extracted MCC in contrast to the commercially available MCC, which is more homogenous. The presence of multiscale filler components is known to produce a synergistic reinforcement in the composites [47, 48, 55]. The composites with xMCC appear to be the hybrid morphology, which results in dramatic reinforcement. These results are in a similar trendline with recent studies by Bandera et al. [40], which demonstrated that the reinforcing capability of commercially available cellulose nanocrystals (prepared by fluidization or grinding commercially available MCC). Ramires et al. [56] discussed the similar reinforcing ability of treated MCC on the fact that the multiscale components of both micro and nanosized in the sample might have resulted in synergy in reinforcement. Several other studies have also demonstrated the synergy in mechanical reinforcement by using different fractions of micro and nanofillers in thermoplastic composites [57].

*3.2. Morphology of the Composites.* The microstructure of the neat EO-EPI and EO-EPI composites are characterized by SEM. The cryo-fracture surface morphologies of the composite films comprising 10 wt% of different MCC are shown in Figure 5. By comparing the EO-EPI/MCC composite with the micrograph of the EO-EPI/xMCC composites at a constant 10 wt% of filler loading, one can observe the differences.

The xMCC are well dispersed and covered by the matrix. Fiber pull-out is observed in all xMCC-containing samples,



(c)

FIGURE 5: Lower (left) and higher (right) magnifications of the SEM micrographs of cryo-fractured surfaces of samples: (a) pure EO-EPI, (b) EO-EPI/10 wt% WMCC composite, and (c) EO-EPI/10 wt% NWMCC composite.





FIGURE 6: Lower (left) and higher (right) magnifications of SEM micrographs of cryo-fractured surfaces of different samples: (a) NWoMCC composite; (b) FMCC composite; and (c) C-MCC composite.

showing that the resulting dramatic and unexpected reinforcement (Figures 5 and 6) could be due to the multiscale nature of the xMCC and stress transferred to xMCC on the application of stress on the composites. In addition, the role of long fibers is known to be more reasonable concerning the short ones in terms of stress transfers, which can result the better reinforcement. Higher stress concentration at the end of short fibers, such as NCCs, can also have a negative impact on the mechanical properties, such as tensile strength, elongation at break, etc., resulting in lower reinforcement than those by xMCC. It also appears that the fibers can be aligned within the composites as observed in NWMCC and other xMCCs with different roughness, which could also have resulted in the difference in mechanical reinforcement, likely due to improved physical bonding to the matrix.

Figure 6 shows the SEM micrographs of the cryo-fractured surface of the EPO-EPI/xMCC, which show good compatibility of fiber and matrix. The micrographs presented in Figure 6 reveals no fiber pull-out. No debonding is observed, which can be attributed to the fine adhesion between the microfibers and the polymer matrix [36, 38, 42]. The Fibers pulled out from the fracture surface are all covered with a matrix, indicating a good interface of compatibility. All other composites with various xMCC from different sources follow a similar pattern. This fine interfacial adhesion between the microfibers and the polymer matrix would result in an improvement in the mechanical properties of the nanocomposites; such phenomena are also observed in other works as well [36].

### 4. Conclusions

This work has introduced various kinds of plant fibers (such as wheat stalk, Fosro, and Nigalo) as good sources of MCC that can be used to reinforce the mechanical properties of polymers and rubbers such as EO-EPI. The results can be summarized as follows:

- (1) The chemical treatments applied on different fibers produced almost similar dimensions of the xMCC powder.
- (2) The solution casting of those fillers (by 10 wt% with) with the EO-EPI matrix produced homogeneous dispersion of the xMCC in the composites, as confirmed by electron micrographs.
- (3) The glassy modulus (E') and rubbery modulus (E'') both were enhanced by xMCC incorporation into the polymer matrix, indicating the reinforcement potential of the fibers.

The composite with 10% of the fillers (NWoMCC and NWMCC) was found to have a higher increase in storage and loss modulus values (E' and E'' values). At lower temperatures (such as at  $-60^{\circ}$ C), E' values enhanced by 2.33 and 2.16 folds, respectively, while the reinforcement in E'' values at 25°C was found to be 84.07 and 23.33 folds, respectively, higher than the neat polymer EO-EPI. It has been demonstrated that the xMCC obtained from different plant sources could be promising materials to enhance the mechanical properties of the EO-EPI.

## Data Availability

All data presented here are the research data which can be provided on request and are correct and compared with other researches which were given due respect in the reference section.

#### **Conflicts of Interest**

The authors declare that there are no conflicts of interest.

#### **Authors' Contributions**

All authors contributed to the present paper remarkably. JG formed the concept of the paper and prepared MCC from all plant fibers selected in this work. JG and JS performed the experimental works. JG wrote the first version of the paper. After consultation with the other authors, she revised the paper further. JS and RA reviewed the paper intensively. JG further improved the quality of the figures and schemes. JS supervised the work for compounding and characterizations. RA further supervised the whole work. The publication has been approved by all co-authors.

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