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Research Paper

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Comparative and Structural Investigation of SAMMAZ-14 Variety Maize Cob Nanocellulose Reinforced High- and Low-Density Polyethylene (HDPE & LDPE) Bio-Composites

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ABSTRACT

This research is to investigate the potential of waste agro -residue as reinforcement for polymers to produce composites as an alternative to synthetic fillers. Bio composites were produced using Sammaz-14 variety maize cobs Nano cellulose (MC-NC)/High Density Polyethylene (HDPE) / high-density polyethylene grafted maleic anhydride (HD-g-MAH) / organic montmorillonite (Org-MMT) and Sammaz-14 variety maize cobs Nano cellulose (MC-NC)/Low Density Polyethylene (LDPE) / Low-density polyethylene grafted maleic anhydride (HD-g-MAH) / organic montmorillonite (Org-MMT). The nanocomposites were compounded with a two-roll mill and fabricated using the horizontal injection molding machine. The structural properties of the composites were investigated. The crystallinity, crystallographic phase and d-spacing as well as dimensional stability of the composites were examined by XRD Effects and impact of alkaline (5 % NaOH) treatment was observed. The effect of the addition of 1 & 5 % of MC-NC with 2 wt % of Org-MMT and 3% maleic anhydride grafted polyethylene and 1 & 5 % Org-MMT in HDPE & LDPE were studied, From the results, 2% of Org-MMT was enough to improve the exfoliation and dispersion of the filler in the matrix and 3% incorporation of HD-g-MAH increased the adhesion and reduce the surface tension between the fiber and matrix. hence, creating excellent interfacial adhesion within the composite's structures. Further investigation confirmed the presence of functional interactions among HDPE/LDPE, MC-NC, Org-MMT, and HD/LD-g-MAH, suggesting enhanced compatibility and dispersion

Keywords - Bio-composites, High-Density Polyethylene, Ligh-Density Polyethylene, Maize cob Nano Cellulose, Organic Montmorillonite, polyethylene grafted maleic anhydride.

I. INTRODUCTION

The increasing environmental concerns associated with synthetic fillers and non-biodegradable reinforcement materials have intensified research into sustainable alternatives derived from agro-waste resources. Agricultural residues, particularly maize cobs, maize husk and maize stalk amongst others represent an abundant, renewable, and low-cost lignocellulosic biomass with significant potential for value-added applications Minerva et al (2026); Gupta et al (2025). The valorisation of such agro-residues into nanocellulose for polymer reinforcement offers a promising way toward the development of environmentally friendly and high-performance sustainable materials.

Polyethylene, High-Density Polyethylene (HDPE) and Low-Density Polyethylene (LDPE), remains some of the most widely used thermoplastics (commodity plastic) due to their excellent chemical resistance, availability, ease of processability, and cost-effectiveness Silva et al (2025). However, inherent limitations in stiffness, dimensional stability, and interfacial compatibility with hydrophilic fillers restrict their performance in structural applications Pradeep et al (2024). The incorporation of nano-scale fillers such as maize cob nanocellulose (MC-NC) and layered silicates like organic montmorillonite (Org-MMT) has been widely explored to enhance the mechanical, dimensional, and structural properties of polyethylene matrices.

Despite the potential improvements offered by nano-fillers, the interfacial incompatibility between hydrophobic polyethylene and hydrophilic cellulose-based reinforcements remains a critical challenge Mishra et al (2025) to overcome this limitation, compatibilising agents such as maleic anhydride grafted polyethylene (HD-g-MAH or LD-g-MAH) are employed to promote chemical interactions, reduce interfacial tension, and improve filler dispersion within the polymer matrix.

Furthermore, alkaline surface treatment (5 % NaOH) maize cob nanocellulose modifies the fibre surface by removing amorphous components and enhancing surface roughness, thereby improving interfacial bonding Mohammadi et al (2024); Baba et al. (2022). The structural behaviour of such multi-component nanocomposites—particularly crystallinity, crystallographic phase evolution, and interlayer spacing—can be effectively analysed using X-ray diffraction (XRD).

Hitherto, most maize cob nanocellulose studies used unspecified maize varieties. The composition of lignin, cellulose, hemicellulose varies significantly by agro-climatic zone and method of cultivation. Sammaz-14 maize is a high yield Nigerian maize variety released by IAR, ABU Zaria, Nigeria. The study establishes a baseline for locally abundant, agro-economically relevant waste for use as reinforcement in bio-composites, thus, making it directly useful for Nigerian and other part of the world. PE-g-MAH and Org-MMT were used to improve filler-matrix interaction as well as adhesion. in turns, add value to HDPE & LDPE by making it useful in structural applications.

This study therefore investigates the structural characteristics and interfacial behaviour of Sammaz-14 maize cob nanocellulose reinforced HDPE and LDPE nanocomposites containing organic montmorillonite and maleic anhydride grafted polyethylene. A comparative evaluation of HDPE- and LDPE-based systems was conducted to determine how matrix architecture influences crystallinity, filler dispersion, intercalation/exfoliation behaviour, and overall structural integrity of the prepared composites. These materials can be used in applications where strength, dimensional (according to the results reported by Mohammadi et al (2024); Baba et al (2020) and structural stability is paramount e.g., building and constructure as well as automotive interior.

II. MATERIALS AND METHODS

2.1 Materials

Nano clay (Org-MMT) 0.885 g/cm³, surface modified with trimethyl ammonium stearyl purchased from sigma Aldrich Germany, Maize cobs (SAMMAZ-14 Maize cob variety) obtained from Institute for Agricultural Research, Federal Ministry of Agriculture and Rural Development, Ahmadu Bello University, Zaria-Nigeria, Maize cob nano cellulose (isolated via 5 % NaOH pre-treatment, at 95 °C, for 4hr followed by ball milling with stainless steel ball of 10 mm to obtain MC-NC within the range 100 nm ± 5nm which has an onset temperature of 281 °C and degradation temperature of 386 °C). Polyethylene (HDPE) grade no: H16178M170306D and HD-g-

MAH (Orevac 18360), (LDPE) grade no. LDF-260GG-E190307E, and LD-g-MAH (Orevac 18360) purchased from Lotte Chemicals, Johor Malaysia.

2.2 Methods

2.2.1 Composite Fabrication

Interfacial compatibility between polymer matrices (HDPE, LDPE & grafted copolymer PE-g-MAH) and nanofiber (MC-NC) is critically important for manipulating nanostructure and various properties of the polymer bio composites. Thus, the need for proper compounding of these materials to achieve a good dispersion of the fillers in the matrix. Composites properties significantly depend on the dispersion of fillers particles in the polymer. In this research, maize cob nano fiber was utilized as functional reinforcing filler, Org-MMT was used as an additive to modify/ improve the properties of the matrix and enhanced the dispersion of the fillers in the matrix whereas, the PE-g-MAH was incorporated as a compatibilizer whose role is to create linkage or site for interaction/bonding between the phases i.e fibre and polymer and hence, provide fibre/matrix interfacial adhesion because of its available oxygen acceptor sites.

Table 1: Composite Formulations

S/ N	HDPE & LDPE (wt.%)	MC-NC (wt.%)	HD-g-MAH & LD-g-MAH (wt.%)	Org-MMT (wt.%)
1	100	0	0	0
2	94	1	3	2
3	94	2	3	1
4	90	5	3	2
5	90	2	3	5

2.2.2 Compounding/Mixing:

The compounding was done using a two-roll-mill (Prep-Mill C.W. Brabender mixer -Hackensack, N.J. PM-3030) with 150 g capacity at a temperature of 190 °C for HDPE and 160 °C for LDPE with a roller nip distance of 1.4 mm, melting 12g PE-g-MAH /10 min and 17g HDPE and LDPE / 10 min at a roller speed of 60 rpm for 20 min.

2.2.3 Crushing/Granulating:

Plastic granulator SLM, 50 FY (Wenzhou Zhingang co. Ltd China) was used for crushing the compounded samples to a smaller size, suitable for use on the mini plastic injection molding machine. The granulated or crushed samples were fabricated using the Nissei horizontal injection molding machine in the polymer processing lab at the Universiti Tun Hussein Onn Malaysia.

2.2.4 Fabrication/ Injection Moulding:

The crushed samples in the form of pellets were fed into the Nissei horizontal Injection Molding machine (NISSEI, NP7Real Mini), Japan) whose temperature was set at (HDPE)140 °C, 160 °C and 190 °C, and (LDPE) 120 °C, 140 °C and 160 °C (for the hopper, barrel i.e heating and mixing chamber and the nozzle temperature respectively) after a complete cycle, the moulded samples were ejected from the mould cavity by the pins. Molding was carried out in a semi-automatic mode, the complete processes happened in approximately 10min/cycle.

2.2.5 Conditioning of Composites:

The fabricated samples were conditioned according to ISO 291 before characterization (XRD).

2.2.6 X-Ray Diffraction (XRD) Analysis/ Procedure for HDPE & LDPE-Composites:

The crystalline structure of the prepared samples was studied using a wide-angle X-ray diffraction (WAXRD) powder diffractometer (RIGAKU-smart lab Japan). The analysis was carried out using CuK α radiation source at 40 kv and 30 mA and wavelength of 1.54Å. The samples were kept on a sample slate and placed under the radiation source with the above conditions. The crystallinity index and crystallographic phase of sample was examined by XRD. Effects and impact of the alkaline treatment and acid depolymerization processes on the

crystallinity index of the materials were investigated from the generated X-ray diffractograms. The X-ray diffraction patterns were recorded from 10° to 60° . To identify the crystalline, amorphous and d-spacing of the bio-composites, the prepared composites were crushed into smaller sizes of about 7 micrometres.

III. RESULTS AND DISCUSSION

3.1 X-Ray Diffraction (XRD) Analysis of HDPE-Based Bio-Composites

X-ray diffraction (XRD) analysis was employed to assess the crystalline structure and phase behavior of polyethylene (HDPE and LDPE) based bio-composites incorporated with varying proportions of maize cob nanocellulose (MC-NC), organo-modified montmorillonite nanoclay (Org-MMT), and PE-grafted maleic anhydride (HDPE-g-MA and LDPE-g-MA). The diffraction patterns corresponding to Figures 1 provide insights into how the addition of organic and inorganic nanofillers influences the crystalline order and interlayer spacing of the polymer matrix.

Figure 1. Shows XRD diffractogram for the pure HDPE. The non-destructive method was used in this study to characterized and get information on the crystal orientations, crystallinity and physical properties of the prepared composites. The d-spacing or lattice spacing (which is the minimum distance in Angstrom between the two atomic planes) Khalid et al (2025) values were also presented below. Each crystallographic structure has its own unique diffraction pattern and d-spacing.

From Figure 1, the XRD pattern of neat HDPE (100 %), which typically exhibits strong diffraction peaks at approximately 21.6° and 23.9° 2θ . These peaks correspond to the (110) and (200) crystallographic planes of the orthorhombic phase of HDPE, respectively Ruud & Russell (2018). Their presence and sharpness indicate a high degree of crystallinity and long-range order inherent to the semi-crystalline nature of HDPE. This pattern serves as the benchmark for evaluating the structural modifications induced by the addition of fillers and compatibilizer, which will be seen in the subsequent Figures.

Figure 2 presented the XRD graph/ pattern for composites containing 2 % maize cob nanocellulose (MC-NC), 1 % organo-modified Montmorillonite nano clay (Org-MMT), 3 % HDPE-grafted maleic anhydride and 94 % of High-density polyethylene (HDPE)

In this pattern, the HDPE crystalline peaks remain visible, although slight peak broadening and minor reductions in intensity are observed. This suggests a reduction in crystallinity due to the partial disruption of the HDPE chain packing by the introduction of nanofillers. A weak, low-angle reflection near $2\theta \approx 2-6^\circ$ may also be present, attributed to the basal spacing of the Org-MMT layers, indicating partial intercalation of polymer chains into the clay galleries Ruud and Russell (2018). The presence of HDPE-g-MA likely facilitates better filler dispersion, contributing to mild exfoliation or intercalation effects as depicted in the diffractogram.

Figure 3 corresponds to the XRD graph/pattern for composites of 2 % maize cob nanocellulose (MC-NC), 5 % organo-modified Montmorillonite nanoclay (Org-MMT), 3 % HDPE-grafted maleic anhydride and 90 % of High-density polyethylene (HDPE).

As depicted, the HDPE peaks are further broadened and reduced in intensity compared to Figure 2, reflecting more pronounced interference with polymer chain crystallization. The higher nanoclay loading intensifies the amorphous halo and possibly introduces a more prominent reflection at low 2θ values, consistent with increased interlayer distance and enhanced intercalation Saha et al (2021). However, the absence of peak disappearance indicates that complete exfoliation of clay layers may not be achieved at this loading, possibly due to agglomeration or incomplete compatibilization despite the presence of HDPE-g-MA in the prepared bio composites formulations.

The Figure presents the XRD profile for a composite with 1 % maize cob nanocellulose (MC-NC), 2 % organo-modified MMT (Org-MMT), 3 % HDPE-grafted maleic anhydride (HDPE-g-MA), and 94 % HDPE. In this formulation, the crystalline HDPE peaks are retained with relatively modest broadening, and no significant shift in peak position is observed. The degree of crystallinity appears better preserved than in Figure 3, likely due to the lower clay content and improved filler dispersion at reduced loading Saha et al (2021). Any observed low-angle peaks related to Org-MMT remain weak, suggesting partial intercalation. The relatively balanced filler

content in this system allows retention of semi-crystalline HDPE morphology while still inducing minor structural perturbations within the composites structure.

Figure 5 shows the XRD pattern of a composite composed of 5 % MC-NC, 2 % Org-MMT, 3 % HDPE-g-MA, and 90 % HDPE. The crystalline peaks of HDPE persist, although more diffused, with increased amorphous background scattering. The high content of MC-NC, a largely amorphous biopolymer (fibre), contributes to reduced overall crystallinity. The Org-MMT peak, if present, may appear slightly more pronounced than in lower MMT formulations, indicating partial intercalation rather than full exfoliation Saha et al (2021); Shelly et al (2024). The resulting pattern suggests that excess cellulose introduces greater disorder, reducing crystal growth and possibly forming hydrogen-bonded filler networks that restrict polymer chain mobility hence, possibly decreasing the mechanical strength of the prepared bio composites.

Across Figures 1 to 5 a progressive trend of decreased peak intensity and broadening is observed with increasing filler content, particularly MC-NC and Org-MMT. The interplay between nanofiller concentration and compatibilization governs the extent of crystallinity retention and structural reorganization in HDPE. While Org-MMT introduces lamellar structures that may intercalate polymer chains, the degree of dispersion critically influences whether exfoliation is achieved. Simultaneously, MC-NC introduces amorphous content that competes with crystalline HDPE domains, particularly at higher concentrations.

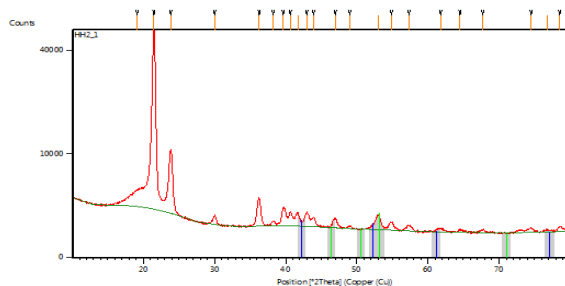


Figure 1. XRD Diffractograms for composites of 100 % High-density polyethylene (HDPE)

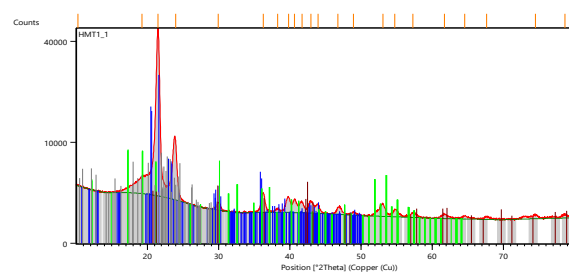


Figure 2. XRD Diffractograms Intensity of $^{\circ}2\theta$ for 94 % HDPE/MC-NC/ 1% Org-MMT and HDPE-grafted maleic anhydride Bio-Composites.

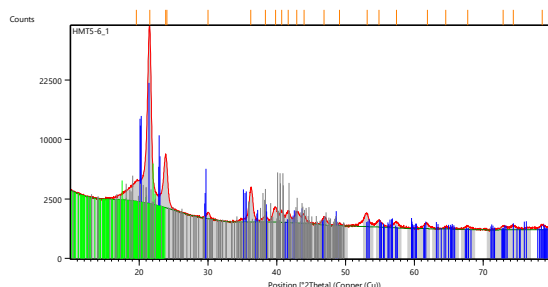


Figure 3. XRD Diffractograms Intensity of $^{\circ}2\theta$ for 90 % HDPE/MC-NC/ 5 % Org-MMT and HDPE-grafted maleic anhydride Bio-Composites.

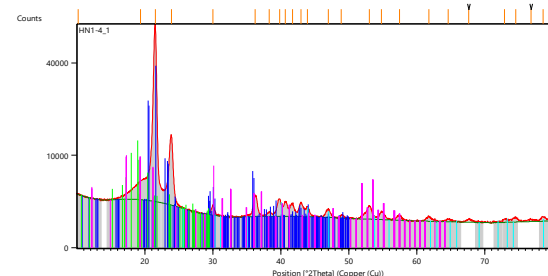


Figure 4. XRD Diffractograms Intensity of $^{\circ}2\theta$ for 94 % HDPE/ 1 % MC-NC/Org-MMT and HDPE-grafted maleic anhydride Bio-Composites.

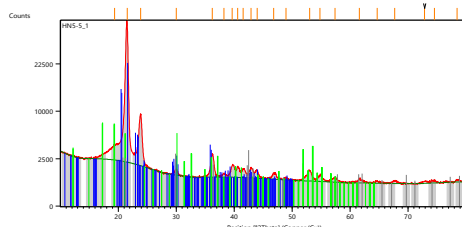


Figure 5. XRD Diffractograms Intensity of $^{\circ}2\theta$ for 90 % HDPE/5 % MC-NC/ Org-MMT and HDPE-grafted maleic anhydride Bio-Composites.

HDPE-g-MA enhances interfacial interactions, allowing better filler integration, yet may not fully compensate for the steric and chemical effects of excessive filler loading Walid et al (2022).

These findings are in line and consistent with earlier studies by Arian et al (2020) are indicating that nanoclay and cellulose fillers can reduce crystallinity and modify polymer lattice organization, depending on loading level and compatibilizer efficiency.

3.2 X-Ray Diffraction (XRD) Analysis of LDPE-Based Bio-Composites

X-ray diffraction (XRD) analysis was conducted to examine the crystalline behavior and structural modifications in low-density polyethylene (LDPE) based bio-composites reinforced with maize cob nanocellulose (MC-NC), organo-modified montmorillonite nanoclay (Org-MMT), and compatibilized using LDPE-grafted maleic anhydride (LDPE-g-MA). Figures 6 to 11 illustrate the XRD patterns of various LDPE composite formulations, highlighting the changes in crystallinity and possible intercalation effects caused by filler incorporation.

Figure 6. Depicted the XRD pattern of neat LDPE (100 %), characterized by broad diffraction peaks at approximately 21.1° and $23.5^\circ 2\theta$, which correspond to the (110) and (200) planes of LDPE's orthorhombic crystal lattice. Compared to HDPE, LDPE exhibits broader and less intense peaks due to its higher degree of branching and lower crystallinity. This baseline spectrum is used to assess the structural influence of nanofillers and compatibilizer in the subsequent/ reinforced composites Walid et al (2022); Depan et al (2021)

Figure 7 represents the XRD pattern of a binary composite containing 2 % MC-NC and 98 % LDPE. The crystalline peaks of LDPE remain largely unchanged, but a slight increase in peak broadening is observed, indicating a minor disruption in crystallite formation due to the incorporation of MC-NC Zikri et al (2018). As MC-NC is predominantly amorphous, its incorporation tends to decrease the degree of crystallinity by introducing non-crystalline domains within the polymer matrix. However, in the absence of a compatibilizer or nanoclay, the extent of disruption remains limited, and no significant shift in peak position is observed.

Figure 8 shows the composites of 2 % MC-NC 1 % organo-modified Montmorillonite nanoclay (Org-MMT), 3 % LDPE-grafted maleic anhydride and 94 % of Low-density polyethylene (LDPE).

In this sample, the crystalline peaks of LDPE persist but with slightly reduced intensity, suggesting that the fillers interfere with regular lamellar packing. A minor reflection at low angles ($2\theta \approx 2-6^\circ$) may appear due to the basal spacing of Org-MMT, suggesting partial intercalation of polymer chains into the clay galleries. The presence of LDPE-g-MA supports improved dispersion of hydrophilic fillers (MC-NC) within the non-polar matrix which may have promote the exfoliation or intercalation of clay, though the persistence of MMT peaks suggests incomplete delamination Mohamed et al (2020)

Figure 9 corresponds to a composite composed of 2 % maize cob nanocellulose (MC-NC), 5 % organo-modified Montmorillonite nanoclay (Org-MMT), 3 % LDPE-grafted maleic anhydride and 90 % of Low-density polyethylene (LDPE)

The LDPE crystalline peaks exhibit noticeable broadening and attenuation, indicating a substantial reduction in crystalline domain size or perfection. The higher nanoclay content increases the likelihood of filler agglomeration Mohamed et al (2020); Ray & Banerjee (2025). But also introduces greater structural interference within the polymer matrix. A more distinct low-angle reflection may be present due to increased intercalated clay content, although complete exfoliation remains unlikely. This composite reflects a typical trade-off where increased filler loading disrupts crystallinity while enhancing interfacial reinforcement.

Figure 10 presents the XRD pattern for a composite containing 1 % maize cob nanocellulose (MC-NC), 2 % organo-modified Montmorillonite nanoclay (Org-MMT), 3 % LDPE-grafted maleic anhydride and 94 % of Low-density polyethylene (LDPE). The pattern resembles that of Figure 8, with slightly sharper LDPE peaks and reduced background noise. This suggests better retention of crystallinity at lower filler loading; it may be attributed to more effective dispersion and minimized steric hindrance. The low-angle Org-MMT peak, if present, remains weak, indicating limited intercalation or partial exfoliation at lower nanoclay loading of 2 % Ray & Banerjee (2025).

Figure 11 displays the XRD pattern for a composite of 5 % maize cob nanocellulose (MC-NC), 2 % organo-modified Montmorillonite nanoclay (Org-MMT), 3 % LDPE-grafted maleic anhydride and 90 % of Low-density polyethylene (LDPE).

In this system, the LDPE crystalline peaks are significantly weakened and broadened, indicating substantial crystallinity reduction.

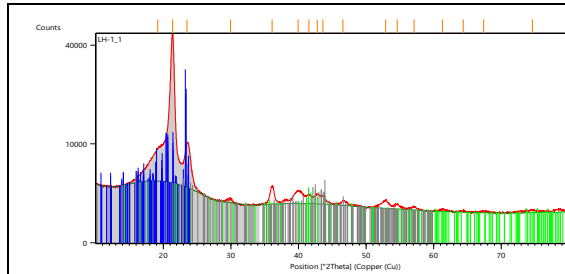


Figure 6. XRD Diffractograms Intensity of 2θ for composites of 100 % Low-density polyethylene (LDPE)

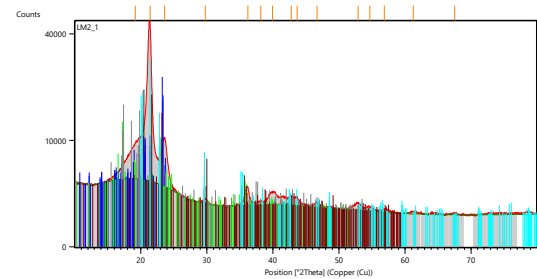


Figure 7. XRD Diffractograms intensity of 2θ for composites of 2 % maize cob nanocellulose (MC-NC) and 98 % of Low-density polyethylene (LDPE)

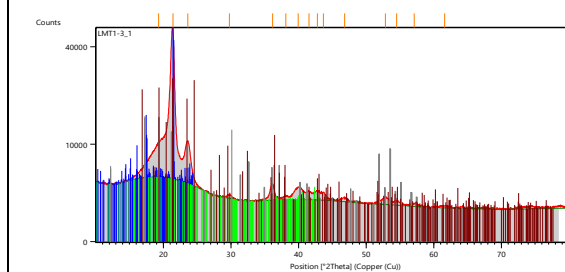


Figure 8. XRD Diffractograms Intensity of 2θ for 94 % LDPE/ MC-NC/ 1 % Org-MMT and LDPE-grafted maleic anhydride Bio-Composites

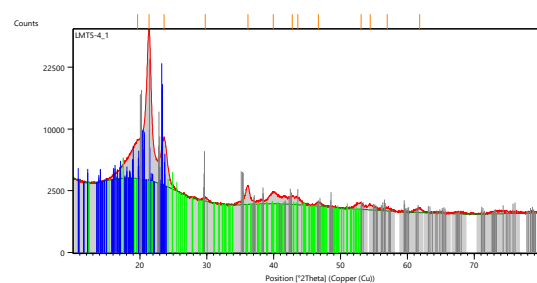


Figure 9. XRD Diffractograms Intensity of 2θ for 90 % LDPE/ MC-NC/ 5 % Org-MMT and LDPE-grafted maleic anhydride Bio-Composites

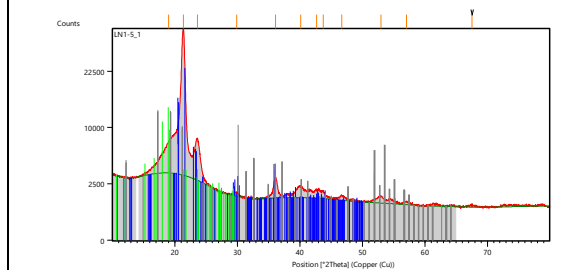


Figure 10. XRD Diffractograms Intensity of 2θ for 94 % LDPE/ 1 % MC-NC/Org-MMT and LDPE-grafted maleic anhydride Bio-Composites

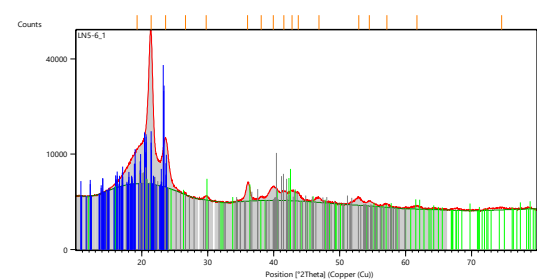


Figure 11. XRD Diffractograms Intensity of 2θ for 90 % LDPE/ 5 % MC-NC/Org-MMT and LDPE-grafted maleic anhydride Bio-Composites.

The high MC-NC content introduces amorphous content that competes with crystalline LDPE domain formation, while the Org-MMT may hinder long-range molecular ordering. The appearance of broad amorphous halos and subdued crystalline peaks reflects increased heterogeneity and reduced lattice regularity, likely a consequence of excessive filler content and hydrogen-bonding interactions between MC-NC particles Hao et al (2024).

From Figures 6 to 11, the degree of crystallinity in LDPE-based composites progressively declines with increased filler content, especially at high loadings of MC-NC and Org-MMT. Peak broadening, intensity loss, and the appearance of low-angle reflections signal the partial intercalation of polymer chains into nanoclay layers Rajapakse et al (2026), while LDPE-g-MA facilitates dispersion but does not fully preserve crystalline order. These XRD features demonstrate how the combined influence of nanofillers and compatibilizer alters LDPE's crystalline structure, echoing findings from related polymer-clay systems Liu et al (2004); Ray & Okamoto (2003).

3.3 Comparative Discussion for HDPE and LDPE structural findings XRD

The comparative XRD analysis of HDPE- and LDPE-based bio-composites reveals distinct differences in crystalline morphology, filler integration, and structural response to MC-NC and Org-MMT incorporation. Neat HDPE (Figure 1) exhibits sharper, well-defined diffraction peaks at approximately 21.6° and 23.9° 2θ , corresponding to the (110) and (200) planes of its orthorhombic unit cell, confirming its higher intrinsic crystallinity and lamellar regularity. In contrast, neat LDPE (Figure 6) presents broader and less intense peaks near 21.1° and 23.5° 2θ , reflecting the polymer's branched architecture and lower degree of crystallinity Rajapakse et al (2026) Upon addition of MC-NC and Org-MMT, both systems display peak broadening and reduced intensity, indicating a general trend of crystalline disruption.

However, HDPE composites retain sharper diffraction features relative to their LDPE counterparts at equivalent filler loadings, suggesting that HDPE's linear chain structure better tolerates the spatial hindrance imposed by fillers. In HDPE composites with higher Org-MMT content (e.g., Figure 3), a more pronounced reduction in crystallinity is evident, although the persistence of key diffraction peaks implies partial intercalation without complete exfoliation. LDPE composites (e.g., Figures 9 and 11), especially those with elevated MC-NC and Org-MMT (5 %) loading, show greater peak attenuation and amorphous halo formation, signifying more severe lattice disorder. This contrast arises from LDPE's lower baseline crystallinity and its greater susceptibility to steric and hydrogen-bonding interference.

The appearance of low-angle reflections in both systems—attributed to the basal spacing of Org-MMT María et al (2025); Shaw (2020) suggests partial intercalation in both matrices, yet the extent of polymer–clay interaction appears more coherent in HDPE, possibly due to better filler dispersion assisted by HDPE-g-MA. While LDPE-g-MA similarly enhances interaction in LDPE composites, the structural irregularity of the matrix limits the extent of ordered filler alignment and crystalline preservation.

Overall, HDPE composites demonstrate more resilient crystallinity and clearer diffraction features under nanofiller inclusion, whereas LDPE composites experience more pronounced crystalline disruption, emphasizing the influence of matrix architecture on structural integrity and filler compatibility in nanocomposite systems.

I.CONCLUSION

This research investigates the technical applicability of utilizing Sammaz-14 maize cob nanocellulose as a sustainable reinforcement for polyethylene-based nanocomposites. The study confirms that matrix structure plays a critical role in governing crystallinity retention, filler dispersion, and structural response upon nanofiller incorporation/loading.

XRD analysis revealed that neat HDPE exhibits higher intrinsic crystallinity characterized by sharp and well-defined diffraction peaks corresponding to its orthorhombic structure, whereas LDPE presents broader peaks due to its branched molecular configuration and lower crystallinity. Upon incorporation of MC-NC and Org-MMT, both polymer systems experienced peak broadening and intensity reduction, indicating partial crystalline disruption. However, HDPE composites retained comparatively sharper diffraction features, demonstrating greater structural resilience against filler-induced steric interference.

The addition of 2 wt% Org-MMT was sufficient to promote improved intercalation and dispersion within both matrices (HDPE & LDPE), while 3 wt% maleic anhydride grafted polyethylene significantly enhanced interfacial adhesion by reducing surface tension and facilitating chemical interactions between the polymer matrix and hydrophilic nanocellulose. Alkaline treatment further improved fiber–matrix compatibility by modifying surface chemistry and enhancing bonding efficiency.

LDPE-based composites exhibited greater crystalline reduction and amorphous halo formation at higher filler loadings (5 %), indicating increased lattice disorder due to its inherently lower baseline crystallinity and greater susceptibility to steric and hydrogen-bonding interactions. Although LDPE-g-MA improved compatibility, the branched structure of LDPE limited ordered filler alignment compared to HDPE systems.

Overall, HDPE-based nanocomposites demonstrated superior structural integrity, clearer diffraction characteristics, and better preservation of crystallinity under nanofiller inclusion compared to LDPE composites. The findings emphasize that effective compatibilization (3% MAH grafting) combined with optimized nanoclay loading (2% Org-MMT) is critical for achieving enhanced dispersion and interfacial bonding in bio-based polyethylene nanocomposites.

This study provides a sustainable materials engineering framework for converting agro-residues into high-performance nanocomposite reinforcements for construction and automotive interior.

AUTHOR(S) CONTRIBUTIONS

Jamila Baba Ali & Abdullahi Danladi, conceptualized the study, designed the methodology, conducted the analysis and wrote the first draft also reviewed/edited the final manuscript. Suleiman Abdulrahman & Abubakar Musa Bunza provided funding and partook in reviewing of the manuscript. A. S. Abdul Rasheed & Aliyu Ishaq supervised the experimental process and partook in the review and editing of the final manuscript.

AI DISCLOSURE

Some part of the discussion of results in this research was paraphrased using ChatGPT with proper citation.

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DECLARATION OF CONFLICT OF INTEREST

All authors declared no conflict of interest and approve this submission.

REFERENCES

- Ali, J. B., Danladi, A., Bukhari, M. M., Nyakuma, B. B., Mamza, P., Mohamad, Z. B., ... Inuwa, I. M. (2022). Extraction and Characterization of Cellulose Nanofibres and Cellulose Nanocrystals from Sammaz-14 Maize Cobs. *Journal of Natural Fibers*, 19(7), 2756–2771. <https://doi.org/10.1080/15440478.2020.1856279>
- Amirkiai, A., Sarmad, M. P., Sadeghi, G. M., Arjmand, M., Abrisham, M., Dehghan, P., & Nazockdast, H. (2020). Microstructural design for enhanced mechanical and shape memory performance of polyurethane nanocomposites: Role of hybrid nanofillers of montmorillonite and halloysite nanotube. *Applied Clay Science*, 198, 105816. <https://doi.org/10.1016/j.clay.2020.105816>
- Baba Ali, J., Danladi, A., Bukhari, M. M., Mamza, P., & Zurina, M. (2020). Effect of Org-MMT on the mechanical and morphological properties of HDPE/HD-g-MAH/maize cobs nanocomposite. *SLU Journal of Science and Technology*, 1(1), 24–32.
- Depan, D., Chirdon, W., & Khattab, A. (2021). Morphological and chemical analysis of low-density polyethylene crystallized on carbon and clay nanofillers. *Polymers*, 13(10), 1558. <https://doi.org/10.3390/polym13101558>
- Gupta, N., Mahur, B. K., Izrayeel, A. M. D., et al. (2022). Biomass conversion of agricultural waste residues for different applications: A comprehensive review. *Environmental Science and Pollution Research*, 29, 73622–73647. <https://doi.org/10.1007/s11356-022-22802-6>

- Lan, H., Wang, J., Cheng, L., Yu, D., Wang, H., & Guo, L. (2024). The synthesis and application of crystalline–amorphous hybrid materials. *Chemical Society Reviews*, 53, 684–713. <https://doi.org/10.1039/D3CS00860F>
- Lorenzo, M. C., Reale, P. D., Colonna, J. I., Larralde, A. L., Lamas, D. G., & Eisenberg, P. (2025). Influence of clay chemistry on the microstructure, water sensitivity, and home-composting disintegration of protein-rich wheat gluten–PBAT blends. *ACS Applied Polymer Materials*, 8(1), 394–404. <https://doi.org/10.1021/acsapm.5c03815>
- Mishra, B., Varshney, S., & Gupta, M. K. (2025). Effect of fillers on the performance of fibre reinforced polymer hybrid composites: A comprehensive review. *Polymer-Plastics Technology and Materials*, 64(14), 2179–2213. <https://doi.org/10.1080/25740881.2024.2415678>
- Mohamed, A., Codou, A., Anstey, A., Mohanty, A. K., & Misra, M. (2020). Studies on the dimensional stability and mechanical properties of nanobiocomposites from polyamide 6-filled with biocarbon and nanoclay hybrid systems. *Composites Part A: Applied Science and Manufacturing*, 129, 105695. <https://doi.org/10.1016/j.compositesa.2019.105695>
- Mohammadi, M., Ishak, M. R., & Sultan, M. T. H. (2024). Exploring chemical and physical advancements in surface modification techniques of natural fiber reinforced composite: A comprehensive review. *Journal of Natural Fibers*, 21(1). <https://doi.org/10.1080/15440478.2024.2408633>
- Noer, Z., Nasruddin, M. N., Bukit, N., Juwairiah, Susilawati, & Ikhwanuddin. (2018). Characterization of low-density polyethylene (LDPE)/carbon black (CB) nanocomposite-based packaging material. *Journal of Physics: Conference Series*, 1120(1), 012066. <https://doi.org/10.1088/1742-6596/1120/1/012066>
- Oueslati, W., Mejri, C., & Amara, A. B. H. (2022). X-ray diffraction profiles modeling method for layered structures reconstruction: Nanoclay structural verification. *IntechOpen*. <https://doi.org/10.5772/intechopen.107017>
- Rajapakse, R. M. G., Malalagama, T., Bandara, V. M. Y. S. U., & Soonmin, H. (2026). Electroactive clay polymer nanocomposites for sustainable device applications. *Journal of Chemical Reviews*, 8(2), 214–240. <https://doi.org/10.48309/JCR.2026.537691.1488>
- Ray, S. S., & Banerjee, R. (2025). Techniques for structural and morphological characterisation of polymer blend nanocomposites. In *Polymer Blend Nanocomposites* (Springer Series in Materials Science, Vol. 354). Springer, Cham. https://doi.org/10.1007/978-3-032-00309-6_4
- Ruud, C. O., & Russell, P. A. (2018). *X-ray diffraction: Analysis of airborne particles by physical methods* (1st ed.). Taylor & Francis.
- Saha, M., Ray, R., Choudhury, A. R., et al. (2021). Impact of exfoliation/intercalation of nanoclay on structure, morphology and electrical properties of poly (ethylene oxide) based solid nanocomposite electrolytes. *Journal of Polymer Research*, 28, 299. <https://doi.org/10.1007/s10965-021-02622-x>
- Sambyal, P., Najmi, P., Sharma, D., Khoshbakhti, E., Hosseini, H., Milani, A. S., & Arjmand, M. (2024). Plastic recycling: Challenges and opportunities. *The Canadian Journal of Chemical Engineering*. <https://doi.org/10.1002/cjce.25531>
- Sharma, M., Chaubey, K. K., Pal, A. K., Thapliyal, S., Kumar, A., Sharma, S., Sanghi, A., Kumar, V., Tripathi, V. L., & Tiwari, S. (2026). *Utilization of maize waste for various applications: Sustainable solutions for agricultural waste* (1st ed.). Taylor & Francis Group.

- Shaw, A. V. (2020). Linking the thermal, morphological and electrical properties of thermoplastic polymer composites containing an organoclay (Doctoral thesis). University of Southampton.
- Shelly, D., Singhal, V., Singh, S., Nanda, T., Mehta, R., Lee, S.-Y., & Park, S.-J. (2024). Exploring the impact of nanoclay on epoxy nanocomposites: A comprehensive review. *Journal of Composites Science*, 8(12), 506. <https://doi.org/10.3390/jcs8120506>
- Shishir, K. H., Islam, M. M., Islam, M. T., Rahaman, M. A., Mustak, M. H., Mostafa, M. G., & Alam, M. A. (2025). X-ray line diffraction pattern profiling of monoclinic sucrose nanocrystal. *Results in Surfaces and Interfaces*, 20, 100595. <https://doi.org/10.1016/j.rsurfi.2025.100595>
- Silva, R., Graf, K., & Ribeiro Okimoto, M. L. (2025). Plastic waste recycling: An overview of the mechanical, chemical, and thermal technologies. *Journal of Engineering and Applied Science*, 72, 251. <https://doi.org/10.1186/s44147-025-00799-2>