Nanocellulose from Goosegrass (*Eleusine indica*) as a Blending Polymer for Pebax Membranes

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**Abstract.** Nanocellulose has gained considerable attention as an environmentally friendly and versatile biomaterial. This study investigated the extraction of nanocellulose from goosegrass (*Eleusine indica*), an abundant and renewable resource, to enhance the mechanical properties of Pebax membranes. Nanocellulose was obtained through sulfuric acid hydrolysis of goosegrass microcellulose and was subsequently incorporated into Pebax to create membrane films. The extracted nanocellulose was characterized using particle size analysis (PSA), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and thermogravimetric analysis (TGA). The mechanical properties of the resulting Pebax-nanocellulose membranes were evaluated using a universal testing machine (UTM). PSA revealed successful nanocellulose isolation with an average particle size of 80.2 nm. FTIR analysis confirmed the removal of non-cellulosic components and the introduction of sulfonate groups onto the nanocellulose surface. XRD analysis demonstrated an increase in crystallinity from 51.87% to 78.12%. The TGA results indicated that the nanocellulose exhibited thermal stability up to 383.19°C. The incorporation of nanocellulose into the Pebax matrix led to improved mechanical properties. This study highlights the potential of goosegrass as a sustainable source of high-quality nanocellulose with promise as a reinforcing agent in membrane applications.

# INTRODUCTION

The growing demand for sustainable and eco-friendly materials has spurred extensive research on renewable resources for various applications. Nanocellulose, a nanostructured form of cellulose derived from plant fibers, has emerged as a promising biomaterial owing to its exceptional properties, including high strength, lightweight nature, biodegradability, and renewability.1–4 Its diverse applications range from reinforcing agents in composites to biomedical materials5 and membrane technology.6–8

Traditionally, nanocellulose production relied on wood pulp or agricultural residue. However, the utilization of underutilized plant resources offers a sustainable and cost-effective alternative. Goosegrass (*Eleusine indica*), a common weed found in tropical and subtropical regions, is a potential feedstock for nanocellulose extraction. This grass is characterized by high cellulose content and rapid growth rate, making it an attractive candidate for valorization.9

Isolating cellulose from goosegrass involves removing non-cellulosic components and then extracting the nanocellulose. Various methods, including chemical,10 mechanical,11 and enzymatic treatments,12 have been employed for nanocellulose preparation. Acid hydrolysis, a widely used chemical method, involves cleaving cellulose chains using strong acids, resulting in nanocellulose formation.1,13–16

This study aimed to investigate the potential of goosegrass as a source of nanocellulose. The extraction process involves acid hydrolysis using sulfuric acid, followed by the characterization of the obtained nanocellulose using various techniques, including PSA, FTIR, XRD, and TGA. Furthermore, the potential applications of goosegrass nanocellulose in membrane technology have been explored by preparing and characterizing Pebax-nanocellulose composite membranes. The mechanical properties of these membranes have been evaluated to assess the reinforcing effect of nanocellulose.

The findings of this research will contribute to sustainable and eco-friendly materials by using abundant and underutilized plant resources. Successful extraction and characterization of nanocellulose from goosegrass will pave the way for its potential applications in membrane development and other advanced materials.

# MATERIALS AND METHODS

## Materials

Goosegrass (*Eleusine indica*) was obtained from local agricultural waste in Sumenep, Indonesia. Sodium hydroxide (NaOH), hydrogen peroxide (H2O2), and hydrochloric acid (HCl, 37%) were obtained from Sigma-Aldrich, USA.

## Microcellulose Isolation

Microcellulose isolation was carried out in several stages, including alkalization, bleaching, and hydrolysis, as described previously.9 Goosegrass powder was subjected to alkali treatment with 3% (w/w) NaOH solution at 100°C for 3 h, followed by washing with distilled water until neutral. The remaining insoluble residue was bleached with H2O2 at 80°C for 2 h and washed again. The residue was then treated with 2 N HCl for 1 h and washed. The resulting residue was oven-dried and stored.

## Nanocellulose Extraction

Nanocellulose was extracted via acid hydrolysis using a 40% (w/v) sulfuric acid solution (fiber:acid ratio 1:50) at 45°C for 45 min with continuous stirring. The reaction was terminated by dilution with water, followed by repeated washing and centrifugation at 11000 rpm for 30 min. A final sonication step (40 kHz, 45 min) was performed to disperse the nanocellulose. The resulting nanocellulose suspension was stored for further characterization and use in membrane fabrication.

## Nanocellulose Characterization

Nanocellulose characterization involves several techniques to assess its properties. Fourier transform infrared spectroscopy (FTIR) was used to identify the functional groups in the raw fibers, microcellulose, and nanocellulose samples. The particle size distribution of the nanocellulose products was determined using a particle size analyzer. The crystallinity index (CI) was calculated using the Segal equation from the X-ray diffraction (XRD) patterns obtained using a diffractometer. Thermal stability was assessed using thermogravimetric analysis (TGA).

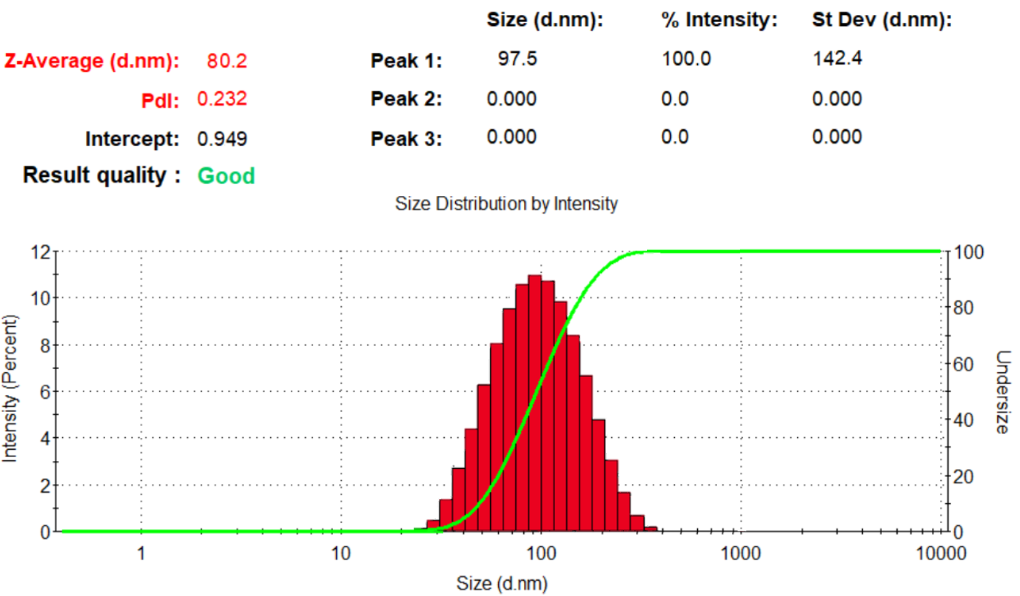
## Fabrication of Pebax–Nanocellulose Membranes and Mechanical Testing

Pebax–nanocellulose composite membranes were fabricated by solvent casting. A 4-wt% Pebax solution was prepared in a 70-wt% ethanol/water co-solvent. Nanocellulose (0.4% and 0.6%) was added, and the resulting mixture was cast onto a glass substrate and dried. The mechanical properties of the membranes were evaluated using a universal testing machine.

# Results and discussion

## Particle size analysis

Particle size analysis (PSA) of the nanocellulose samples revealed a monopodal distribution with a Z-average size of 80.2 nm, as shown in Figure 1. This indicates a relatively homogeneous sample, with most particles falling within a narrow range around the average. The polydispersity index (PdI) of 0.232 further supports this, suggesting a moderate width of size distribution.17 The absence of significant peaks at other particle sizes confirms the absence of larger or smaller particle populations. The cumulative undersize curve shows a steep rise, indicating that the majority of particles are smaller than 100 nm. Overall, the PSA results suggest that the nanocellulose sample is well-dispersed and consists primarily of particles approximately 80 nm in size.



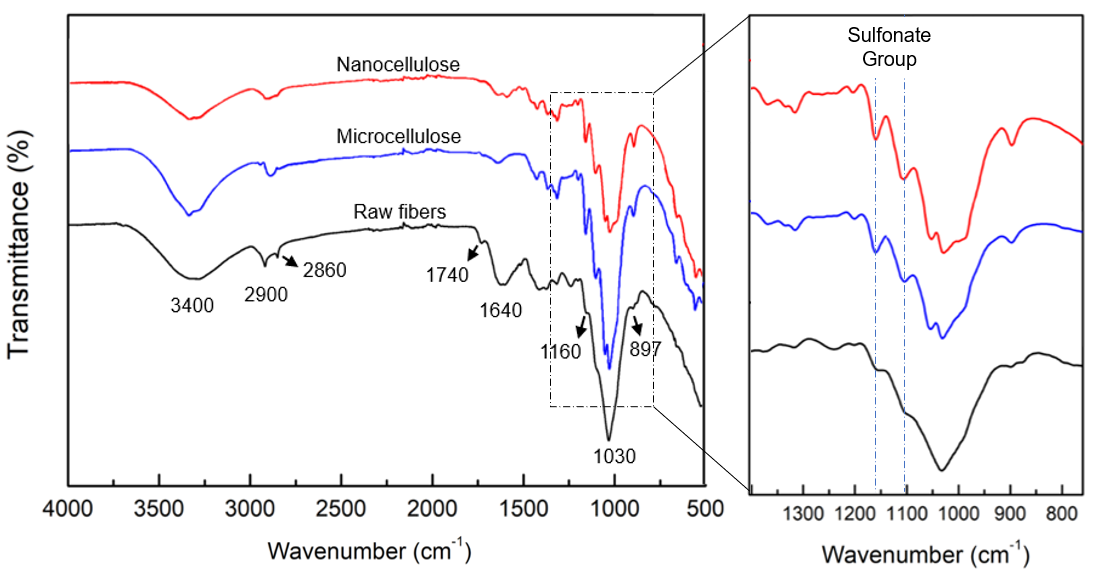
**Figure 1.** Size distribution of nanocellulose determined by PSA measurement

## Functional Group Analysis

The FTIR spectra of raw fibers, microcellulose, and nanocellulose revealed distinct patterns due to processing (Figure 2). Raw fibers exhibited a broad peak around 3400 cm-1 characteristic of -OH stretching vibrations, primarily from cellulose and hemicellulose.9 Peaks at 2860-2900 cm-1 were attributed to C-H stretching vibrations from aliphatic groups within the cellulose structure.18–20 A peak at 1740 cm-1 suggested the presence of carbonyl (C=O) groups, likely from hemicellulose or lignin.

Microcellulose retained similar peaks as raw fibers, but with a noticeable decrease in intensity at 1740 cm-1, indicating a reduction in hemicellulose content. Nanocellulose shared the broad -OH peak, C-H peaks, and the peak at 1030 cm-1 with microcellulose, confirming the preservation of the fundamental cellulose structure. Sharper peaks, particularly around 1030 cm-1 (C-O stretching) and 897 cm-1 (β-glycosidic linkages), suggested an increase in crystallinity compared with microcellulose. The complete disappearance of the 1740 cm-1 peak indicated the removal of almost all hemicellulose during nanocellulose production. Meanwhile, the increase of peaks between 1100-120 cm-1 corresponds to S=O on sulfonate groups due to acid hydrolysis as previously reported.21

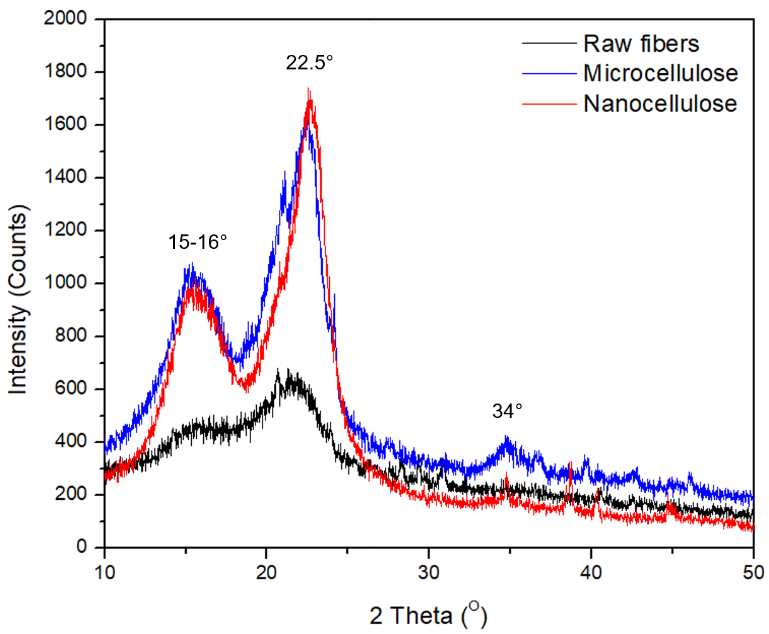
Overall, FTIR analysis revealed the successful isolation of cellulose from raw fibers and a possible increase in nanocellulose crystallinity, enhancing its mechanical and thermal properties.



**Figure 2.** FTIR spectra of raw fibers, microcellulose, and nanocellulose

## Crystallinity Analysis

The XRD analysis of raw fiber, microcellulose, and nanocellulose provided insights into their crystallinity (Figure 3). The XRD pattern of raw fibers showed a broad peak around 15-16°, characteristic of amorphous regions within cellulose, indicating a lower degree of crystallinity (51.87%). The microcellulose XRD pattern exhibited a sharper peak at 22.5°, indicating a higher degree of crystallinity (75.76%) due to the removal of amorphous components.9,22 The nanocellulose XRD pattern was similar to that of microcellulose but with a slightly more intense peak, suggesting a further increase in crystallinity (78.12%). Overall, the XRD analysis demonstrated the effectiveness of the extraction and processing methods in increasing the cellulose crystallinity from raw fibers to nanocellulose.



**Figure 3.** XRD diffraction patterns of raw fibers, microcellulose, and nanocellulose

## Thermal Analysis

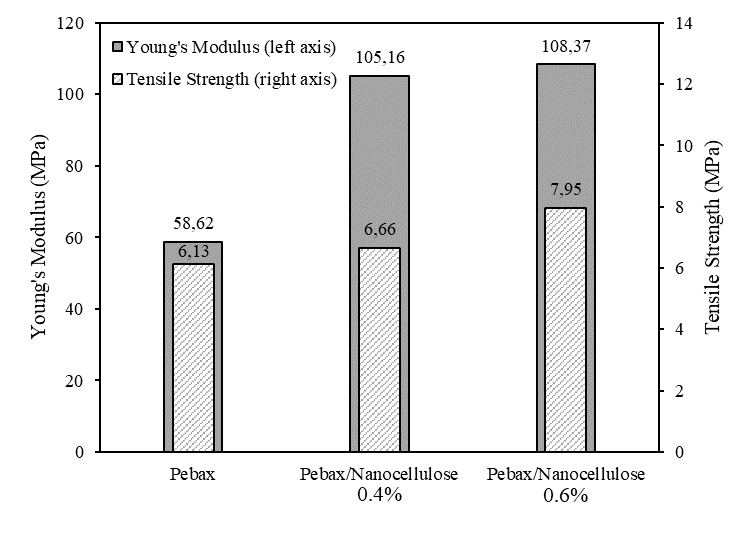
The TGA and DTG curves provided insights into the thermal degradation behavior of the samples (Figure 4). All samples exhibited similar initial weight losses below 100°C due to moisture evaporation.23,24 The major weight loss occurred in the range of 200-400°C, corresponding to cellulose decomposition. Nanocellulose exhibited the highest thermal stability, with the most significant weight loss occurring at higher temperatures compared to microcellulose and raw fibers. The final degradation temperature of nanocellulose was 383.19°C, followed by microcellulose at 360.67°C and raw fibers at 314.15°C. The increased thermal stability of nanocellulose can be attributed to its higher crystallinity, reduced amorphous content, and increased surface area.9

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| (a) | (b) |

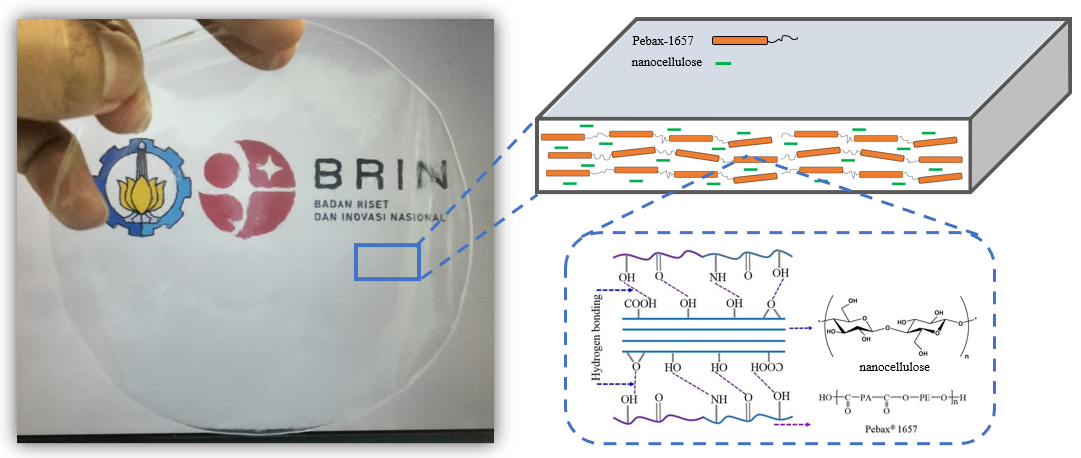
**Figure 4.** TGA (a) and DTG (b) curves of raw fibers, microcellulose, and nanocellulose.

## Mechanical Properties of Pebax/Nanocellulose Membranes

The incorporation of nanocellulose into Pebax membranes significantly affected their mechanical properties. The tensile strength and Young modulus increased with increasing nanocellulose content (Figure 5). This enhancement can be attributed to the strong interfacial interactions between nanocellulose and the Pebax matrix, as well as the reinforcing effect of the rigid nanocellulose fibers.25 Hydrogen bonding between the hydroxyl groups on the nanocellulose surface and the amide groups in Pebax as shown in Figure 6 likely contributed to the improved mechanical properties.7,26 In addition to hydrogen bonding, weak van der Waals forces also contribute to the interaction between the nanocellulose and Pebax 1657 molecules. These forces arise from temporary fluctuations in electron distribution and further strengthen the composite structure.



**Figure 5.** Tensile strength and elongation at break of the pebax/nanocellulose membranes



**Figure 6.** Schematic representation of hydrogen bonding interactions between nanocellulose and Pebax 1657

Overall, the mechanical properties of the Pebax/nanocellulose membranes indicate a good balance between strength and flexibility. The addition of nanocellulose improved the tensile strength and stiffness of the membranes while maintaining adequate elongation at break for most applications. The strong interfacial interactions between nanocellulose and Pebax, facilitated by hydrogen bonding, make their composites promising candidates for gas separation membrane applications. The dense and interconnected network formed by these materials can significantly reduce the permeability of gases, enhancing the selectivity and efficiency of gas separation processes. Additionally, the improved mechanical properties and thermal stability of these composites enable their use in demanding environments, making them suitable for various industrial applications.

# CONCLUSION

This study successfully demonstrated the extraction of high-quality nanocellulose from goosegrass, which is an abundant and underutilized plant resource. The extracted nanocellulose was characterized by its small particle size (80.2 nm), high crystallinity (78.12%), and excellent thermal stability (up to 383.19°C). The incorporation of nanocellulose into Pebax membranes significantly improved their mechanical properties, including their tensile strength and Young modulus. This study highlights the potential of goosegrass as a sustainable source of nanocellulose for developing high-performance, eco-friendly membrane materials. Further research is warranted to investigate the complete characteristics and performances of these membranes for gas separation.

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# References

1. M. Asrofi, H. Abral, A. Kasim, A. Pratoto, M. Mahardika, J.W. Park, and H.J. Kim, “Isolation of Nanocellulose from Water Hyacinth Fiber (WHF) Produced via Digester-Sonication and Its Characterization,” Fibers Polym. **19**(8), 1618–1625 (2018).

2. M. Mahardika, H. Abral, A. Kasim, S. Arief, and M. Asrofi, “Production of Nanocellulose from Pineapple Leaf Fibers via High-Shear Homogenization and Ultrasonication,” Fibers **6**(2), 28 (2018).

3. D. Rahmadiawan, H. Abral, W.H. Yesa, D. Handayani, N. Sandrawati, E. Sugiarti, A.N. Muslimin, S.M. Sapuan, and R.A. Ilyas, “White Ginger Nanocellulose as Effective Reinforcement and Antimicrobial Polyvinyl Alcohol/ZnO Hybrid Biocomposite Films Additive for Food Packaging Applications,” J. Compos. Sci. 2022, Vol. 6, Page 316 **6**(10), 316 (2022).

4. A.S. Norfarhana, R.A. Ilyas, and N. Ngadi, “A review of nanocellulose adsorptive membrane as multifunctional wastewater treatment,” Carbohydr. Polym. **291**, 119563 (2022).

5. S. Jiji, K. Maharajan, and K. Kadirvelu, “Recent developments of bacterial nanocellulose porous scaffolds in biomedical applications,” Nanocellulose Mater., (2022).

6. J. Torstensen, R.M.L. Helberg, L. Deng, Ø.W. Gregersen, and K. Syverud, “PVA/nanocellulose nanocomposite membranes for CO2 separation from flue gas,” Int. J. Greenh. Gas Control **81**, (2019).

7. Z. Dai, J. Deng, Y. Ma, H. Guo, J. Wei, B. Wang, X. Jiang, and L. Deng, “Nanocellulose Crystal-Enhanced Hybrid Membrane for CO2Capture,” Ind. Eng. Chem. Res. **61**(25), 9067–9076 (2022).

8. Z. Jahan, M.B.K. Niazi, M.B. Hägg, and Ø.W. Gregersen, “Cellulose nanocrystal/PVA nanocomposite membranes for CO2/CH4 separation at high pressure,” J. Memb. Sci. **554**, 275–281 (2018).

9. A.W. Pratama, M. Mahardika, N. Widiastuti, B. Piluharto, R.A. Ilyas, S.M. Sapuan, D. Amelia, and A. Firmanda, “Isolation and characterization of highly thermal stable microcrystalline cellulose derived from belulang grass (Eleusine indica),” Case Stud. Chem. Environ. Eng. **9**, 100743 (2024).

10. A.W. Pratama, H.S. Addy, N. Widiastuti, A.R. Widyanto, A. Ratnasari, D. Indarti, and B. Piluharto, “Cellulose Nanofibrils from Corncobs and Their Nanocomposite with Alginate : Study of Swelling Behavior,” Biointerface Res. Appl. Chem. **14**(1), 4–21 (2024).

11. O. Dovjuu, S. Kim, A. Lee, S. Baek, J. Kim, J. Noh, S. Huh, B. Choi, Y. Sung, and H. Jeong, “Structural characterization of the crystalline nanocellulose and nanocellulose-reinforced carbon buckypaper,” Diam. Relat. Mater. **106**, 107821 (2020).

12. D. Zielí Nska, K. Szentner, A. Wa´skiewiczwa´skiewicz, S. Borysiak, Z. Nska, D.; Szentner, K.; Wa´skiewiczwa´skiewicz, and A.; Borysiak, “Production of Nanocellulose by Enzymatic Treatment for Application in Polymer Composites,” Materials (Basel). **14**(9), 2124 (2021).

13. R. Ratna, N. Arahman, A.A. Munawar, and S. Aprilia, “Extraction, Isolation, and Characterization of Nanocrystalline Cellulose from Barangan Banana (Musa acuminata L.) Peduncles Waste,” Indones. J. Chem. **23**(1), 73–89 (2023).

14. A.A. Oun, and J.W. Rhim, “Isolation of cellulose nanocrystals from grain straws and their use for the preparation of carboxymethyl cellulose-based nanocomposite films,” Carbohydr. Polym. **150**, 187–200 (2016).

15. S. Soleimani, A. Heydari, and M. Fattahi, “Isolation and Characterization of Cellulose Nanocrystals from Waste Cotton Fibers Using Sulfuric Acid Hydrolysis,” Starch **74**(11–12), 2200159 (2022).

16. Y. Wu, C. Luo, T. Wang, Y. Yang, Y. Sun, Y. Zhang, L. Cui, Z. Song, X. Chen, X. Cao, S. Li, and G. Cai, “Extraction and characterization of nanocellulose from cattail leaves: Morphological, microstructural and thermal properties,” Int. J. Biol. Macromol. **255**, 128123 (2024).

17. S. Rao, Y. Song, F. Peddie, and A.M. Evans, “Particle size reduction to the nanometer range: a promising approach to improve buccal absorption of poorly water-soluble drugs,” Int. J. Nanomedicine **6**, 1245–1251 (2011).

18. E. Syafri, J. Jamaluddin, N.H. Sari, M. Mahardika, L. Suryanegara, R. Sinaga, F. Yudhanto, R. Zainul, A. Nugroho, and A. Khan, “Effect of ultrafine grinding and ultrasonication duration on the performance of polyvinyl alcohol (PVA) agave gigantea cellulose micro fiber (CMF) bio-composite film,” J. Nat. Fibers **20**(1), 2192545 (2023).

19. O.O. Sadare, K.O. Yoro, K. Moothi, and M.O. Daramola, “Lignocellulosic Biomass-Derived Nanocellulose Crystals as Fillers in Membranes for Water and Wastewater Treatment: A Review,” Membranes (Basel). **12**(3), (2022).

20. R. Vijay, D. Lenin Singaravelu, A. Vinod, M.R. Sanjay, S. Siengchin, M. Jawaid, A. Khan, and J. Parameswaranpillai, “Characterization of raw and alkali treated new natural cellulosic fibers from Tridax procumbens,” Int. J. Biol. Macromol. **125**, 99–108 (2019).

21. A.W. Pratama, B. Piluharto, D. Indarti, T. Haryati, and H.S. Addy, “Pengaruh Konsentrasi Asam Terhadap Sifat Fisik dan Muatan Permukaan Selulosa Termodifikasi,” ALCHEMY J. Penelit. Kim. **15**(2), 315–328 (2019).

22. A.W. Pratama, B. Piluharto, M. Mahardika, N. Widiastuti, A. Firmanda, and M.N.F. Norrrahim, “Comparative study of oxidized cellulose nanofibrils properties from diverse sources via TEMPO-mediated oxidation,” Case Stud. Chem. Environ. Eng. **10**, 100823 (2024).

23. M.N. Norizan, M.H. Moklis, A.H. Alias, A.I. Rushdan, M.N.F. Norrrahim, K. Abdan, and N. Abdullah, “Treatments of Natural Fibre as Reinforcement in Polymer Composites-Short Review,” Funct. Compos. Struct., (2021).

24. E. Syafri, Jamaluddin, Harmailis, S. Umar, M. Mahardika, D. Amelia, R. Mayerni, S. Mavinkere Rangappa, S. Siengchin, T.R. Sobahi, A. Khan, and A.M. Asiri, “Isolation and Characterization of New Cellulosic Microfibers from Pandan Duri (Pandanus Tectorius) for Sustainable Environment,” J. Nat. Fibers **19**(16), 12924–12934 (2022).

25. T. Narkkun, W. Kraithong, S. Ruangdit, C. Klaysom, K. Faungnawakij, and V. Itthibenchapong, “Pebax/Modified Cellulose Nanofiber Composite Membranes for Highly Enhanced CO 2 /CH 4 Separation,” ACS Omega **8**(48), 45428–45437 (2023).

26. P. Ajkidkarn, and H. Manuspiya, “Novel bacterial cellulose nanocrystals/polyether block amide microporous membranes as separators for lithium-ion batteries,” Int. J. Biol. Macromol. **164**, 3580–3588 (2020).