

CONFERENCE PROCEEDING

Synthesis of Regenerated Cellulose Nanofiber/glycerol to produce Bioplastic Composite

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ABSTRACT

Nowadays, people always use plastics in daily life as their needs to facilitate the carrying of large quantities of goods. As the population of people in the world increase, it has resulted in the number of plastics used constantly increasing as well. Moreover, the number of plastic wastes is also increasing as it takes a very long time to be degraded. Therefore, the process of making biodegradable plastics is necessary to reduce the number of plastic wastes. The aims of this study was to investigate the synthesis of regenerated cellulose and glycerol at different reaction times for bioplastic composite production. The EFB cellulose nanofibers were blended into the frozen solid of green solvent and stirred with glycerol at different times. A bioplastic membrane was formed after the solutions were cast, coagulated, and dried. FTIR analysis proved that the spectra of the plasticization cellulose display significant peaks in the range of 3000-3600 cm^{-1} and the intensity increases with increasing reaction time between cellulose and glycerol. The morphology of cellulose has changed after regeneration process. The results indicate this production of bioplastic composite had saved the earth from being littered with plastic wastes as it is more eco-friendly for the environment and users.

Keywords: *Biodegradable plastics; FTIR; green solvent; plasticization*

INTRODUCTION

Plastic is one of the most abundant waste on earth and has caused many environmental problems because of its difficulty in biodegradability. Therefore, bioplastics have been developed in the present study and are widely used in industry as an alternative to reduce the use of conventional plastics. (Thiruchelvi *et al.*, 2021). Cellulose, the most common organic polymer, is an extensively used material in the manufacture of bioplastics because of its good strength, toughness, transparency and high surface gloss. However, there are some shortcomings due to its partially crystalline structure and hydrogen bonds which results in difficulty in the removal of molecules and the close packing of cellulose molecular chains through strong hydrogen bonding leads to stiff molecular chains. (Wang *et al.*, 2013)

Therefore, plasticization, a simple and useful method, was done to improve the chemical and physical properties of cellulose. Glycerol was used as a plasticizer and results in higher thermal stability, better film formation and tensile strength since glycerol is a hydrophilic substance and acts as a water container agent. (Teixeira *et al.*, 2021). Hence, this study aims to incorporate glycerol in cellulose for plasticization to achieve high quality, flexible and long-lasting cellulose-based polymer.

MATERIALS AND METHODS

Materials

Oil palm EFB cellulose nanofibers at desired sizes of 106 to 500 μm were purchased from Szetech Engineering Sdn Bhd (Selangor, Malaysia). Cellulose dissolution process was carried out using lithium hydroxide, LiOH and urea (Sigma Aldrich). The coagulation bath was prepared containing 5% sulphuric acid, H_2SO_4 (Sigma Aldrich). Glycerol was purchased containing 99.5% USP grade.

Dissolution and Regeneration of Cellulose/glycerol in Green Solvent

A green aqueous solvent containing 4.6 % LiOH, 15 % urea and 80.4 % distilled water was prepared and pre-cooled at $-13\text{ }^\circ\text{C}$ in a refrigerator. Consequently, the EFB cellulose nanofibers was blended into the frozen solid by extensively stirred at room temperature to obtain a transparent cellulose solution. Glycerol of 5% and was added into the cellulose solution and stir homogeneously until cellulose/glycerol solution were formed at different reaction time of 5, 10 and 20 minutes for plasticization process. The blended solutions were cast on a glass plate and immediately inserted into coagulation bath containing 5% H_2SO_4 solution. After a minute of solvent transfer between solution and the acid, a piece of transparent membrane was formed. Ultimately, the cellulose membrane was air dried to form a bioplastic membrane.

RESULTS AND DISCUSSION

Physical Properties

Figure 1 shows the physical structure of the samples. The sample of EFB cellulose in the form of wet cellulose (Figure 1(a)) was transform from Cellulose I (EFB cellulose) into cellulose II (regenerated cellulose membrane) as can be seen in Figure 1(b) after undergone regeneration process respectively. The regenerated cellulose film then was air-dried, and it was reconstructed into cellulose bioplastic (Figure 1(c)).



Figure 1. (a) wet EFB cellulose (b) regenerated cellulose/glycerol membrane and (c) regenerated cellulose/glycerol bioplastic

FTIR Analysis

The FTIR spectrum of cellulose and cellulose/glycerol at different reaction time are shown in Figure 2. The spectra of cellulose at wavenumber of $3000\text{--}3600\text{ cm}^{-1}$ are due to the O–H stretching vibration of the hydroxyl groups (Foster *et al.* 2018; Nguyen & Lee 2021). The wavenumber around $2882\text{--}2933\text{ cm}^{-1}$ is due to the C–H stretching vibrations. Meanwhile, the spectra of the plasticized cellulose (cellulose/glycerol) display significant peaks in the range of $3000\text{--}3600\text{ cm}^{-1}$ and the intensity increases with increasing reaction time between cellulose and glycerol. This can be attributed to the large number of hydroxyl groups in glycerol. The existence of double peaks at 2933 cm^{-1} and 2882 cm^{-1} are due to the C–H stretching vibration in glycerol molecules, while the absorption peaks at 999 cm^{-1} and 1033 cm^{-1} are associated with its C–O–C and C–O stretching vibrations, respectively. Hence, these results suggest that glycerol

has reacted with cellulose (Nguyen & Lee 2021).

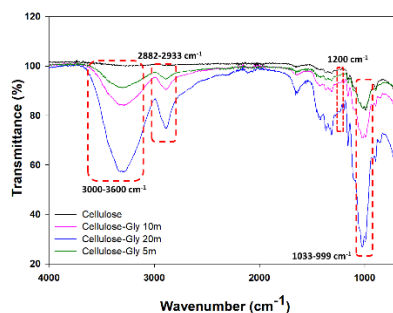


Figure 2. FTIR spectrum of cellulose and cellulose-glycerol reacted at 50, 10 and 20 minutes

Morphology Study

Figure 3 (a) shows SEM images on surface morphology of the cellulose and regenerated cellulose. The surface morphology cellulose shows single fiber arranged between one another. Meanwhile, regenerated cellulose shows smooth and porous structure which due to the drying process where water trapped has evaporated which has resulted in the formation of voids (Kaco *et al.*, 2014).

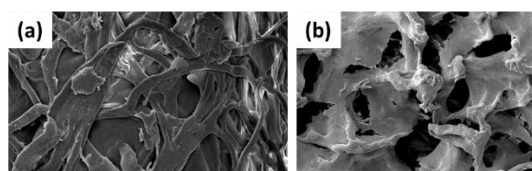


Figure 3. Surface morphology of (a) cellulose and (b) regenerated cellulose

CONCLUSION

Cellulose and glycerol have been reacted to produced plasticized regenerated cellulose/glycerol at different reaction time for bioplastic composite production. FTIR result shows the cellulose functional groups were changed after the existence of glycerol. Meanwhile, morphology of cellulose has alternated after regeneration process. Hence, this cellulose/glycerol bioplastic is potential to be used as bioplastic for food packaging and others application due to improve flexibility and suitable for wound dressing.

ACKNOWLEDGEMENT

The authors would like to thank Universiti Sains Islam Malaysia (PPPI/KGI/0119/051000/16019) and Universiti Kebangsaan Malaysia-Yayasan Sime Darby Chair for Sustainability (UKM-YSD-2021-006) for the financial support.

REFERENCES

- Foster , E.J., Moon, R.J. & Agarwal, U.P. (2018) Current characterization methods for cellulose nanomaterials. *Chemical Society Reviews*, 47,2609–2679
- Nguyen, S.V. & Lee, B.K. (2021). Microfibrillated cellulose film with enhanced mechanical and water-resistant properties by glycerol and hot-pressing treatment. *Cellulose*, 28, 5693–5705
- Kaco, H., Zakaria, S., Chia, C.H. & Zhang, L. (2014). Transparent and printable regenerated kenaf cellulose/PVA film. *BioResources* 9(2), 2167-2178
- Teixeira, S. C., Silva, R. R. A., de Oliveira, T. V., Stringheta, P. C., Pinto, M. R. M. R., & Soares, N. D. F. F. (2021). Glycerol and triethyl citrate plasticizer effects on molecular, thermal, mechanical, and barrier properties of cellulose acetate films. *Food Bioscience*, 42, 101202
- Thiruchelvi, R., Das, A., & Sikdar, E. (2021). Bioplastics as better alternative to petro plastic. *Materials Today: Proceedings*, 37, 1634–1639

Wang, Q., Cai, J., Zhang, L., Xu, M., Cheng, H., Han, C. C., Kuga, S., Xiao, J., & Xiao, R. (2013). A bioplastic with high strength constructed from a cellulose hydrogel by changing the aggregated structure. *Journal of Materials Chemistry A*, 1(22), 6678