Progress in Engineering Application and Technology Vol. 3 No. 2 (2022) 046–057
© Universiti Tun Hussein Onn Malaysia Publisher's Office

PEAT

Homepage: http://publisher.uthm.edu.my/periodicals/index.php/peat
e-ISSN : 2773-5303

Thermal And Mechanical Properties of Regenerated Nanocellulose Reinforced Polylactic Acid Composites: A Systematic Review

Nurhanani Nor[1], Mazatusziha Ahmad[1]*, Mohd Nazrul Roslan[1]

[1]Department of Chemical Engineering Technology, Faculty of Engineering Technology,
Universiti Tun Hussein Onn Malaysia, 84600 Pagoh, Johor, MALAYSIA

*Corresponding Author Designation

DOI: https://doi.org/10.30880/peat.2022.03.02.005
Received 26 January 2022; Accepted 20 July 2022; Available online 10 December 2022

Abstract: Regenerated nanocellulose (NC) is produced by converting natural nanocellulose to soluble cellulose derivatives and dissolving in ionic liquid (ILs) solution. The main objective of the study was to analyze the effect of different categories of nanocellulose and size on thermal and mechanical properties of polylactic acid (PLA) composites. The effect of nanocellulose on thermal stability and mechanical properties of (PLA) composites was investigated. In this study, PRISMA method has been applied to identify accurate and reliable articles. The qualitative analysis covers 22 Systematic Review studies. Through the PRISMA method, finalization studies of 20 reports were done and further assessment on selected articles were evaluate. Composite with good interfacial adhesion and cellulose distribution between PLA and NC results to the high thermal and mechanical properties. Modification by ILs shows enhancement in the thermal and mechanical properties of NC/PLA composite. The nanocellulose type and size greatly affects these properties. Further, composite with higher thermal degradation temperature (Tonset and Tmax), results in better thermal stability. A systematic review from the research provides comprehensive information to determine the effectiveness of regenerated nanocellulose film as reinforcement agent in PLA composite. More detail is needed to standardize the used of modification type of ILs. Empirical studies are recommended to include sufficient statistical findings so that a meta-analysis could be conducted.
.

Keywords: Nanocellulose, Polylactic Acid, Composite, Thermal, Mechanical, Ionic Liquid

---

Corresponding author: maza@uthm.edu.my

1. Introduction

The increasing awareness on global environmental issues among consumers, industry and government results in increasing demands for products created from renewable and sustainable resources. Natural fiber-reinforced polymers (NFRP) composite which categorized as green composite materials provides an alternative for replacement of petroleum-based polymeric materials. Kenaf, jute, sisal, pulp, and bamboo fiber are the most commonly studied of a variety of natural sources where the cellulose was extracted. Regenerated nanocellulose is created by converting natural nanocellulose to a soluble cellulosic derivative and dissolving in a solution. Some ionic liquids have the ability to break down the strong intra- and intermolecular interactions found in nano cellulose and dissolve it. Withdrawn of it causes pollution emission to the environment and pretreatment by ionic liquid (ILs) is used to replace harmful organic solvents. Various studies have been carried out to discover more effective nanocellulose from ionic liquid pretreatment incorporated polymer composite. NFRPs manufactured from three frequently used thermoplastic polymer matrices, such as polypropylene (PP), polyethylene (PE), and polylactic acid (PLA) are discussed by Niu et al., (2021). However, PLA has been used as a matrix in bio-composites with natural fiber reinforcement. It is due to excellent mechanical properties, transparency, biodegradability, fat and oil resistance, and renewable resources. PLA-based composites have higher Young's modulus, tensile strength, and thermal stability than polypropylene (PP) based composites.

Fibrillating original fibers into nano and micron sized fibers can reduce defect points along the fibers due to surface modification when dissolved in ILs [16]. Reddy et al.,2014 reported self-reinforced microcellulose composite sheets with average tensile strength, modulus, and elongation of 135 MPa, 8150 MPa, and 3.2 percent, respectively. However, Niu et al., 2021 reported tensile strength, strain, and Young's modulus of CNF/PLA film are 230 MPa, 13.00 %, and 5.4 GPa, respectively. The properties of the nanofibrillated cellulose (NFC) increased dramatically than microfibrillated cellulose (MFC). Thermal stability is important for long-term durability, short time in degradation and anti-flammable. Thermal stability is indicated by thermal degradation temperature. This study will analyze the effect of categories and size of nanocellulose on the thermal and mechanical properties of polylactic acid (PLA) composites.

2. Materials and Methods

The PRISMA method was used to select related articles from credible resources such as ScienceDirect, Scopus, SpringerLink, Emerald Insight, Taylor Francis, SAGE, Oxford Journals, and Wiley Online Library.

2.1 PISMA

Preferred reporting items used in disability areas for systematic review and meta- Analysis or known as PRISMA are intended as guidelines for writing a systematic review. It enables the researchers to research accurately and reliably on thermal stability and mechanical properties concerning nanocellulose polymer composites.

2.2 Sources

Scopus and Web of Science (WOS) have been the principal database used in these studies. One of the most important abstract literature and citation database examined by peers. This database gives a comprehensive overview of science's progress worldwide, with clever tooling for research, visualization, and interpretation.

2.3 Eligibility and exclusion criteria

Only papers relating to the regenerated nanocellulose reinforced PLA composite properties have been recovered. Literature search in this study has been filtered by the following criteria; language, document type, source type and year of publication.

2.4 Process of systematic review

In an integrative systematic review that collects the articles by qualitative, quantitative, and mixed methodologies, a wide overview of papers and several research methodologies is possible [1,4]. The associated literature was accessed, searches have been set up to be titled, abstract or keyword. Few of these keywords have been utilized to obtain the related existing literature. The search string in the Scopus and WOS database is shown in Table 1 below. 45 articles had been screened and exclusive pieces resulted for some reasons indicated earlier as in Figure 1 [1,4,12]. The selection of 20 articles was reduced because it concentrated heavily on the subject of interest. Another has been omitted from the scale of eligibility due to not articles, non-English, and documents published more than 10 years ago.

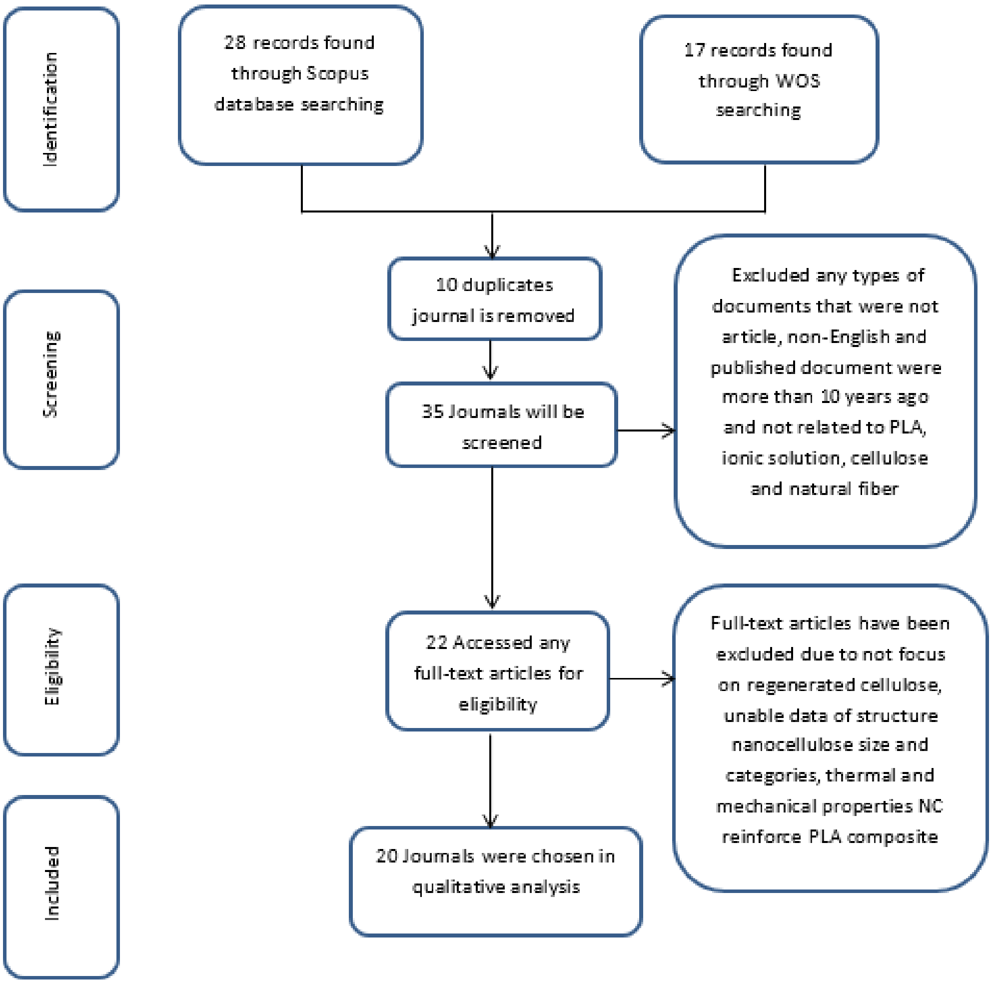

Figure 1: The flow diagram of the study

Table 1: The search string used for systematic literature reviewing

Journal database	Search string	Frequency of hits
Scopus	(TITLE-ABS-KEY ("ionic pretreatment" OR "ionic liquid") AND TITLE-ABS-KEY ("plant fiber" OR "natural fiber" OR "cellulose") AND TITLE-ABS-KEY ("thermal" OR "mechanical" OR "properties") AND TITLE-ABS-KEY ("polymer composite" OR "PLA" OR "polylactic acid"))	28
Web of Science (WOS)	TITLE- ARTICLE TERM ("ionic pretreatment" OR "ionic liquid") AND ("plant fiber" OR "natural fiber" OR "cellulose") AND ("thermal" OR "mechanical" OR "properties") AND ("polymer composite" OR "PLA" OR "polylactic acid"))	17

2.5 Data abstraction and analysis

Raw data such as temperature and PLA composite foam composition has been extracted in this investigation. The content analysis of the nominated papers is identified for meta-data and carried out to answer the study's investigative questions. Full paper analysis has only been performed after extensive analysis of abstracts.

3. Results and Discussion

Regenerated nanocellulose (NC) reinforce PLA composite have successfully organized. 20 studies were selected and pointed out for systematic review. Analysis focused on the size structure, degradation temperature, and strength of NC/PLA composite. Further investigation was made to inspect the effect of NC on the thermal stability and mechanical properties of composites film.

3.1 The effect of nanocellulose structure on the degree of crystallinity, thermal stability and mechanical properties

Nanocellulose is dissolved with an ionic liquid to improve the surface area and reduce the hydrophilic by breaking the hydrogen bonds [9]. The smaller size of nanocellulose led to a stronger interaction with PLA. When the size reduces due to modification by ILs, the surface area will increase. The increase of surface area was an essential factor to consider since it enhances the adhesion between two components. It was also indicating the improvement in thermal and mechanical properties of composite materials. The Microfibrillated cellulose (MFC) is similar to cellulose nanofibrils (CNF), with the only difference being the size of the cellulosic particles utilized. The orientated fibrils in regenerated MFC were uniform in size compared to before modification by ILs. The smallest size was CNC, which was treated by emm ILs less than 220 nm. Zhao et al. (2016) recorded that following modification changed CNC structure. It becomes shorter, and the sport of sphere becomes dominating in ACN/ILs.

The porosity may be related to enhancing chain flexibility and improving the functionality of PLA fiber while decreasing the size and increasing surface area. Table 2 shows the effect of composition polymer/nanocellulose on size between 3 authors. If the content of PLA in composite PLA/MCF/ILs decreases, the particle size decreases. However, when composition was 1/17 the size increases this due to dominance of cellulosic fibrils with non-uniform sizes or interference between the PLA matrix [7]. Yet, The MCC incorporation PLA is the smallest size compared to others.

Table 2: The effect composition of nanocellulose reinforcement composite on size

Authors	Types of nanocellulose	Sample	Composition of Polymer/cellulose (wt%)	Display structure	Size (μm)
(Khaw et al., 2019)	MCC	PLA/MCC	99/1	visible with non-uniform dispersion	4.48
		PLA/MCC/ILs	94/1	Not visible	N/A
(Shamshina et al., 2018)	MCF	PLA/MCF/ILs 6	17/17	N/A	125 ± 27
		PLA/MCF/ILs 5	13/17		172 ± 33
		PLA/MCF/ILs 4	8/17		116 ± 42
		PLA/MCF/ILs 3	5/17		106 ± 4
		PLA/MCF/ILs 2	4/17		90 ± 2
		PLA/MCF/ILs 1	1/17		122 ± 8
(Guo et al., 2012)	MCC	PLA/MCC/ILs	N/A	N/A	0.006 to 0.052

Table 3: The comparisons crystalline parameters of PLA and different types of nanocellulose/PLA with and without ILs

Authors	Types of nanocellulose	Sample	Glass transition temperature, Tg (°C)	Melting temperature, Tm (°C)	Crystallization temperature Tc (°C)	Xc (%)
(Khaw et al., 2019)	MCC	PLA	60.58	154.48	119.61	17.73
		PLA/ILs	55.09	139.20	98.77	25.26
		PLA/MCC	63.00	149.50	112.32	28.92
		PLA/MCC/ILs	54.39	141.00	101.69	34.68
(Luan et al., 2013)	ACN	PLA	60.00	N/A	N/A	N/A
		PLA/ILs/ACN	141.96			
(Zhao et al., 2016)	ACN	PLA	60.7 ± 0.2	166.4 ± 0.3	132.2 ± 0.8	1.6 ± 0.4
		PLA/ACN I/ILs	61.7 ± 0.2	165.8 ± 0.4	123.1 ± 0.6	3.6 ± 0.2
		PLA/ACN II/ILs	60.1 ± 0.8	164.2 ± 0.4	120.4 ± 0.4	3.4 ± 1.0
(Shamshina et al., 2018)	MFC	PLA	66	N/A	N/A	N/A
		MFC/IL	N/A	N/A		
		PLA/MFC/IL	N/A	182		

Table 3 gave more information about the degree of crystallinity, Xc % based on crystalline parameters. PLA has a glass transition temperature (Tg) of 50 to 80 °C and a melting temperature of 170 to 180 °C, depending on the amount of residual monomer. When MCC/ACN is applied to PLA, it increases the film rigidity and raises the temperature over that PLA. This has a greater effect because the molecular weight of the polymer chain has increased or the mobility of the polymers has decreased. The decrease in Tg of the composites indicates that ILs is an effective solvent for disentangling polymer chains during heating and shearing. The presence of ILs reduces the amorphous area, and their content leads to an increase in crystallinity.

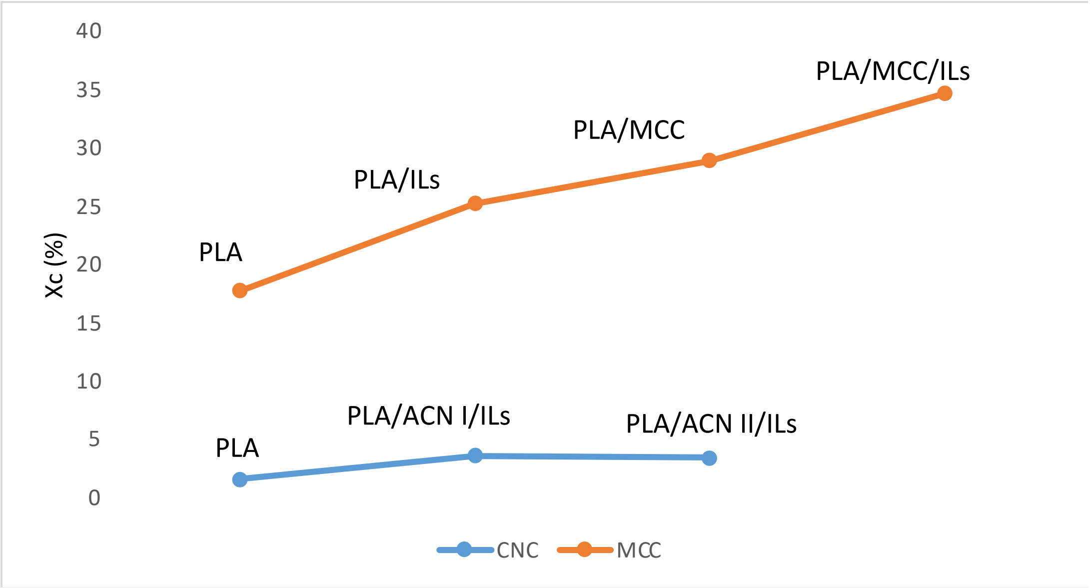

Figure 2: Comparison trend degree of crystallinity of different categorizes of nanocellulose between two authors

PLA/ ACN/ ILs had a higher Tg but lower Tm and Tc values than PLA/MCC/ILs, according to the retreatment composite film. Higher crystallinity was associated with lower Tc, particularly in PLA composites. MCC has greater crystallinity than ACN because to the higher presence, although the mechanical characteristics improve. Processing technique has an impact on the amount of crystallinity in a material. Plastics having a larger percentage of crystalline areas take longer to solidify in melt processing. When the temperature rises over Tg, amorphous materials lose strength quickly, but semicrystalline materials retain their mechanical capabilities until the temperature reaches Tm. Figure 3.1 indicates the degree of crystallinity trend with different categorizations of nanocellulose. MCC has greater crystallinity than ACN because to the higher presence, although the mechanical characteristics improve with length. Processing technique has an impact on the amount of crystallinity in a material. When the temperature rises over Tg, amorphous materials lose strength quickly, but semicrystalline materials such as PLA/CEL retain their mechanical capabilities until the temperature reaches Tm.

3.2 Thermal properties of nanocellulose reinforced polylactic acid (NC/PLA)

Thermogravimetric analysis (TGA) measurements were commonly used to study the thermal characteristics of retreatment cellulose/polymer. Thermal stability was studied because it was a critical factor in determining the film's application at high temperatures. In their native condition, cellulose have a crystalline and amorphous structure. Yet, the most hydrophobic cellulose crystals have the highest thermal stability [2]. Due to the increased hydrophobic, weight loss happens at the tested temperature range. Grafting with PLA side chains may disturb the crystalline structure of cellulose, resulting in lower thermal stability. Indicate that chemical modification, such as dissolution in ILs, has occurred to enhance the properties [6,14,15].

Table 4: The effect of degradation temperature parameters with different types of nanocellulose

Authors	Type of nanocellulose	Sample	Tonset (◦C)	Tmax (◦C)	Temperature decomposition (◦C)
(Fox et al., 2016)	CNC	CNC	181	N/A	<200
(Yu et al., 2020)	N/A	PLA	285.4	339.3	N/A
		PLA/PILs	275.5	351.2	
(Khaw et al., 2019)	MCC	PLA	323	N/A	
		MCC	308		315-400
(Luan et al., 2013)	MFC/CNC	PLA	363.53	384.41	N/A
		MFC	340.32	360.15	
		PLA/ILs/ACN	295.55	400.57	
(Zhao et al., 2016)	CNC	PLA	334 ± 1.6	358 ± 0.6	N/A
		CNC I	148 ± 2.0	169 ± 0.5	
		CNC II	155 ± 1.5	171 ± 0.4	
		ACN I/ILs	222 ± 2.2	254 ± 0.8	
		ACN II/ILs	238 ± 1.8	254 ± 0.6	
		PLA/ACN I/ILs	348 ± 1.5	372 ± 0.5	
		PLA/ACN II/ILs	350 ± 2.0	373 ± 1.0	
(Xian et al., 2018)	MCC/MFC	MCC	310	368	N/A
		MFC	245	335	
		PLA	333	373	
		PLA /MCC	315	352	
(Shamshina et al., 2018)	MFC	PLA	245	300	N/A
		MFC	248	356	
		PLA/MFC	294	360	
(Xu et al., 2019)	MFC	PLA/MFC/IL	N/A	N/A	244-293
		MFC/IL			220
		PLA			332
(Zhang et al., 2014)	MFC	MFC	N/A	345.7	346
		MFC/IL		286.3	N/A
		PLA		N/A	226.6-311.2
		MFC/PLA			348.5-361.6

Extrapolated onset decomposition temperatures (Td/Tonset) and the maximum decomposition temperature (Tmax) were used to define the decomposition process. Due to the high repeatability of the results, these were adopted as the thermal stability measurement standard. Tmax is the temperature at which the maximum rate of breakdown appears. A different result of neat PLA film indicates in the table. Luan et al., (2013) shows the highest degradation temperature (Tonset and Tmax) 363.53◦C and 384.41 ◦C compared to others. It leads to higher thermal stability and the differences of each PLA due to differences in resources [11]. Figure 3, 4, 5 where the bar graft identifies the degradation temperature onset of different types of material cellulose which show lower than PLA temperature. However, when incorporation cellulose with PLA composites the Tonset is higher than cellulose.

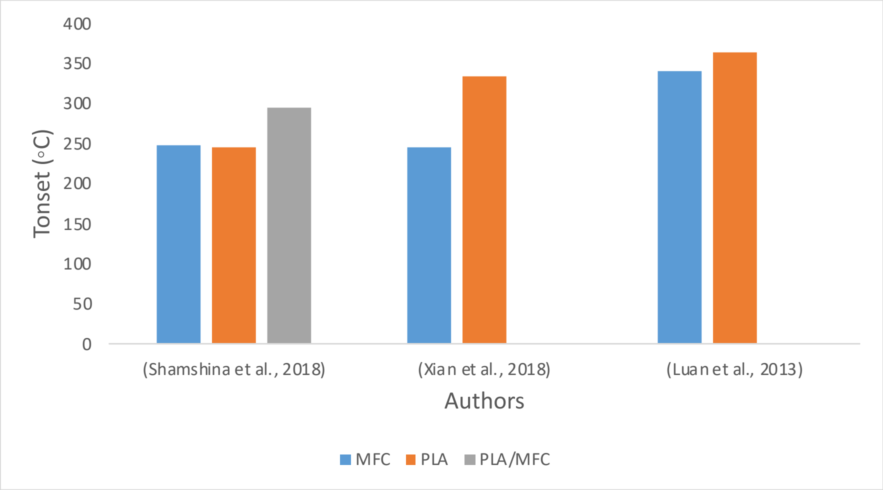

Figure 3: The comparison of MFC degradation onset before and after reinforcement with PLA composite

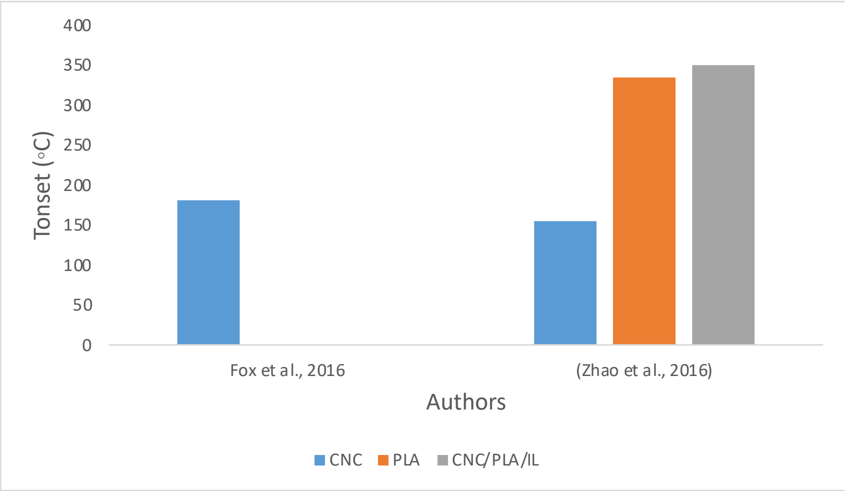

Figure 4: The comparison of CNC degradation onset before and after reinforcement with PLA composite

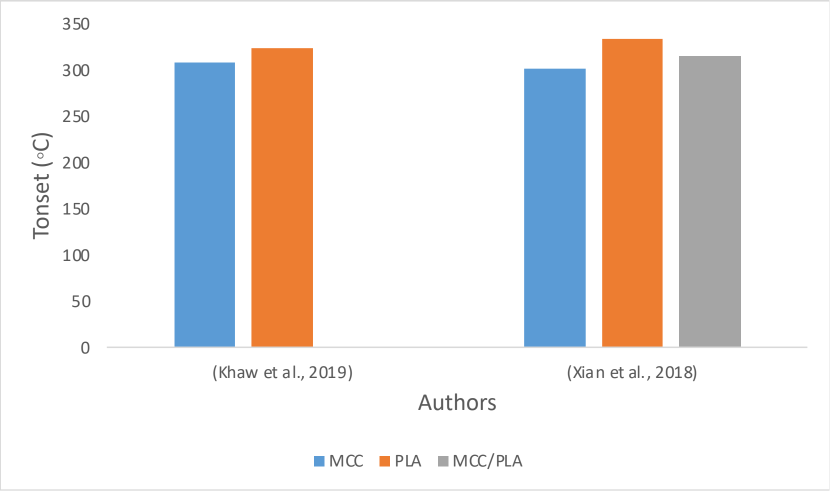

Figure 5: The comparison of MCC degradation onset before and after reinforcement with PLA composite

Moreover, for ANC II composite film when dissolve with ILs it recorded the highest Tonset was 350.00 °C but Tmax lower 27.57 °C compared to Luan et al., (2013). CNC was modified using acetic anhydride called (ACN). Zhang et al., (2014) described the reduced data MCC treat with ILs. When cellulose is treated with ILs, the molecular chain of the cellulose is disrupted when it is swollen or dissolved in the ILs, reducing the polymerization degree and thermal stability of the regenerated cellulose. Nonetheless, due to the acid sulfate groups in CNC, the retreatment CNC in ILs measured the lowest data, which was less than 200.00 °C. The presence of acid sulfur groups reduces the thermal stability of the dehydration process in general. Even after washing, some Na+ ions remained on the crystal surfaces, which might be the cause of their poor thermal stability.

It shows that the Tonset 350 ± 2.00 °C and Tmax 400.57 °C have the best thermal stability. The greater the Tonset and Tmax, the better the thermal stability. When the temperature rose, it was more likely to degrade. The Tonset rose as the length of the grafted PLA chains grew longer. However, when cellulose is dissolved with ILs, the particle length of the cellulose is reduced. The production of PLA long-chain branches on the backbone of cellulose disrupts segment mobility and induces chain entanglement, according to Zhang et al., (2014). Because a short or rigid polymer chain barely tangles, pulling the chains apart in solution or over a surface is simple. The negative effects of ILs are also minimized by using biocomposite composite film. Incorporation's cellulose/PLA blend's thermal stability has also been significantly increased. Because of the high interfacial adhesion, the biocomposites thermal stability was improved [17,18].

3.3 Mechanical Properties of nanocellulose reinforced polylactic acid (NC/PLA)

The material mechanical qualities are crucial for NC/PLA applications. As a result, the NC/PLA composite films' tensile strength, Young's modulus, and elongation at break were examined The ILs, which are recognized as effective cellulose solvents, expanded the compact structure of cellulose fiber. The strength of composite materials is largely determined by the interfacial area. Each fiber has its own contact with the matrix, and the strength of each fiber is determined by its surface area. Because of the tiny NC and its large specific surface area, as well as the development of interfacial adhesion between PLA and NC, the tensile modulus was improved [8, 17].

Table 5 stated the data result of composition, hardness, young modulus, E tensile strength, σ and elongation at break, ε of researchers. For the hardness, fewer data focus on it, but Khaw et al. (2019) reported the incorporation of ILs a little bit reduced the hardness of neat PLA, 21.1 to 10.5 MPa and PLA/MCC composites. However, the hardness value raised steadily when the two composites were incorporated after modification with ILs 10.5 to 14. Microhardness is increased by lowering the size of fillers. The shape, thermal stability, crystallization, and glass-transition temperatures of PLA composite films with and without IL were found to correspond to the mechanical hardness. The neat PLA that had the highest young’s modulus was 1780 ± 48 MPA than Yu et al., (2020). It is because of the differences of molecular weight PLA be used.

Table 5: The Comparison Between Composition, Hardness, Young Modulus, Tensile Strength and Elongation at Break

Author	Sample	Composition	Hardness	Young’s modulus (MPA)	Tensile strength MPa	Elongation at break mm/mm
(Khaw et al., 2019)	PLA	100:0	21.1	N/A	N/A	N/A
	PLA/ ILs	95:5	10.5			
	PLA/MCC	99:1	19.8			
	PLA/MCC/ILs	94:1:5	14			
(Yu et al., 2020)	PLA	100:0	N/A	11	0.9	0.32
	PLA/PILs	95:5		97.32	3.68	0.55
	PLA/PILs	90:10		95	3.5	0.58
	PLA/PILs	85:15		87	3.3	0.7

	PLA/PILs	80:20		81	3.28	0.77
(Xian et al., 2018)	PLA	100:0	N/A	206.00±22.89	67.35±0.67	6.40±0.61
	PLA/MCC	92:8		262.90±30.69	64.88 ±4.64	5.13±0.17
(Xu et al., 2019)	MFC/ILs	N/A	N/A	N/A	30	N/A
	PLA/MFC/ILs	99:1			36	
	PLA/MFC/ILs	97:3			46	
	PLA/MFC/ILs	95:5			39	
	PLA/MFC/ILs	9:1			24	
	PLA/MFC/ILs	7:3			19	
	PLA/MFC/ILs	1:1			15	
(Shamshina et al., 2018)	MFC/ILs	N/A	N/A	4200±200	71±2	N/A
	PLA/MFC/ILs	1:0.1		6400±300	83±5	
	PLA/MFC/ILs	1:0.25		5800±300	110±12	
	PLA/MFC/ILs	1:0.3		5900±300	112±7	
(Zhang et al., 2014)	MFC/ILs	N/A	N/A	N/A	N/A	7.5
	PLA/MFC/ILs					13.4
(Montes et al., 2016)	PLA	100:0	N/A	1780 ± 48	52.5 ± 4.2	7.28 ± 0.41
	PLA/F-CNC/ILs	44:1;55		1815 ± 176	54.7 ± 5.1	5.30 ± 0.31
(Dormanns et al., 2016)	MFC	100:0	N/A	6600 ± 500	67.3 ± 7.9	2.0 ± 0.9

Yu et al, if the ILs composition high than PLA the E and σ decrease but ε increase but it is still higher than pure PLA. Xian et al, (2018) perform when the MCC is introduced in PLA, E would increase but reduce in σ and ε. the addition of either MCC or other nanocellulose categorized would cause a large number of stress concentration points. It would reduce the material tension and reduce the elongation at break. So, the modification by ILs plays the main role in improving mechanical properties.

When the composition of nanocellulose increases the E and σ will decrease. Even though nanocellulose in small size and its large specific surface area improve E but the increase of σ and ε must be investigated. PLA/MFC/ILs with composition ratio 97:3 show a high peak of tensile strength but lower than 1:0.1. The PLA/MFC/ILs by Shamshina et al. (2018) was the best film in mechanical properties. This is due to the highest value in young modulus and tensile strength. Though, PLA/MFC/ILs have better elongation at break 13.4 mm/mm than at the composition 44:1;55. The highest value may be due to the composition that is used for making up the film. Even though, the PLA/F-CNC/ILs lower than PLA/MFC/ILs in terms of ε but it shows an enhance in mechanical properties. The optimum mechanical properties of Young’s modulus were 5900±300 MPa, Tensile strength 112±7 MPa while elongation at break 13.4 mm/mm in this study.

The nanocellulose/PLA blend fiber's branching structure encouraged chain entanglement and allowed for substantially more chemical bond breakdown. It would result in enhanced mechanical characteristics by allowing for more efficient stress transfer under stress conditions. The presence of micrometric particles and particles of nanometric size affected the elongation at the break parameter. The smaller the distribution of sizes, the better. The breaking elongation of the film was much greater than that of pure cellulose film.

4. Conclusion

Nanocellulose categories and size are very important parameters for analyzing optimum mechanical and thermal properties. Many research journals focus on size, thermal and mechanical properties needed. Limitation of this study appeared to achieve the objective of analysis. Future study correlated on data to achieve the objective above. The optimum mechanical properties of Young's modulus were 5900±300 MPa, Tensile strength 112±7 MPa while elongation at break 13.4 mm/mm in this study. PLA/MFC/ILs was the best film form to enhance the mechanical properties. The highest degradation temperature (T_{onset} and T_{max}) 363.53 °C and 384.41°C compared to others. The greater the T_{onset} and T_{max}, the better the thermal stability. Analysis of nanocellulose reinforced plastic (PLA) found that composition plays a big role in improving properties of composite film. The findings provided comprehensive information and act as reference for research on thermal and mechanical properties of PLA. Future study needed to be run more on composition because it also plays an important role in improved composites.

Acknowledgement

The authors would also like to thank the Faculty of Engineering Tecnology, Universiti Tun Hussein Onn Malaysia for its support.

References

[1] Beller, E. M., Glasziou, P. P., Altman, D. G., Hopewell, S., Bastian, H., Chalmers, I., Gøtzsche, P. C., Lasserson, T., & Tovey, D. (2013). PRISMA for Abstracts: Reporting Systematic Reviews in Journal and Conference Abstracts. PLoS Medicine, 10(4). https://doi.org/10.1371/journal.pmed.1001419

[2] Fox, D. M., Rodriguez, R. S., Devilbiss, M. N., Woodcock, J., Davis, C. S., Sinko, R., Keten, S., & Gilman, J. W. (2016). Simultaneously Tailoring Surface Energies and Thermal Stabilities of Cellulose Nanocrystals Using Ion Exchange: Effects on Polymer Composite Properties for Transportation, Infrastructure, and Renewable Energy Applications. *ACS Applied Materials and Interfaces*, *8*(40), 27270–27281. https://doi.org/10.1021/acsami.6b06083

[3] Guo, Y., Wang, X., Shu, X., Shen, Z., & Sun, R. C. (2012). Self-assembly and paclitaxel loading capacity of cellulose-graft- poly(lactide) nanomicelles. *Journal of Agricultural and Food Chemistry*, *60*(15), 3900–3908. https://doi.org/10.1021/jf3001873

[4] Harris, J. D., Quatman, C. E., Manring, M. M., Siston, R. A., & Flanigan, D. C. (2014). How to write a systematic review. *American Journal of Sports Medicine*, *42*(11), 2761–2768. https://doi.org/10.1177/0363546513497567

[5] Khaw, Y. Y., Chee, C. Y., Gan, S. N., Singh, R., Ghazali, N. N. N., & Liu, N. S. (2019). Poly(lactic acid) composite films reinforced with microcrystalline cellulose and keratin from chicken feather fiber in 1-butyl-3-methylimidazolium chloride. *Journal of Applied Polymer Science*, *136*(24), 1–10. https://doi.org/10.1002/app.47642

[6] Luan, Y., Wu, J., Zhan, M., Zhang, J., Zhang, J., & He, J. (2013). “One pot” homogeneous synthesis of thermoplastic cellulose acetate-graft-poly(l-lactide) copolymers from unmodified cellulose. *Cellulose*, *20*(1), 327–337. https://doi.org/10.1007/s10570-012-9818-x

[7] Miao, C., & Hamad, W. Y. (2013). Cellulose reinforced polymer composites and nanocomposites: A critical review. Cellulose, 20(5), 2221–2262. https://doi.org/10.1007/s10570-013-0007-3

[8] Montes, S., Azcune, I., Cabañero, G., Grande, H. J., Odriozola, I., & Labidi, J. (2016). Functionalization of cellulose nanocrystals in choline lactate ionic liquid. Materials, 9(7), 1–12. https://doi.org/10.3390/ma9070499

[9] Mokhena, T. C., Sefadi, J. S., Sadiku, E. R., John, M. J., Mochane, M. J., & Mtibe, A. (2018). Thermoplastic processing of PLA/cellulose nanomaterials composites. Polymers, 10(12). https://doi.org/10.3390/polym10121363

[10] Niu, X., Huan, S., Li, H., Pan, H., & Rojas, O. J. (2021). Transparent films by ionic liquid welding of cellulose nanofibers and polylactide: Enhanced biodegradability in marine environments. Journal of Hazardous Materials, 402(September 2020), 124073. https://doi.org/10.1016/j.jhazmat.2020.124073

[11] Oza, S., Ning, H., Ferguson, I., & Lu, N. (2014). Effect of surface treatment on thermal stability of the hemp-PLA composites: Correlation of activation energy with thermal degradation. *Composites Part B: Engineering*, *67*, 227–232. https://doi.org/10.1016/j.compositesb.2014.06.033

[12] Page, M. J., Mckenzie, J. E., Bossuyt, P. M., Boutron, I., Hoffmann, T. C., Mulrow, C. D., Shamseer, L., Tetzlaff, J. M., Akl, E. A., Brennan, S. E., Chou, R., Glanville, J., Grimshaw, J. M., Hróbjartsson, A., Lalu, M. M., Li, T., Loder, E. W., Mayo-wilson, E., Mcdonald, S.Yhec, C. (2021). The PRISMA 2020 statement: An updated guideline for reporting systematic reviews. Journal of Clinical Epidemiology, 134, 178–189. https://doi.org/10.1016/j.jclinepi.2021.03.001

[13] Reddy, K. O., Zhang, J., Zhang, J., & Rajulu, A. V. (2014). Preparation and properties of self-reinforced cellulose composite films from Agave microfibrils using an ionic liquid. Carbohydrate Polymers, 114, 537–545. https://doi.org/10.1016/j.carbpol.2014.08.054

[14] Salama, A., & Hesemann, P. (2020). Recent Trends in Elaboration, Processing, and Derivatization of Cellulosic Materials Using Ionic Liquids. *ACS Sustainable Chemistry and Engineering*, *8*(49), 17893–17907. https://doi.org/10.1021/acssuschemeng.0c06913

[15] Shamshina, J. L., Zavgorodnya, O., Berton, P., Chhotaray, P. K., Choudhary, H., & Rogers, R. D. (2018). Ionic Liquid Platform for Spinning Composite Chitin-Poly(lactic acid) Fibers. ACS Sustainable Chemistry and Engineering, 6(8), 10241–10251. https://doi.org/10.1021/acssuschemeng.8b01554

[16] Tanpichai, S., & Wootthikanokkhan, J. (2018). Reinforcing abilities of microfibers and nanofibrillated cellulose in poly(lactic acid) composites. *IEEE Journal of Selected Topics in Quantum Electronics*, *25*(2), 395–401. https://doi.org/10.1515/secm-2016-0113

[17] Xian, X., Wang, X., Zhu, Y., Guo, Y., & Tian, Y. (2018). Effects of MCC Content on the Structure and Performance of PLA/MCC Biocomposites. *Journal of Polymers and the Environment*, *26*(8), 3484–3492. https://doi.org/10.1007/s10924-018-1226-3

[18] Xu, A., Wang, Y., Gao, J., & Wang, J. (2019). Facile fabrication of a homogeneous cellulose/polylactic acid composite film with improved biocompatibility, biodegradability and mechanical properties. *Green Chemistry*, *21*(16), 4449–4456. https://doi.org/10.1039/c9gc01918a

[19] Yu, Q., Qin, Y., Han, M., Pan, F., Han, L., Yin, X., Chen, Z., Wang, L., & Wang, H. (2020). Preparation and characterization of solvent-free fluids reinforced and plasticized polylactic acid fibrous membrane. *International Journal of Biological Macromolecules*, *161*, 122–131. https://doi.org/10.1016/j.ijbiomac.2020.06.027

[20] Zhang, Y., Li, X., Yang, Y., Lan, A., He, X., & Yu, M. (2014). In situ graft copolymerization of l-lactide onto cellulose and the direct melt spinning. *RSC Advances*, *4*(65), 34584–34590. https://doi.org/10.1039/c4ra02727b

[21] Zhao, J., Zhao, Y., Wang, Z., & Peng, Z. (2016). Effect of polymorphs of cellulose nanocrystal on the thermal properties of poly(lactic acid)/cellulose nanocrystal composites. *European Physical Journal E*, *39*(12). https://doi.org/10.1140/epje/i2016-16118-2