

The Effect of Paddy Straw Content on Properties of Regenerated Cellulose Biocomposite Films

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Abstract. Regenerated cellulose (RC)/paddy straw (PS) biocomposite films was prepared by addition of PS powder in partially dissolving microcrystalline cellulose into an 8% lithium chloride/N,N-dimethylacetamide (LiCl/DMAc) solution. The effect of paddy straw on tensile properties and crystallinity index (CrI) of RC/PS biocomposite films were studied. The results obtained revealed that the tensile strength and modulus of elasticity of RC/PS biocomposite films compared with pure regenerated cellulose was significantly improved at 5 wt% PS loading but decrease with the increment of PS. The increase in crystallinity index at 5 wt% proved the enhancement affected by the incorporation of PS.

1. Introduction

The development of biopolymer from renewable resources has become the objective to substitute the non-biodegradable petrochemical synthetic polymer [1,2]. Cellulose is a promising resource for developing the sustainable, biodegradable and environmentally friendly product as it is a dominant component in the vast majority of plant form with outstanding properties such as high flexibility, good thermal and chemical stability, biocompatibility, non-toxicity and low cost [3,4]. The utilization of cellulose not only protects the environment but also helps to curb the dependence on fossil fuels while maintaining the consumption of it [5biocam].

However, cellulose is difficult to process because of its crystalline form and the close packing and numerous number of hydrogen bonds makes it neither melt nor soluble in water or any common solvent [6]. Currently, several solvent have been used for regeneration of cellulose process. Most studies related to regenerated cellulose composite describe use of LiCl/DMAc as the solvent system for the dissolution of cellulose. There are some other non-derivatising cellulose solvent such as mineral acids, various molten hydrates and ionic liquid that also have been used in the processing of regenerated cellulose composite [6-8]. Incorporation of filler into polymer matrix has been proved to increase the polymer properties. Rice is the main agriculture crop that generate huge amount of agriculture waste in the form of straw accounting around more than 100 MT annually in India. Globally, paddy waste is the major crop left over annual generation of more than 770 MT [9]. Paddy



straw being a lignocellulose predominantly contains cellulose (32-47%), hemicellulose (19-27%) and lignin (5-24%) which are associated with each other through complex bonding [10].

In this study, the effect of PS powder loading on tensile properties and crystallinity index (CrI) of RC/PS biocomposite were investigated.

2. Eperimental

2.1. Materials

Paddy straw (PS) was obtained from Kilang Beras Bernas Kuala Perlis, Perlis. The collected agro residue was thoroughly washed and dried at 50 °C. The dried paddy straw was ground into powder and sieved. The average size of PS powder was 53 µm as measured by a Malvern particle size analyzer. The DMAc and LiCl were supplied by Merck and Acros Organic, while microcrystalline cellulose (MCC) was supplied by Sigma-Aldrich.

2.2. Preparation of RC/PS Biocomposite Films

Microcrystalline cellulose requires activation before dissolution in LiCl/DMAc. Activation procedure requires MCC to be immersed in distilled water followed by acetone and DMAc for 1 hour at room temperature. Activated MCC was dissolve in 8% (w/v) LiCl/DMAc. The cellulose solution was stirred for 1 hour until transparent solution was obtained. The viscous cellulose solution was casted on the glass plate for regeneration process to take place. The RC/PS biocomposite film was washed with distilled water and dried at room temperature. The formulation of RC/PS biocomposite films with different PS loading is shown in Table 1.

Table 1. Formulation of RC/PS biocomposite films

Materials	RC biocomposite films	RC/PS Biocomposite films
MCC (wt.%)	3	3
PS (wt.%)	-	5, 10, 15, 20

2.3. Tensile Testing

Tensile properties such as tensile strength, modulus of elasticity and elongation at break can be obtained from tensile test. Tensile testing was conducted using Instron 5569 Universal testing system by referring to ASTM D 882. A cross-head speed of 10 mm/min was used and test was carried out at room temperature. Five samples were tested with size of 50 x 15 mm was cut from each of RC biocomposite films.

2.4. X-Ray Diffraction

Crystallinity index (CrI) of RC biocomposite film samples was determined by X-ray diffraction method by using Bruker Advance model system. The pattern with Cu K α 1=1.5406Å at 30kV and 10mA were recorded from 2° to 35°. CrI as defined by Segal was used as a measurement of crystallinity.

$$\text{CrI (\%)} = (I_{002} - I_{\text{AM}}) / I_{002} \times 100 \quad (1)$$

Where I_{002} is the maximum intensity of the (022) lattice diffraction peak and I_{AM} is the intensity scattered by the amorphous part of the sample. The diffraction peak for plane (002) is located at the diffraction angle of around $2\theta=21^\circ\text{-}22^\circ$ and the intensity scattered by amorphous part was measured at lowest intensity at a diffraction angle around 16°

3. Result and Discussion

3.1. Tensile Properties

Figure 1 and 2 shows the tensile properties of RC/PS biocomposite films containing different paddy straw weight content. The tensile strength and modulus of elasticity are significantly increase with the incorporation of PS powder up to 5 wt% and decreases with the continuous addition of PS in RC biocomposite films. This could be attributed to high aspect amount of PS ratio in the system that leads to agglomeration causing poor dispersion in the RC matrix. The improvement in mechanical properties of RC/PS biocomposite film over pure RC film could be due to better filler interaction thus enhanced the properties for stress transfer. The elongation at break of RC/PS biocomposite films is shown in Figure 2. As the PS content further increase, the elongation at break reduced. This is due to the restriction of RC chain movement by the PS powder during deformation.

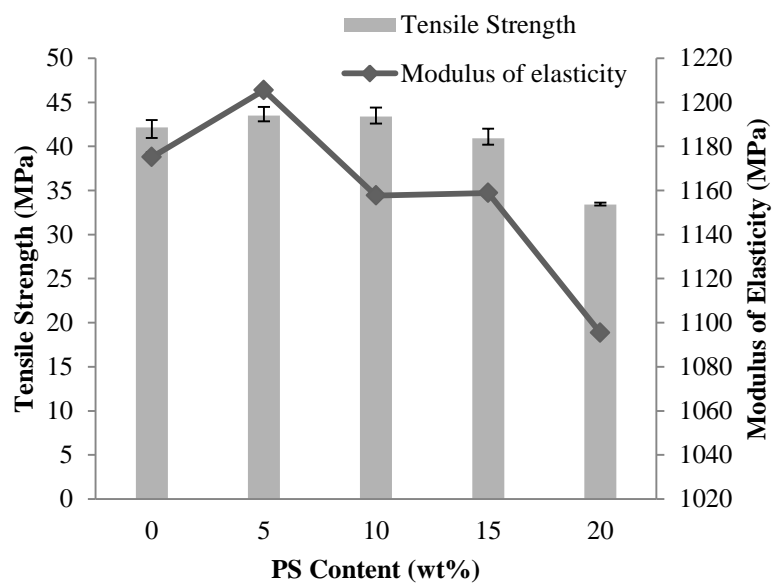


Figure 1: The effect of PS on tensile strength and modulus of elasticity of RC/PS biocomposite films

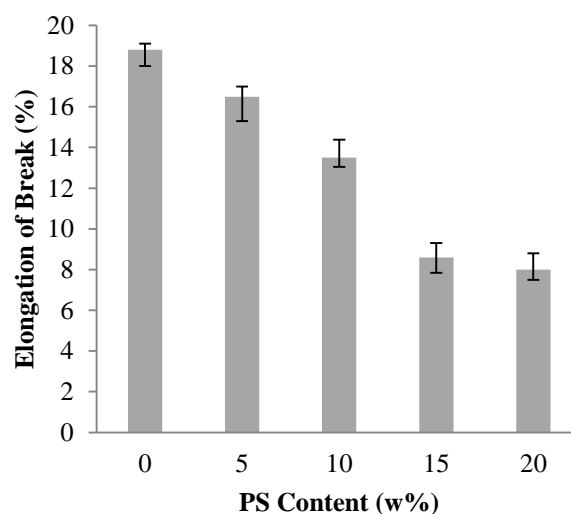


Figure 2: The effect of PS on the elongation at break of RC/PS biocomposite films

3.2. X-Ray Diffraction

XRD patterns of RC and RC/PS biocomposite films with different PS content are shown in Figure 3. The regenerated cellulose films exhibit characteristic peaks at $2\theta = 11^\circ$ - 12° which correspond to (110) plane, while $2\theta = 20^\circ$ and $2\theta = 22^\circ$ correspond to (110) and (020) planes. The diffraction peaks of RC/PS5 and RC/PS15 biocomposite films are very much similar to the pure RC film. This shows that PS was distinctly formed by the regenerated cellulose chain. The RC/PS biocomposite films with 15 wt% of PS show highest CrI as presented in Table 2. This could be attributed to the further increment of PS in the RC matrix increases the crystalline materials in the biocomposite films. Good interaction between PS and RC matrix also promotes the increment in crystallinity index of RC/PS biocomposite films.

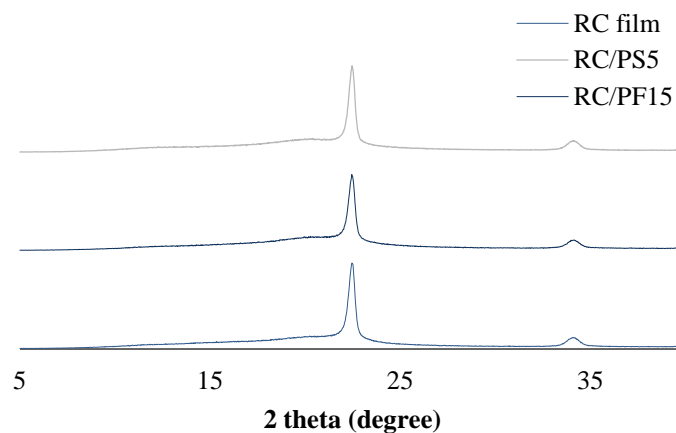


Figure 3: The effect of PS on the crystallinity index (CrI) of RC/PS biocomposite films

Table 2:

RC/PS biocomposite films	Crystallinity index (CrI)
RC/PS film 0 wt%	45.9
RC/PS film 5 wt%	46.0
RC/PS film 15 wt%	47.3

4. Conclusion

The incorporation of paddy straw in the regenerated cellulose biocomposite films improves the tensile strength and modulus of elasticity at 5 wt% of PS and decreased at higher PS loading. The elongation at break decreases with the increasing amount of PS loading in the biocomposite films. Crystallinity index (CrI) increases with the increment of PS content.

Acknowledgement

This research was supported by the Ministry of Higher Education (MOHE). The authors would like to thank the Fundamental Research Grant Scheme (FRGS), 9003 00601 for financial support.

References

- [1] Soheil Moghaddam M, Wahit M, U 2013 Development of regenerated cellulose/halloysite nanotube bionanocomposite with ionic liquid *International Journal of Biological Macromolecules* **58** 133-139.

- [2] Silva R D Vongsanga K Wang X Byrne N 2015 Development of a novel regenerated cellulose composite material *Carbohydrate Polymers* **121** 382-387
- [3] Soheilmoghaddam M Wahit M U Mahmoudian S Hanid N A 2013 Regenerated cellulose/halloysite nanotube nanocomposite films prepared with an ionic liquid *Materials Chemistry and Physics* **141** 936-943
- [4] Soheilmoghaddam M Wahit M U Whye W T Akos N I Pour R H Yussuf A A 2014 Bionanocomposites of regenerated cellulose/zeolite prepared using environmentally benign ionic liquid solvent *Carbohydrate Polymers* **106** 326-334
- [5] Pullawan T Wilkinson A N Eichhorn S J Discrimination of matrix-fibre interaction in all-cellulose nanocomposite *Composites Science and Technology* **70** **16** 2325
- [6] Duchemin B J C Newman R H Staiger M P 2009 Structure-property relationship of all cellulose composites *Composite Science and Technology* **69** 1225-1230
- [7] Huber T Mussig J Curnow O Pang S Bickerton S Staiger M P 2012 A critical review of all-cellulose composites *Journal of Materials Science* , **47**(3), 1171-1186.
- [8] Kauldhar B S Yadav S K 2018 Turning waste to wealth: A direct process for recovery of nano-silica and lignin from paddy straw agro-waste *Journal of Cleaner Production* **194** 158-166.
- [9] Kaur K Phutela U G 2016 Enhancement of paddy straw digestibility and biogas production by sodium hydroxide-microwave pretreatment *Renewable Energy* **92** 178-184.