

**TENSILE AND THERMAL PROPERTIES OF OIL  
PALM EMPTY FRUIT BUNCH REGENERATED  
CELLULOSE BIOCOMPOSITE FILMS USING  
IONIC LIQUID**

**NUR LIYANA IZYAN BT. ZAILUDDIN**

**UNIVERSITI MALAYSIA PERLIS**

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by

**NUR LIYANA IZYAN BT. ZAILUDDIN**

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## LIST OF ABBREVIATIONS

ABE	Acetone-Butanol-Ethanol
ACC	All-cellulose composites
ATR	Total reflectance
BMA	Butyl methacrylate
CN	Cellulose nanoparticle
CPH	Cocoa pod husk
DMAc	Dimethylacetamide
DMSO	Dimethylsulfoxide
DMI	1,3-dimethyl (2-imidazolium)
DP	Degree of polymerization
C <sub>2</sub> H <sub>5</sub> OH	Ethanol
FTIR	Fourier Transform Infrared Spectroscopy
H <sub>2</sub> SO <sub>4</sub>	Sulphuric acid
H <sub>3</sub> PO <sub>3</sub>	Phosphoric acid
IL	Ionic liquid
IR	Infrared Spectroscopy
LiCl	Lithium chloride
LODP	Leveling-off degree of polymerization
MAA	Methylacrylic acid
MCC	Microcrystalline cellulose
MFC	Microfibrillated cellulose
MMT	Montmorillonite
NaClO <sub>2</sub>	Sodium chlorite
NaOH	Sodium hydroxide
NFC	Nanofibrillated cellulose
NMMO	N-methylmorpholine-N-oxide
NMR	Nuclear Magnetic Resonance

OH	Hydroxyl groups
OPEFB	Oil palm empty fruit bunch
PF	Paraformaldehyde
PP	Polypropylene
RC	Regenerated Cellulose
RTIL	Room-temperature ionic liquids
SEM	Scanning Electron Microscopy
SRPM	Self-Reinforced Polymeric Matrix
TGA	Thermogravimetric Analysis
TSIL	Task specific ionic liquid
XRD	X-Ray Diffraction

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## LIST OF SYMBOLS

%	Percentage
wt%	Weight percentage
$\theta$	Theta
°C	Degree Celsius
°	Degree
CrI	Crystallinity Index
$T_{\text{dmax}}$	Temperature maximum degradation
$T_{300}$	Temperature degradation at 300 °C
$T_{600}$	Temperature degradation at 600 °C
$\mu\text{m}$	Micrometer
v/v	Volume per volume
w/v	Mass per volume
g	Gram
ml	Milliliter
mm	Millimeter
min	Minute
mg	Milligram

## Sifat-sifat Tensil dan Terma Filem-Filem Biokomposit Tandan Buah Kosong Minyak Sawit Selulosa Dijana Semula Menggunakan Cecair Berion

### ABSTRAK

Filem-Filem biokomposit selulosa dijana semula (SDS) daripada tandan buah kosong minyak sawit (TBKMS) dan selulosa berhablur mikro (SBM) disediakan menggunakan cecair berion. N, N Dimetilacetamida (DMAc) dan Litium Klorida (LiCl) digunakan sebagai sistem pelarut untuk melarutkan selulosa dijana semula pada suhu bilik. Separa terlarut SDS dan seterusnya bertukar kepada domain matrik terbenam diperkuat SDS yang tidak terlarut. Di dalam kajian ini kandungan SDS berbagai dari 1, 2, 3, dan 4 wt% dan SBM adalah ditetapkan pada 3 wt%. Kesan kandungan TBKMS dan modifikasi kimia menggunakan asid Butilmetakrilat (ABM) dan asid Metakrilat (AMA) ke atas pembelauan X-Ray, sifat-sifat tensil, morfologi, sifat-sifat termadan, FTIR filem biokomposit telah dikaji. Didapati pada kandungan 2 wt% TBKMS menunjukkan indeks penghabluran (IP), kekuatan tensil dan elastisiti modulus yang paling tinggi biokomposit filem SDS, tetapi pemanjangan pada takat putus adalah lebih rendah daripada kandungan-kandungan TBKMS yang lain. Suhu penguraian maksimum pengurangan berat  $T_{dmax}$  dan pengurangan berat pada suhu 300 °C ( $T_{300}$ ) berkurang dengan meningkatnya kandungan TBKMS, manakala pengurangan berat pada suhu 600 °C ( $T_{600}$ ) didapati meningkat. Kajian morfologi filem biokomposit TBKMS dijana semula menunjukkan pada kandungan 2 wt% TBKMS mempunyai penyebaran SDS yang lebih baik di dalam matrik. Modifikasi kimia TBKMS menggunakan ABM atau AMA menunjukkan peningkatan sifat-sifat filem biokomposit SDS dirawat. Indeks penghabluran, kekuatan tensil dan elastisiti modulus filem biokomposit SDS dirawat dengan ABM atau AMA adalah lebih tinggi dibandingkan dengan filem-filem biokomposit SDS tidak dirawat.  $T_{dmax}$  filem-filem biokomposit SDS dirawat dengan ABM atau AMA adalah lebih tinggi dibandingkan filem-filem biokomposit SDS tidak dirawat. Pada suhu penguraian  $T_{300}$  dan  $T_{600}$  menunjukkan filem-filem biokomposit SDS dirawat mempunyai pengurangan berat yang lebih rendah daripada filem-filem biokomposit SDS tidak dirawat. Ini menunjukkan bahawa filem-filem biokomposit SDS dirawat mempunyai kestabilan terma yang lebih baik. Peningkatan interaksi antara muka dan penyebaran filem-filem biokomposit SDS dirawat dengan ABA atau AMA telah dibuktikan dengan kajian SEM. Spektra FTIR filem-filem biokomposit RC dirawat menunjukkan pembentukan tindak balas pengesteran diantara selulosa dari TBKMS dan SBM dengan ABM atau AMA. Filem-filem biokomposit SDS dirawat dengan ABM mempunyai indeks penghabluran, kekuatan tensil, elastisiti modulus dan kestabilan terma yang tinggi dibandingkan filem-filem biokomposit SDS dirawat dengan AMA.

## Tensile and Thermal Properties of Oil Palm Empty Fruit Bunch Regenerated Cellulose Biocomposite Films using Ionic Liquid

### ABSTRACT

The regenerated cellulose (RC) biocomposite films from oil palm empty fruit bunch (OPEFB) and microcrystalline cellulose (MCC) were prepared using ionic liquid. N, N Dimethylacetamide (DMAc) and Lithium Chloride (LiCl) were used as solvent system to dissolve the regenerated cellulose, at room temperature. The partially dissolve of RC and subsequently convert onto matrix domain embedding the reinforcement of the non-dissolve RC. In this study, the OPEFB contents were varied from 1, 2, 3 and 4 wt% and MCC were fixed at 3 wt%. The effect of OPEFB contents and chemical modification using Butylmethacrylate acid (BMA) and Methacrylic acid (MAA) on X-Ray diffraction, tensile properties, morphology study, thermal properties and FTIR of RC biocomposite films were investigated. It was found that at 2 wt% of OPEFB contents showed the highest crystallinity index (CrI), tensile strength and modulus of elasticity of RC biocomposite films, but lower elongation at break than other OPEFB contents. The temperature at maximum rate of weight loss ( $T_{dmax}$ ) and weight loss at temperature 300 °C ( $T_{300}$ ) decreased with increasing OPEFB contents, while weight loss at temperature 600 °C ( $T_{600}$ ) increased. The morphology study of OPEFB RC biocomposite films exhibited at 2 wt% contents of OPEFB has better dispersion of RC into the matrix. The chemical modification of OPEFB using BMA or MAA indicated enhance the properties of treated RC biocomposite films. The  $T_{dmax}$  of treated RC biocomposite films with BMA or MAA were higher than the untreated RC biocomposite films. At temperature  $T_{300}$  and  $T_{600}$  degradation showed that treated RC biocomposite films with BMA or MAA have lower weight loss. This indicated that treated biocomposite films had better thermal stability. The enhanced of interfacial interaction and dispersion of treated RC biocomposite films with BMA or MAA were proven by SEM study. The FTIR spectra of treated RC biocomposite films indicated the formation of esterification reaction between cellulose from OPEFB and MCC with BMA or MAA. The treated RC biocomposite films with BMA have higher crystallinity index, tensile strength, modulus of elasticity and thermal stability compared to treated RC biocomposite films with MAA.

# CHAPTER 1

## INTRODUCTION

### 1.1 Research Background

Polymers from renewable resources have gain attentions worldwide due to their biodegradability and potential to substitute petrochemical which are diminishing (Guansen et al., 2012). The research and development of material biodegradable polymer has drawn considerable attention due to a new sustainable development concept (Zhang et al., 2012; Soheilmoghaddam et al., 2014). Cellulose as renewable resources in nature, is amongst the most widely used natural fibers (Chen et al., 2006; Kadokawa et al., 2009; Klemm et al., 2005; Kosam et al., 2008). However, the preparation of cellulose can be quite ambitious or strenuous. This is because the natural polymer does not melt or dissolve in usual solvent due to its inter and intra hydrogen bond, particularly crystalline structure, which strictly limiting its processing and applications (Chi & Zhang, 2006; Ruan et al., 2008; Zadagen et al., 2010; Han et al., 2013).

Regenerated cellulose is known as the man-made cellulose. In other word, it is a chemical dissolution of insoluble natural cellulose followed by the recovery of the material from the solution. The regenerated cellulose fibers have been made according to various processes yielding with a wide range of mechanical properties. The textile fibers with a low modulus, and high elongation at break, fibers for technical applications such as tire yarns with an intermediate modulus and strength, and fibers

with a high modulus and low elongation at break (Norholt et al., 2001). Regenerated cellulose fibers can be used to produce applications ranging from films, membranes and sponges (Klemm et al., 2005).

Recently, a new type of solvent, ionic liquid (ILs) has been the focus of interest due to their potential to be eco-friendly. In addition, they also exhibit characteristics including low melting points, low flammability, non-volatility, non-explosiveness, thermal stabilities and ease of recycling. Hence, ILs has the potential to replace traditional volatile organic solvents (Gatowshi et al., 2003; Mahmoudian et al., 2012). Cellulose can dissolve in several solvent systems which include heavy metal-ceramic complex solutions, concentrated metal salt, cold NaOH solution, thiocyanate/amine, dimethylacetamide (DMAc)/lithium chloride (LiCl), *N*-methylmorpholine-*N*-oxide (NMMO/H<sub>2</sub>O) system and concentrated H<sub>2</sub>SO<sub>4</sub> and H<sub>3</sub>PO<sub>3</sub> (Haibo et al., 2007).

The used of ILs can be seen in many forms ranging from media for organic synthesis and catalysts to lubricants and recently have generated much interest as replacements for environmentally, damaging volatile organic solvents (Berg et al., 2005). The emergence of ILs caters an extensive opportunity for the development of cellulose chemistry. The evolution of cellulose dissolution has been confirm by the use of ILs based on their unique and exclusive properties, such as low melting points, negligible vapor pressure, non-volatile and inflammable as well as wide electrochemical windows (Huddleston et al., 2001; Zhen et al., 2012).

Some researchers, it were found that cellulose can swell and dissolve in several solvents such as *N*-methylmorpholine-*N*-oxide (NMMO) (Biganska et al., 2002; Zhao et

al., 2007), *N*-dimethylacetamide/lithium chloride (DMAc/LiCl), (Zhang et al., 2012), 1-butyl-3-methylimidazolium chloride (Soheilmoghaddam & Wahit, 2013; Soheilmoghaddam et al., 2013; Mahmodian et al., 2012; Han et al., 2013; Chen et al., 2009; Liu et al., 2011), dimethyl sulfoxide/paratormaldehyde (DMSO/PF) (Jiang et al., 2012), 1,3-dimethyl (2-imidazolium)/lithium chloride (DMI/LiCl) (Tamai et al., 2004) and NaOH/urea solution (Kuo & Lee, 2009).

Encompassed by the solvents mentioned above, the two-component DMAc/LiCl solvent system is the most frequent used solvent for homogeneous cellulose modification and also dissolution of different types of cellulose without significant degradation (Granström, 2009; Vladimir et al., 2012). The dissolution process can be achieved in two ways which are by altering the ratio between the solvent and cellulose. In addition to that, an activation step is required for both processes (Granström, 2009). Since the solvent exhibit the advantage in dissolving cellulose, a variety of cellulose analysis by various methods was studied such as Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron Microscope (SEM) (Araki et al., 2006).

The groundwork of regenerated cellulose fibers through this dissolution method need to undergo the activation procedure in which the fiber is penetrated with a polar medium. Skipping the activation step may lead to the dissolution of cellulose to take up to several months (Tim et al., 2012). A study by Daisuke et al. (2008) showed that the DMAc-treated celluloses dissolved more rapidly as compared to the dissolution of the acetone-treated celluloses and the untreated one. It has been reported that the solubility of cellulose increases with LiCl content (Tim et al., 2012). Govindan et al. (2014) have conducted a researched on preparation and characterization of regenerated cellulose

using ionic liquid whereby the dissolution of micro crystalline cellulose (MCC) using DMAc/LiCl has improve the tensile strength and modulus of elasticity of the biocomposite films.

Palm oil fiber is one lignocellulosic material which is abundant in Malaysia. The use of palm oil biomass has taken the focus of researchers and scientists to exploit their properties. Palm oil is widely grown in Malaysia and its vast production has also lead to the vast production of biomass waste such as oil palm empty fruit bunch (OPEFB). Since the overloaded of these wastes that scattered the land, it is an idea to convert this useful waste into valuable product. OPEFB has showed potential as being natural fiber reinforcement in biocomposite sectors. The composition of OPEFB can be divided into cellulose (59.7 %), lignin (18.1 %), and hemicelluloses (22.1 %) (Geng, 2013).

Due to the fact that the cellulose content in OPEFB fiber is the most dominant constituent, it is best to further the study on the use of this cellulose to produce new bio-based product that is simply eco-friendly to the environment. One of the advantages is that the cellulose in OPEFB can be used to be regenerated into the production of biocomposite films. In order to form the regenerated cellulose, cellulose from OPEFB must dissolve in suitable solvents (Hassan et al., 2010; Chang, 2014).

To obtain the cellulose component prior to the dissolution in DMAc/LiCl solvent, usually pretreatment in the form of physical or chemical is carried out. The significant of conducting pretreatment is to break the lignin and the crystalline structure to obtain more cellulose content and thus, enhancing the dissolution ability of the solvent (Kihlman, 2012). Ariffin et al. (2008) reported the effect of physical, chemical

and thermal pretreatments of the oil palm empty fruit bunch (OPEFB) with increased in cellulose content and lignin while hemicelluloses decreased.

## **1.2 Problem Statements**

The study using ionic liquid with commercial microcrystalline cellulose and type of inorganic material has been reported in previous study (Mahmoudian et al., 2012; Soheilmoghaddam & Wahit, 2013; Soheilmoghaddam et al., 2014). However, the utilization of OPEFB as biocomposite regenerated cellulose has not yet been studied by using ionic liquid. Thus, this is one of many new methods can be used to utilize the use of OPEFB to produce green products such as biocomposite films with low cost and environmental friendly. However, OPEFB consist of complex structure of cellulose, lignin and hemicelluloses and is regarded as a material that is difficult to digest. To produce regenerated cellulose, extraction of cellulose from OPEFB is essential to separate the cellulose from the lignin and hemicelluloses that can prevent the dissolution or penetration of solutions. The study using regenerated cellulose DMAc/LiCl solvent is not yet reported. In order to improve the properties of biocomposite films, the chemical treatment of regenerated cellulose OPEFB was investigated.

### 1.3 Objectives

The objectives of this research are:

1. To study the effect of different OPEFB contents of regenerated cellulose biocomposite films using *N*-dimethylacetamide/lithium chloride (DMAc/LiCl) based on the X-ray diffraction, tensile properties, morphology, thermal properties and Fourier transform infrared spectroscopy analysis.
2. To investigate the effect of different chemical treatments such as, butyl metacrylate (BMA) and methacrylic acid (MAA) of OPEFB on the properties of regenerated cellulose OPEFB biocomposite films.

### 1.4 Scope of Study

The scope of this study involves several testings such as tensile properties, morphology, thermal properties, Fourier transform infrared (FTIR) and X-Ray diffraction (XRD). Tensile properties are used to measure the tensile strength, elongation at break and modulus of elasticity. Scanning electron microscope (SEM) is an imaging technique used to observe the morphology of the regenerated cellulose biocomposite films. Thermogravimetric analysis (TGA) is used to determine the thermal properties of the regenerated cellulose biocomposite films. Fourier transform infrared (FTIR) is used to identify the functional groups of regenerated cellulose biocomposite films. X-Ray diffraction (XRD) is explained to study the crystallinity of regenerated cellulose biocomposite films.

## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 Regenerated Cellulose

Regenerated cellulose is known as the man-made cellulose which was introduced in 1846 where Karl-Friedrich Schönbein discovered cellulose nitrate. In addition to that, cellulose xanthate or commonly known as Viscose was discovered in 1891 by Cross, Bevan, and Beadle (Roder et al., 2009).

Recently, it has been reported of a highly oriented cellulose fiber with high modulus and tenacity properties spun from an anisotropic solution in phosphoric acid using an air gap. Liquid crystalline solutions are known to be good precursors for high modulus/high tenacity yarns. The advantage of using a liquid crystalline solution for fiber production is the location orientation order of the chains is already at such a level they can be transformed into highly oriented fibers without the necessity of an after-treatment, which is required in spinning from isotropic melts or solutions of flexible polymers (Norholt et al., 2001).

The steps in cellulose regeneration started when some parts of the cellulose began to swell when dissolve in a particular solvent. This swollen cellulose will be transformed into the matrix phase covering the non-dissolved part of cellulose which acts as the reinforcement. After partial dissolution of cellulose, the solvent will be removed by using a coagulant (for example water) followed by evaporative drying. This

cellulose regeneration is crucial as it controls the precipitant of the final cellulose phase. The rate of regeneration tends to decrease with cellulose phases of higher crystallinity. This is attributed to the dissolved cellulose chains having more time to order themselves into a lower energy configuration (Huber et al., 2012). A study by Gindl & Keckes (2005) showed that 10 % of regenerated cellulose matrix is formed around 90 % of the non-dissolved cellulose after cellulose dissolution.

The applications of regenerated cellulose are often used to produce films, and sponges (Klemm et al., 2005). For instance, viscose fibers (Rayon) have excellent properties for a broad product range, from wet-strength cotton-like textile fibers like Modal fibers to technical fibers in the form of cord which is known as Rayon commonly used in high-performance tires. Up to now, the viscose technology is still being adopted in the production of film which is particularly important for food casing products (Klemm et al., 2005).

Currently, a great variety of man-made cellulose fibers is known but only a few are produced in industrial scale. For example, viscose (CV, staple and filament), modal (CMD), lyocell (CLY), cupro (CUP), acetate (AC), and triacetate (CTA). Cellulose fibers such as CV and CMD can be produced either by cellulose derivatisation followed by regeneration which involves the destruction of the derivative or by dissolution in a non-derivatising solvent like NMMO or ionic liquids (Roder et al., 2009).