



**CHARACTERIZATION AND BIODEGRADATION
STUDY ON CELLULOSE NANOCRYSTALS ISOLATED
FROM RICE STRAW / POLYVINYL ALCOHOL
NANOCOMPOSITES**

by

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LIST OF ABBREVIATIONS

BC	Bacterial cellulose
C	Carbon
Ca	Calcium
CNC	Cellulose nanocrystals
CNF	Cellulose nanofibrils
CO ₂	Carbon dioxide
CrI	Crystallinity index
Cu	Copper
DMAc	Dimethylacetamide
DSC	Differential scanning calorimetry
dTG	Derivative thermogravimetric
EA	Ethanedioic acid
Eb	Elongation at break
FAO	Food and Agriculture Organization of the United Nations
FESEM	Field Emission Scanning Electron Microscope
FTIR	Field Emission Scanning Electron Microscope
H	Hydrogen
H ₂ O	Water
H ₂ SO ₄	Sulfuric acid
HV	Hydroxyvalerate
KBr	Potassium bromide
LiCl	Lithium chloride
MCC	Microcrystalline cellulose

MVP	Moisture vapor permeation rate
N	Nitrogen
NaClO ₂	Sodium hypochlorite
NaOH	Sodium hydroxide
O ₂	Oxygen
OPH	PVOH hydrolase
PBS	Polybutylene succinate
PCL	Polycaprolactone
PE	Polyethylene
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoates
PHB	Polyhydroxybutyrate
PHBV	Poly(hydroxy butyrate-co-valerate)
pKa	Acid dissociation constant
PLA	Poly(lactic acid)
PQQ	Pyrrroloquinoline quinone
PS	Polystyrene
PU	Polyurethane
PVADH	PVA dehydrogenase
PVC	Polyvinyl chloride
PVOH	Polyvinyl alcohol
rRNA	Ribosomal ribonucleic acid
S	Sulfur
SAO	Secondary alcohol oxidase
SEM	Scanning electron microscope

TEM	Transmission electron microscopy
TEMPO	2,2,6,6-tetramethylpiperidine 1-oxyl radical
TGA	Thermogravimetric analysis
USA	United States
UV	Ultraviolet
XRD	X-ray powder diffraction

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LIST OF SYMBOLS

%	percentage
wt%	weight percentage
g	gram
kg	kilogram
mg	milligram
°C	degree celcius
mm	millimeter
nm	nanometer
µm	micrometer
MPa	mega pascal
GPa	giga pascal
h	hour
L	liter
ml	milliliter
wt%	weight percentage
g/cm ³	gram per cubic centimeter
g/m ²	gram per meter square
kV	kilovolt
cm ⁻¹	reciprocal wavelength
cm ₃	cubic centimeter
ΔH _{exp}	experimental melting enthalpy value of the crystalline phase
ΔH _{100%}	theoretical melting enthalpy value of PVOH crystalline phase
J/g	joule/gram

A	area
t	time
T ₁₀	temperature for 10% weight loss
T ₃₀	temperature for 30% weight loss
T _{MAX}	temperature for maximum weight loss
T _M	melting temperature
μ	micro
°	degree
α	alpha
β	beta
Δ	heat

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Kajian Ciri-Ciri dan Biodegradasi Terhadap Nanokristal Selulosa yang Diasingkan Daripada Jerami Padi / Polivinil Alkohol Nanokomposit

ABSTRAK

Penyelidikan ini meneliti pengekstrakan selulosa dari jerami padi, yang merupakan produk sisa pertanian untuk menghasilkan CNC dan penggunaannya untuk penghasilan filem nanokomposit yang boleh biodegradasi dan mesra alam. CNC yang diekstrak dari jerami padi melalui kaedah rawatan alkali siklik dan pemutihan muncul sebagai kristal panjang yang ditentukan dengan nisbah aspek tinggi 41. Polivinil alkohol (PVOH), polimer biodegradasi dan biokompatibel yang popular dipilih sebagai matriks polimer. Filem bionanokomposit PVOH / CNC yang dihasilkan dengan menggunakan kaedah tuangan dengan peratusan yang berbeza untuk menentukan peratusan CNC optimum. Didapati bahawa penggabungan CNC telah meningkatkan kekuatan tegangan sebanyak 60.4%, suhu degradasi maksimum hingga 287 °C, kadar kebolehtelapan wap air dan rintangan pembengkakan dan kelarutan PVOH / CNC. Jumlah CNC optimum didapati pada 3% peratus. Untuk mengatasi masalah hidrofilik dan mempelbagaikan penggunaan bionanokomposit, bionanokomposit yang lebih serba boleh dikembangkan dengan pengenalan asid etanedioik (EA) sebagai penghubung silang. Nanokomposit bersilang menunjukkan peningkatan yang ketara di mana kekuatan tegangan meningkat sebanyak 104.8%, suhu degradasi maksimum hingga 364 °C, kadar kebolehtelapan wap air, kelarutan dan rintangan bengkak. Akhirnya, kajian biodegradasi adalah penyelidikan yang menarik untuk pengembangan pembuangan sampah plastik yang lebih efisien dan mesra alam. Karya ini juga bertujuan untuk mengkaji tingkah laku biodegradasi dan bakteria yang merosakkan bionanokomposit PVOH / CNC yang berhubung silang dan tidak berhubung silang melalui kaedah penguburan tanah semula jadi. Bacteria yang merosot diasingkan dan dikenal pasti sebagai *Bacillus cereus* strain CCM 2010 dan *Bacillus cereus* strain ATCC 14579 dari penjujukan gen 16S rRNA. Perubahan kumpulan berfungsi sebelum dan sesudah biodegradasi disahkan melalui Fourier-transform inframerah spektroskopi. Ujian tegangan menunjukkan bahawa kekuatan tegangan dan pemanjangan sehingga putus berkurang apabila masa pengebumian tanah meningkat. Kajian morfologi menunjukkan sejauh mana kemerosotan permukaan nanokomposit sebelum dan selepas penguburan tanah, di mana penambahan EA menunjukkan kemerosotan minima. Berat nanokomposit juga menurun setelah berlaku hubungan silang. Suhu lebur dan kristalinitas meningkat dengan penambahan CNC tetapi dikurangkan setelah hubungan silang. Suhu lebur dan kristaliniti semua bionanokomposit meningkat selepas biodegradasi. Biodegradasi bionanokomposit disimpulkan dalam urutan penurunan berikut: PVOH/CNC > PVOH/EA/CNC > PVOH > PVOH/EA.

Characterization and Biodegradation Study on Cellulose Nanocrystals Isolated from Rice Straw / Polyvinyl Alcohol Nanocomposites

ABSTRACT

This research investigates the extraction of cellulose from rice straw, which is an agricultural waste product to produce CNC and its utilization for the development of biodegradable and environment friendly nanocomposite films. CNC extracted from rice straw through cyclic alkaline and bleaching treatment method appeared as long, well-defined rodlike crystals with high aspect ratio of 41. Polyvinyl alcohol (PVOH), a popular biodegradable and biocompatible polymer was chosen as the polymer matrix. PVOH/CNC bionanocomposite films were made through employing solution casting method with different weight percentages to determine the optimum CNC weight percentage (wt%). It was found that 3 wt% CNC incorporation vastly improved the tensile strength by 60.4%, maximum degradation temperature to 287 °C, water vapor permeability rate and swelling and solubility resistance of PVOH/CNC. The optimum CNC amount was found to be at 3 wt%. In order to address the issue of hydrophilicity and diversify the use of the bionanocomposite, a more versatile bionanocomposite was developed with the introduction of ethanedioic acid (EA) as a crosslinker. Crosslinked nanocomposite displayed significant improvements where tensile strength increased by 104.8%, maximum degradation temperature to 364 °C, water vapor permeability rate, solubility and swelling resistance. Finally, biodegradation study is an appealing investigation for the development of a more efficient and environmentally friendly plastic waste disposal. The current work also aimed to study the biodegradation behavior and its degrading bacterias of noncrosslinked and crosslinked PVOH/CNC bionanocomposites through natural soil burial method. The degrading bacterias were isolated and identified to be *Bacillus cereus* strain CCM 2010 and *Bacillus cereus* strain ATCC 14579 from 16S rRNA gene sequencing. The changes in functional groups before and after biodegradation were confirmed through Fourier-transform infrared spectroscopy. Tensile test revealed that the tensile strength and elongation at break reduced as time of soil burial increases. Morphological study showed the extent of surface deterioration of bionanocomposites before and after soil burial, where the addition of EA displayed lessened deterioration. Total weight loss also decreased after crosslinking occurred. Melting temperature and crystallinity increased with addition of CNC but reduced after crosslinking. Melting temperature and crystallinity of all nanocomposites increased after biodegradation for all bionanocomposites. Biodegradation of the bionanocomposites were concluded to be in the following decreasing order: PVOH/CNC > PVOH/EA/CNC > PVOH > PVOH/EA.

CHAPTER 1 : INTRODUCTION

1.1 Introduction

According to Plastics Europe (2019), the world plastic production is increasing each year where in the year 2018, the world plastic production reached an estimated value of 359 million tonnes from 348 million tonnes in 2017. Recently, filler materials for polymers obtained from agricultural waste products are popular in consequence to the public concern on reduction on waste generation. This issue can be solved through recovering and converting these wastes into profitable products, hence the “Waste to Wealth” terminology. Numerous types of fillers obtained from natural products such as rice husk, banana frond flour and tropical fruits have been directly incorporated in PVOH matrices and the biodegradability of the PVOH composites was proven (Arora, Kumar, & Kumar, 2012; Mat Suki, Ismail, & Abdul Hamid, 2014; Zhong, Ismail, Abdul Aziz, & Abu Bakar, 2011). The fabrication of poly (butylene succinate) based composites had shown improvement in tensile strength and elongation at break but at a high loading of 10 wt% of with rice straw flour (RSF) but with reduction in uniformity of filler dispersion within the polymer matrix (Bhattacharjee, Chakraborty, Kashyap, Gupta, & Katiyar, 2021). Wang et al. (2018) demonstrated that the adhesion of unextracted rice straw incorporated with high density polyethylene were found to be poor due to impurities involved. The mechanical properties and hygroscopicity results were inferior when compared to the composites with extracted rice straw.

Cellulose based PVOH composites were found to provide better results but the results are less satisfactory (A. Ali, 2017; Cazón, Vázquez, & Velazquez, 2018b; J. Chen

et al., 2017; Sreekumar, Manirul Haque, Afzal, Sadique, & Al-Harhi, 2019). Hence, nanocellulose based PVOH nanocomposites were created to address the discrepancies and the results obtained are far superior (Cazón, Velazquez, & Vázquez, 2019; Espinosa et al., 2019; Kohli, Garg, & Jana, 2019; W. Wang et al., 2020). For this study, biodegradable synthetic polymer (PVOH) was incorporated with nanocellulose, a renewable and naturally occurring polymer which is available in abundance. Cellulose nanocrystals (CNC) was chemically extracted from rice straw, an underutilized and poorly managed agricultural waste product with compelling cellulose content as the nanofiller material of interest while at the same time converting waste to wealth.

PVOH/CNC composite is known to be hydrophilic which limits its widespread application. Sonker et al. (2017) detailed that in order for PVOH to be accepted and used for extensive range of applications, it must be crosslinked to make it less hydrophilic. Glutaraldehyde (GA) and glyoxal are most commonly used as crosslinkers for polymers which contain hydroxyl and amine groups such as PVOH and cellulose (Ben Halima, 2016; Gadhawe, Mahanwar, & Gadekar, 2019; Gahlawat, Sen, & Das, 2015; T. Hou et al., 2019; K.J. Saunders, 2012; Shanshan Liu, Ge, Zou, & Chen, 2019; Park, Hwang, Yoon, Lee, & Lee, 2015). Although both of them are effective in crosslinking polymers, they are relatively toxic (Q. Liu et al., 2012). Crosslinkers such as epichlorhydrine (K. Das et al., 2010; Sreedhar, Chattopadhyay, Karunakar, & Sastry, 2006), formaldehyde (Frone, Nicolae, Gabor, & Panaitescu, 2015; Shuai Liu et al., 2015), hexamethoxymethylmelamine (L. Chen, Imam, Gordon, & Greene, 1997; Zhong, Ismail, Aziz, & Bakar, 2011) and sodium trimetaphosphate (Chaouat et al., 2008; Morandim-Giannetti, Silva, Magalhães, Schor, & Bersanetti, 2016) has also been extensively researched but they are not preferable due to their toxicity. Therefore, less toxic

crosslinkers are of uttermost interest in crosslinking reaction, particularly to maintain the biodegradability of the polymers and to expand its application.

In this research, a naturally occurring dicarboxylic acid namely ethanedioic acid (EA) was used as a crosslinker to form hydroxyl group linkages between the hydroxyl group of PVOH in order to improve on the tensile and water barrier properties of the PVOH/CNC nanocomposites. High concentrations of ethanedioic acid is usually found in leafy vegetable such as spinach and rhubarb or beverages including chocolate or tea (Abratt & Reid, 2010; Yongliang Chen, Ye, Li, Li, & Li, 2018; Gadd et al., 2014; Prasad & Shivay, 2017; Wijerathna et al., 2018). According to Uren (2018), the versatility and ease of availability favours its use as a green compound in product improvement and the welfare of the ecosystem as it is non-toxic.

The distinctiveness of PVOH as a synthetic polymer as a biodegradable polymer is regarded as a unique trait among the other synthetic polymers. As a matter of fact, PVOH has been reported to be the only vinyl polymer that can be consumed by some microorganisms to be utilized as their source of energy (Kim, Sohn, Lim, Lee, & Park, 2003; Sakai, Hamada, & Watanabe, 1986; Suzuki, Ichihara, Yamada, & Tonomura, 1973). Sizable amounts of post utilized neat PVOH were released into the environment because of the soaring mass use and production and it is very tough to remove PVOH from effluents and landfills. PVOH degradation through biological means can solve these issues through utilizing specific microorganisms that can degrade PVOH (Fukae et al., 1994; Hashimoto & Fujita, 1985; Lee & Kim, 2003). Thus, the search for microorganisms that are able to feed on PVOH by utilizing it as a carbon source is paramount. In this

study, the biodegradation of PVOH nanocomposites were studied through soil burial method and its responsible degrading bacteria was identified.

1.2 Problem statement

Owing to the abundance and underutilised agriculture biomass such as rice straw, these rice straw had gained significant interest as it can be included as a value-added product. The direct utilization of rice straw or microcrystalline cellulose extracted from rice straw (MCC) as filler materials in polymers, however, does not provide the best reinforcement ability due as the lower surface area and severe agglomeration issues (Ng, Sin, Bee, Tee, & Rahmat, 2017). This led towards the discovery of nanosized crystalline cellulose, namely cellulose nanocrystals (CNC) extraction from MCC as reinforcement filler in biodegradable polymers which can provide a larger specific surface area with more active surface groups to react (Dufresne, 2019; Lokanathan et al., 2017).

Polyvinyl alcohol (PVOH) is one of the most popular biodegradable synthetic polymer. Nevertheless, high cost of production and weak biodegradation of PVOH had become the limiting factor for widespread usage in comparison with other synthetic polymer materials especially in the cost-challenging consumer market (Choo, Ching, Chuah, Julai, & Liou, 2016a; A. Das, Uppaluri, & Das, 2019). In response towards this concern, PVOH can be economically feasible when cheap and renewable natural polymers such as cellulose obtained from agricultural waste were used as reinforcement fillers. On the other hand, hydrophilicity of the resultant polymer nanocomposites is another drawback which reduces its viability for extensive daily applications.

Crosslinking of the polymer nanocomposites, preferably from non-toxic and naturally occurring source, is one of the solution to overcome this particular problem.

It is also important to elucidate the biodegradation and its degrading microorganism of non-crosslinked and crosslinked nanocomposites as the findings on this study is not well explored. Although the polymer biodegradation of PVOH has been reported, the findings on biodegradation of PVOH and its crosslinked state has not been detailed, and experiments reported on the biodegradation of PVOH by specific PVOH degrading microorganisms were scarce and ambiguous due to different experimental conditions. These limitations indicated the opportunity for the search for more microorganisms that are capable of degrading PVOH.

From all of the points described above, an effort has been done throughout this research in evaluating the properties of CNC extracted from rice straw, comparing the physical, tensile, thermal and water barrier properties of non-crosslinked and crosslinked PVOH/CNC rice straw nanocomposite, investigating the biodegradability of non-crosslinked and crosslinked PVOH/CNC rice straw nanocomposite after natural soil burial. In order to detail the cause of biodegradation, responsible degrading bacterias will be isolated and identified through 16S rRNA gene sequencing.

1.3 Research objectives

This research was performed with the following objectives:

1. To evaluate on the properties of CNC extracted from rice straw.
2. To evaluate on the tensile, thermal, chemical and physical properties of non-crosslinked and crosslinked PVOH/CNC rice straw nanocomposite.
3. To investigate the biodegradability of non-crosslinked and crosslinked PVOH/CNC rice straw nanocomposite after natural soil burial.
4. To identify the degrading bacteria for crosslinked PVOH/CNC rice straw nanocomposite.

1.4 Scope of study

In the proposed study, MCC and CNC was extracted from the straw of the paddy plant collected in Arau, Perlis. The properties of MCC and CNC were analysed through FTIR, XRD, morphology and TGA analysis. The CNC weight percentage used to fabricate PVOH/CNC nanocomposites was in the range of 1 wt% to 7 wt%. The grade of PVOH used was fully hydrolyzed (99+ %) and the tacticity of PVOH used was in atactic form. Physical properties of PVOH/CNC nanocomposite were assessed through XRD SoI and SwI MVP. Thermal properties of PVOH/CNC nanocomposite were assessed through TGA analysis. Chemical properties of PVOH/CNC nanocomposite were determined through FTIR analysis. Tensile properties of PVOH/CNC nanocomposite were determined through tensile strength, elongation at break and Young's modulus. Biodegradability of PVOH/CNC nanocomposite were determined through FTIR, surface