CHARACTERISATION OF IN-SITU MODIFIED BACTERIAL CELLULOSE USING 2-ACETYL-1-PYRROLINE

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Bioprocess Engineering)

Faculty of Chemical and Energy Engineering
Universiti Teknologi Malaysia

APRIL 2017

Specially dedicated to my beloved and supporting family:

my parents; Pa’e and Mariam, Abdu Rahim and Che Latifah

husband, Asrul Asmawi

and my sons;

Adam Hakimi, Ammar Husni and Aisy Husaini

ACKNOWLEDGEMENT

First and foremost, I would like to take this opportunity to express my deepest gratitude and appreciation to my supervisor, Prof Dr. Ida Idayu Muhamad for her advice, encouragement and dedicated guidance. Without her advice and consult, this thesis project would not be produced on time.

Furthermore, my heartfelt appreciations are also extended to academic and technical staff of Bioprocess and Polymer Engineering Department for providing technical help, and guidelines throughout the whole research.

To all my friends especially FoBERG members, thank you for always being there, sharing every moment together. To all of them, I want to express my sincere gratitude.

Last but not least, I would like to express my earnest appreciation to my beloved parents, family and all people who involved directly and indirectly for their support, understanding and love throughout this unpredictable and meaningful journey.

ABSTRACT

Bacterial cellulose (BC) is a biopolymer with high purity of cellulose and excellent mechanical properties. Increased interest in the use of natural polymer makes BC as an excellent alternative for plant cellulose. Furthermore, the morphology and properties of BC can be easily altered by incorporation with additives that are not specifically required for the growth of the bacteria in fermentation media. This unique property of BC opens a new gate for development of new cellulose composites with desired properties by incorporation of selective suitable materials. In this research, bacterial cellulose-pyrroline (BC-P) composites were developed where BC was modified using in-situ technique. 2-acetyl-1-pyrroline from *Pandanus amaryllifolius* (pandan) was added into the fermentation medium of BC at five different concentrations. The impact on the characteristics of BC-P composites produced and their performance in hexavalent chromium (Cr (VI)) removal from aqueous solution of Cr (VI) were studied. The results showed that the increase of 2-acetyl-1-pyrroline concentration from 0 to 13 mg/L in the medium resulted in the increase of wet weight from 18.41 g to 45.85 g and dry weight from 1.31 g to 4.75 g. The physicochemical properties of BC-P composites produced were studied. For microstructure study using field emission scanning electron microscopy analysis, all composites showed similar interwoven organized fibrils network. Small particles of pyrroline could be seen on the fibrils surface where the particles number increased with the increase of 2-acetyl-1-pyrroline concentration. Mechanical testing revealed that high concentration of 2-acetyl-1-pyrroline gave better tensile strength of BC-P up to 48.20 MPa with crystanillity of 85.27%. Adsorption experiments with aqueous solution of Cr (VI) were carried out where the results showed proportional removal of Cr (VI) from 58.65% to 83.06% with increase of 2-acetyl-1-pyrroline concentration. Several factors were studied to obtain suitable parameter for the adsoption process. In comparison to native BC, BC-P composites had succesfully achieved up to 87% removal of Cr (VI) from aqueous solution of Cr (VI). The most suitable conditions for the adsorption using BC-P composites were at 180 min contact time with adsorbent dosage of 0.25 g, initial Cr (VI) concentration of 75 g/ml and pH 3. Mathematical modelling for adsorption kinetics and isotherm were well correlated with the pseudo second-order (R^2>0.96) and the Freundlich isotherm model (R^2> 0.99), respectively. In conclusion, addition of 2-acetyl-1-pyrroline using in-situ technique was successfully proven to be able to modify the characteristic of BC for producing new BC-P composites. Furthermore, the ability of BC-P composites as a biosorbent were proven by successfully achieved up to 87% efficiency of Cr (VI) ions removal from aqueous solution of Cr (VI).

ABSTRAK

Selulosa bakteria (BC) adalah biopolimer yang mempunyai ketulenan selulosa yang tinggi dan sifat-sifat mekanikal yang baik. Peningkatan minat terhadap penggunaan polimer semulajadi menjadikan BC sebagai alternatif terbaik untuk selulosa tumbuhan. Tambahan pula, morfologi dan sifat-sifat BC dapat diubah dengan mudah melalui penggabungan dengan bahan tambah yang tidak diperlukan untuk pertumbuhan bakteria di dalam media fermentasi. Sifat yang unik ini membuka ruang untuk pembangunan komposit selulosa baru dengan ciri-ciri yang dikehendaki melalui penambahan bahan-bahan terpilih yang sesuai. Dalam kajian ini, komposit bakteria selulosa-pirolina (BC-P) telah dibangunkan di mana BC telah diubah suai menggunakan teknik in-situ. 2-asetil-1-pirolina dari *Pandanus amaryllifolius* (pandan) telah ditambah ke dalam medium fermentasi BC pada lima kepekatan yang berbeza. Kesan penambahan terhadap ciri-ciri komposit BC-P yang dihasilkan dan prestasi dalam penyingkiran kromium heksavalen (Cr (VI)) daripada larutan Cr (VI) telah dikaji. Keputusan menunjukkan peningkatan kepekatan 2-asetil-1-pirolina dari 0 hingga 13 mg/L di dalam medium menyebabkan peningkatan berat basah daripada 18.41 g kepada 45.85 g dan berat kering daripada 1.31 g kepada 4.75 g. Sifat-sifat fizikokimia bagi komposit BC-P yang terhasil turut dikaji. Untuk kajian struktur mikro menggunakan analisis mikroskop elektron imbasan pancaran medan, semua komposit menunjukkan jalinan rangkaian gentian halus yang tersusun. Zarah kecil pirolina dapat dilihat pada permukaan gentian halus di mana bilangan zarah meningkat dengan peningkatan kepekatan 2-asetil-1-pirolina. Ujian mekanikal menunjukkan kepekatan 2-asetil-1-pirolina yang tinggi memberikan kekuatan tegangan yang lebih baik sehingga 48.20 MPa dengan kadar kehabluran sehingga 85.27%. Eksperimen penjerapan dengan larutan Cr (VI) dilakukan di mana keputusan menunjukkan penyingkiran Cr (VI) meningkat daripada 58.65% hingga 83.06% dengan peningkatan kepekatan 2-asetil-1-pirolina. Beberapa faktor dikaji untuk mendapatkan parameter yang sesuai untuk proses penjerapan. Berbanding dengan BC asal, komposit BC-P telah berjaya mencapai sehingga 87% penyingkiran Cr (VI) daripada larutan cecair Cr (VI). Keadaan yang paling sesuai untuk penjerapan menggunakan komposit BC-P adalah pada masa sentuhan 180 minit, dengan dos penjerap 0.25 g, kepekatan awal Cr (VI) 75 g/mL dan pH 3. Pemodelan matematik menunjukkan proses penjerapan adalah mengikut model pseudo kedua (R^2> 0.96) dan model isoterma Freundlich (R^2> 0.99). Kesimpulannya, penambahan 2-asetil-1-pirolina menggunakan teknik in-situ terbukti berjaya dapat mengubah ciri BC untuk menghasilkan komposit BC-P baharu. Tambahan pula, kebolehan komposit BC-P sebagai penjerap bio telah berjaya dibuktikan dengan pencapaian kecekapan sehingga 87% penyingkiran ion Cr (VI) daripada larutan cecair Cr (VI).

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E	-	Removal efficiency
M	-	Molarity
N	-	Newton
µl	-	Microliter
g	-	Gram
°C	-	Degree celcius
h	-	Hour
eV	-	Electron volt
kV	-	Kilo volt
wt	-	Weight
v/v	-	Volume per volume
λ	-	Wavelength
1/n	-	Adsorption intensity
b	-	Affinity of the binding sites in biosorbent/ Langmuir constant (L/mg)
C_e	-	Metal residual concentration in solution
C_o	-	Initial concentration (mg/L).
h	-	$k_2 q_e^2$ (mg $g^{-1}min^{-1}$) when the initial adsorption rate t = 0
k_2	-	rate constant of second order adsorption (g $mg^{-1}min^{-1}$).
K_f	-	Freundlich constant that indicates the adsorption capacity
qe	-	Amount of metal ions adsorbed per unit weight of adsorbent at equilibrium
qt	-	Amount of metal ions adsorbed per unit weight of adsorbent at any time *t*

LIST OF ABBREVIATIONS

2AP	-	2-acetyl-1-pyrroline
BC	-	Bacteria cellulose
BC-P	-	Bacteria cellulose-pyrroline
CFU	-	Colony forming unit
CMC	-	Carboxymethyl cellulose
Cr(VI)	-	Hexavalent chromium
DPC	-	Diphenyl carbazide
EB	-	Elongation at break
FESEM	-	Field Emission Scanning Electron Microscope
FTIR	-	Fourier-Transform Infra Red
GC-MS	-	Gas Chromatography-Mass Spectrophotometer
GC-FID	-	Gas Chromatography-Flame Ionization Detectot
GDH	-	Glucose dehydrogenase
HS-SPME	-	Headspace-Solid Phase Microextraction
TMP	-	2-4-6-trimethylpyridine
Q_0	-	Langmuir constant related to adsorption capacity
TS	-	Tensile strength
XRD	-	X-Ray Powder Diffraction
YM	-	Young’s modulus

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CHAPTER 1

INTRODUCTION

1.1 **Research Background**

Cellulose from bacteria has more advantages than the cellulose found in plants. These advantages provide plenty room for the use of the bacterial cellulose (BC) in various fields. The most significant advantage of the BC compared to plant cellulose is its purity. The BC from fermentation is produced in a pure form without lignin and hemicelluloses (Tyagi and Suresh, 2015). It is highly hydrophilic with high mechanical strength. As BC is 100 percent pure and produced in hydrophilic matrix forms, its extensive fibrils and high mechanical strength are maintained throughout the formation. The BC can be produced using different methods and substrates whereby the properties can be modified based on the application. Soluble or insoluble particles in the media, such as living cells and chemicals can readily be incorporated in the growing matrices during the synthesis (Serafica *et al*., 2002). These properties open the opportunities in many fields mainly in medical field, wastewater treatment, paper and also audio industries.

Addition of other materials that are not needed in the growth of bacteria in the fermentation medium is proven to affect the yield and properties of BC. (Ruka *et al.,* 2013). This modification was done either by using in-situ or ex-situ techniques (Shah *et al.*, 2013). Several researchers have shown the ability to improve the production of BC while others have successfully altered the properties of the cellulose by adding certain substrates or by manipulating the operating conditions. For examples, Park *et al.* (2013) reported that addition of magnetite nanoparticles and polyaniline enhanced the thermal stability of BC while da Silva *et al.* (2016) reported that addition of polyethylene glycol improved hydrophilicity properties of BC.

This unique ability of BC allowing a series of potential applications as a new novel BC composite can be produced with modified properties based on its function. These applications range from high mechanical strength of hydrogel with addition of genipin (Dayal *et al.,* 2016; Nakayama *et al.,* 2004), incorporation of aloe vera as wound dressing (Saibuatong and Phisalaphong, 2010), antimicrobial film with addition of silver nanoparticles (Yang *et al.,* 2012; Wu *et al.,* 2014) to the use of BC as membranes such as cellulose acetate membranes reinforced with BC sheet (Gindl and Keckes, 2004). Development of electromagnetic nanocomposite with incorporation of magnetite nanoparticles and polyaniline was reported by Park *et al.* (2013). Juncu *et al.* (2015) suggested the novel uses of BC for drug delivery with the addition of carboxylmethyl cellulose. Furthermore, Shanshan *et al.* (2012) demonstrated that the production of BC membrane in N-methylmorpholine N-oxide that had better mechanical and barrier properties.

Conservation of the forest, particularly trees, is essential in managing the global warming. However, excessive use of trees for cellulose-based products such as paper, biofuels and construction materials have continuously depleting the forest resources (deforestation) which lead to global warming problem. At the same time, natural polymers such as cellulose have attracted many attentions due to the effect of environmental pollution of the synthetic polymer (Pei *et al.*, 2013). Previously, plant-derived celluloses were widely used. For example, cellulose fibres have been used as eco-composite plastics in agricultural fields (Gonzalez-Sanchez *et al*., 2014), biosorbent for heavy metal removal (O'Connell *et al*., 2008), nanofiller for biofilm (Salehudin *et al*., 2014; Slavutsky and Bertuzzi, 2014), and other newly developed degradable composites (Piccinno *et al*., 2015). However, in recent time, BC is started to be used because of its excellent and promising properties (Ashori *et al.,* 2012). Table 1.1 shows the development of BC and its application starting from dessert or *nata* in the 1990's followed by various other applications throughout the year.

Table 1.1: Development in bacterial cellulose research

Development	**Details**	**Reference**
1950's Research on bacterial strain, fermentation medium and fermentation condition	Synthesis of cellulose by *Acetobacter xylinum*	Hestrin and Schramm. (1954)
	Improved cellulose production using *Acetobacter xylinum* mutant	De Wulf *et al.* (1996)
	Improved production of cellulose with Acetobacter SP.LMG 1518 in submerged culture	Vandamme *et al.* (1998)
	Production of BC from fructose in continuous culture	Naritomi *et al.* (1998)
	Optimization of fermentation condition of *Acetobacter xylinum* in shaking culture	Son *et al*. (2001)
	Increased production of BC in synthetic media under shaking condition	Son *et al*. (2003)
	Production of BC from persimmon vinegar	Kim *et al*. (2006)
1990's Research on characterisation of BC	Mechanical properties of bacterial sheets	Yamanaka *et al.* (1989)
	Characterisation of BC produced by *Acetobacte*r *pasteurianus* strain	Bertocchi *et al.* (1997)

	Characterisation on mechanical properties of BC and chitosan blends	Wu *et al.* (2004)
	Microbial cellulose structure in stationary and agitated culture	Czaja *et al.* (2004)
	Characterisation of water in BC.	Gelin *et al*. (2007)
2000 Research on new application of BC	*Nata de coco* as dessert	Budhiono *et al.* (1999)
	Electronic paper displays made from microbial cellulose	Shah and Brown (2005)
	BC as wound healer	Czaja *et al.* (2006)
	Antimicrobial films from BC	Gao *et al.* (2014)
	BC for skin repair materials	Fu *et al.* (2011)
	BC as carrier for drug delivery system	Amin *et al.* (2012)
2009 Research on modification of BC and its application	Modification of BC using nano aloe vera for wound dressing	Saibuatong and Phisalaphong (2010)
	Hybrid BC nanocrystals and silver nanoparticles	George *et al.* (2014)
	Modification of BC using magnetic field	Fijalkowski *et al.* (2015)
	Modification of BC - alginate composites for scaffold in tissue engineering.	Kirdponpattara *et al.* (2015)
	Surface modification of BC using trimethylsilylation for oil-water separation	Sai *et al.* (2015)
	Modification of BC structure under ultrasonic irradiation	Paximada *et al.* (2016)

Environmentally friendly products are becoming increasingly important. Therefore, the production of cellulose through microbial pathways is advantageous due to the ability of the bacteria to produce pure cellulose, devoid of other contaminating polysaccharides (Jozala *et al.,* 2016). This makes the isolation and purification steps relatively simple and not requires energy or chemical intensive processes. The interests for BC in wastewater were proven by several researchers that successfully produced biosorbent for heavy metal removal from BC and its composites. Chen *et al.* (2009) added the carboxymethyl cellulose (CMC) into the medium in order to produce carboxymethylated BC whereas Wang *et al*. (2015) successfully used coated BC for the removal of copper and lead. Modification of BC with amino group is also capable to remove copper and lead (Lu *et al*., 2014) while Zhu *et al*. (2011) produced spherical BC as adsorbent for chromium, mangan and lead. There was also a report on the use of BC as an adsorbent for dye removal after modification with phthalocyanine (Chen and Huang, 2015).

Pandanus amaryllifolius is a tropical plant which is commonly known as "pandan". Pandan is widely used in south-east Asian cooking as a flavour. The characteristic aroma of pandan is mainly caused by the aromatic compound of 2-acetyl-1-pyrroline (2AP) which gives jasmine rice and basmati rice their typical smell. The 2AP can be formed by Maillard reactions during heating of food such as in baked bread (Hofmann and Schieberle, 1998). Laohakunjit and Kerdchoechuen (2007) reported the pandan leaves as one of the best natural source of 2AP.

The 2AP in pandan had been widely used in food processing industries for flavouring (Yahya *et al.,* 2011) and antioxidant (Jong and Chau, 1998; Nor *et al.,* 2008). However, it was believed that pandan has the ability to remove heavy metals from wastewater. Abdullah and Loo (2006) reported the use of pandan leaves to remove plumbum and copper ions while Ngadi *et al*. (2015) had successfully removed chromium using hybrid chitosan-pandan as adsorbent. The capability of pandan as an adsorbent for heavy metal may be due to the presence of cyclic nitrogen (-N=) and ketone in 2AP structure. This is based on the studies by Birch and Bachofen (1990) and Le Cloirec *et al*. (2003) which reported that these two functional groups are responsible for metal biosorption. Other than that, pandan is also reported as adsorbent for dye in wastewater (Yahya *et al*., 2015) and to remove excess nutrient from nutrient rich water (Han *et al*., 2014).

1.2 Problem Statement and Hypothesis

BC (BC) has been studied widely and innovations have been developed in many fields. However in Malaysia, the usage is limited to food industry only. This may be due to fewer researches on the implementation of BC. The ability of BC to be easily modified during the synthesis process enables the development of BC composite for different usage. Therefore, in this research, 2AP from pandan extract was integrated into BC to produce a BC-P composites for heavy metals removal in wastewater.

The development of biomaterial-based products started to gain attention as it has high potential application in many fields such as medical, bioseparation, tissue engineering and food processing (Liang *et al*., 2007). In wastewater treatment, cellulosic materials from various natural sources especially plants were widely employed as adsorbent (Wei *et al.,* 2015; Hokkanen *et al.,* 2016). The cellulose-based adsorbent is very suitable to be used as it is cheap, abundant and available worldwide (O'Connell *et al.,* 2008). However, the use of trees for the production of paper and construction materials have continuously depleted the forest resources and lead to highly needed of cellulose alternative. In this respect, BC can be said as the best choice to be the alternatives for plant cellulose.

Heavy metals are considered as serious environmental contaminant as it can be very harmful for human being and environment. Different processes had been used for treatment of heavy metal in wastewater such as oxidation and reduction, chemical precipitation, filtration, evaporation, ion exchange and reverse osmosis (Kikuchi and Tanaka, 2012). However, most of them were less efficient and costly (Krishnani *et al.,* 2008). Moreover, the production of toxic or chemical sludge from the accumulation of solid waste makes it non-environmentally friendly (Siet, 2013).

In this context, research on economical approach as alternative for current method is meaningful. In light of the above, biological materials such as BC has emerged as an alternative. In this research, modified BC was prepared by incorporation of 2AP from pandan extract using in-situ technique. The bacterial cellulose-pyrroline (BC-P) composites were used as biosorbent for Cr (VI) removal from aqueous solution of Cr (VI). Using in-situ technique offers simpler and faster way for producing BC composites. Moreover, since BC and pandan are biodegradable, natural and very abundant on earth, the current work provides an alternative for sustainable and cheaper materials for heavy metal removal in wastewater treatment.

1.3 Objectives of the Study

The main objective of this study was to investigate the potential of in-situ modification method during fermentation for producing bacterial cellulose-pyrroline (BC-P) composites and its performance as biosorbent for heavy metal removal from wastewater. This involved several specific objectives which are:

1. To develop and produce BC-P composites by incorporating 2AP from pandan extract into native BC using in-situ modification method.
2. To characterise the properties of BC-P composites produced
3. To investigate the adsorption performance of heavy metal from aqueous solution of Cr (VI) using developed BC-P composites.

1.4 Scope of the Study

The scopes of the research works is comprised of 3 different objectives as mentioned in section 1.3. In order to achieve the objectives of this study, the work will include the following scopes.

1. To extract the 2AP from pandan using maceration technique. Using this method, dried pandan leaves were soaked in hot water as a solvent.
2. To identify and quantify the presence of 2AP in the extracts. Pandan extract was sent to Gas Chromatography-Mass Spectrophotometer (GC-MS) for identification of 2AP while the quantification of 2AP in the extracts was done using Gas Chromatography – Flame ion detector (GC-FID).
3. To produce BC-P composite through in-situ modification with different 2AP concentration using static fermentation. Analysis were done in order to study the effects of different 2AP concentration to the cellulose weight, appearance, sugar content in the medium and bacterial growth in the medium.
4. To characterise the properties of the BC-P composites using FESEM, FTIR, mechanical test and study the effect of different 2AP concentration to BC-P composites properties
5. To determine the optimum conditions for effective removal of Cr (VI) as heavy metal ion in aqueous solution of Cr (VI). One-factor-at-time approach was used to study the effect of 2AP concentration, contact time, initial concentration of adsorbate, pH and adsorbent dosage to Cr (VI) removal.
6. To determine the model of Cr (VI) removal using BC-P composites by adsorption isotherm and kinetics study. Two models were selected for adsorption isotherm which was Langmuir and Freudlich Model while for adsoption kinetics, Pseudo first and second order Model were used ..

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