



**TENSILE AND THERMAL PROPERTIES OF
COCONUT SHELL POWDER FILLED
POLYLACTIC ACID BIOCOMPOSITES**

by

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LIST OF ABBREVIATIONS, SYMBOLS, SPECIALIZED, NOMENCLATURES

3-APE	3-aminopropyltriethoxysilane
AA	Acrylic acid
ASTM	American society testing method
ATR	Attenuated total reflectance
CO ₂	Carbon dioxide
CSP	Coconut shell powder
DP	Degree polymerization
DSC	Differential scanning calorimetry
FTIR	Fourier transform infrared spectroscopy
HDPE	High density polyethylene
LDPE	Low density polyethylene
MA	Maleic acid
PDLA	Poly(D-lactic acid)
PDLLA	Poly(D,L -lactic acid)
PE	Polyethylene
PHA	Polyhydroxyalkanoate
php	Part per hundred polymer
PLA	Polylactic acid
PLLA	Poly (L-lactic acid)

PP	Polypropylene
PS	Polystyrene
SEM	Scanning electron microscopy
T_c	Crystallization temperature
T_g	Glass transition temperature
TGA	Thermogravimetry analysis
T_m	Melting temperature
TPS	Thermoplastic starch
UHMWPE	Ultra high molecular weight polyethylene
X_c	Degree of crystallization
ΔH_f	Heat fusion of polymer biocomposites
ΔH_F^0	Heat fusion of 100% crystalline matrix

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Sifat-Sifat Tensil dan Terma Biokomposit Serbuk Tempurung Kelapa Terisi Polilaktik Asid

ABSTRAK

Di dalam penyelidikan ini kesan kandungan serbuk tempurung kelapa (STK) dan modifikasi kimia terhadap sifat-sifat mekanik, morfologi dan sifat-sifat terma telah dikaji. Biokomposit serbuk tempurung kelapa terisi polilaktik asid telah disediakan menggunakan pencampur Brabender Plastograf EC PLUS pada suhu 180°C dan kelajuan rotor 50 rpm. Keputusan menunjukkan bahawa dengan semakin meningkatnya kandungan STK telah mengurangkan kekuatan tensil dan pemanjangan pada takat putus, tetapi meningkatkan modulus elastisiti biokomposit PLA/STK. Penambahan STK pada 30 bsp telah meningkatkan suhu penghabluran kaca (T_g) dan penghabluran (X_c) biokomposit. Suhu penguraian maksimum (T_{dmax}) biokomposit PLA/STK meningkat dengan peningkatan kandungan STK. Pada kandungan STK yang tinggi, kestabilan terma yang lebih baik telah dicapai. Kajian SEM membuktikan bahawa interaksi antaramuka yang lemah diantara hidropilik STK dan hidropobik PLA. Pelbagai jenis modifikasi-modifikasi kimia seperti asid akrilik (AA), asid maleik (MA) dan 3-aminopropiltriethoxysilana (3-APE) telah dikaji. Biokomposit dirawat dengan AA, MA, 3-APE mempunyai kekuatan tensil dan modulus elastisiti yang lebih tinggi, kecuali pemanjangan pada takat putus yang lebih rendah dibandingkan dengan biokomposit yang tidak dirawat. Modifikasi kimia telah meningkatkan T_g , X_c dan kestabilan terma biokomposit yang dirawat. Analisis FTIR menunjukkan modifikasi kimia STK telah mengurangkan kumpulan hidroksi pada STK. Interaksi antaramuka yang lebih baik diantara STK dan PLA telah dibuktikan dengan kajian SEM.

***Tensile and Thermal Properties of Coconut Shell Powder Filled Polylactic Acid
Biocomposites***

ABSTRACT

In this research the effect of coconut shell powder (CSP) content and chemical modification on mechanical properties, morphology and thermal properties of polylactic acid (PLA)/CSP biocomposites were studied. Coconut shell powder filled polylactic acid biocomposites was prepared using Brabender Plastograph EC PLUS mixer at temperature 180°C and rotor speed 50 rpm. The results show that the increasing of CSP content has decreased the tensile strength and elongation at break but increased the modulus of elasticity of PLA/CSP biocomposites. The addition of CSP at 30 php has increased the glass transition temperature (T_g) and crystallinity (X_c) biocomposites. The decomposition temperature of maximum (T_{dmax}) of PLA/CSP biocomposites was raised with increasing of CSP content. The higher CSP content exhibited better thermal stability. The SEM study was proven that poor interfacial interaction between hydrophilic CSP and hydrophobic PLA. The different types of chemical modification such as acrylic acid (AA), maleic acid (MA) and 3-aminopropyl triethoxysilane (3-APE) were investigated. The treated biocomposites with AA, MA, and 3-APE have higher tensile strength and modulus of elasticity except elongation at break lower compared to untreated biocomposites. The chemical modification enhanced the T_g , X_c and thermal stability of treated biocomposites. The FTIR analysis show the chemical modification of CSP was reduced the hydroxyl group of CSP. The better interfacial interaction between CSP and PLA was proof by SEM study.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Biocomposites are defined as the materials made by combining natural filler and petroleum based or biodegradable polymers. Biocomposites derived from natural fiber and crop or bioderived plastic (biopolymer/bioplastic) is likely to more eco-friendly and such composites are termed as green composites (John & Thomas, 2008). For the past few decades, most of the researches are focusing on green and eco-friendly materials with the term such as “renewable”, “recyclable”, “degradable” and “sustainable”, due to the increasing of the environmental awareness (John & Anandjiwala, 2009; Satyanarayana et al., 2009). Therefore, many researches today was efforts on developing a new class of fully biodegradable “green” composites (biocomposites) by combining natural filler with biodegradable polymer (Averous & Digabel, 2006; John & Thomas, 2008).

Over the last few years, many researches are initial the biocomposites by incorporating natural filler into petroleum based polymer, like polyethylene (PE) (Fronaco & Gozalez, 2005; Pickering et al., 2003; Pradhan et al., 2004), and polypropylene (PP) (Gwon et al., 2010; John & Anandjiwala, 2009; Park et al., 2008).

Biocomposites can be produced via conventional processing method of thermoplastic, those processing involved twin screw extruder (Ashori & Nourbakhsh, 2010; Gwon et al., 2010; Kaboorani, 2010), single screw extruder (Rahman et al., 2010), Brabender Mixer (Danyadi et al., 2010; Morreale et al., 2008), Z-blade mixer (Romisuhani et al., 2010; Salmah et al., 2011a; Salmah et al., 2011b; Salmah et al., 2012), Haake Rheolmixer (Nachtigall et al., 2007; Tserki et al., 2006), sigma blade thermal mixer (Kalam et al., 2010), thermo kinetic mixer (Luz et al., 2008), high speed cascade mixer (Bledzki et al., 2010b) and internal mixer (Renner et al., 2010). Nevertheless, the biocomposites with petroleum based polymer were partially biodegradable. The biodegradation of those biocomposites was involved natural filler, and the polymer matrix will remain as residual, which bring the pollution to environment. Although, the recycling of those polymer was the solution to the environmental problem, but just small number of petroleum based polymer was actually recyclable (Wu, 2009). After a few decades, petroleum based polymer will face a problem of the limitation of the petroleum resource. The processing of petroleum based polymer was contributed to carbon dioxide (CO₂) footprint, which giving an environmental impact. Furthermore, the cost of petroleum based polymer resin was keeping increased with the price of petroleum continually increased during the past few decades (Steinbuechel, 2005).

The shortage of petroleum resources, high cost petroleum, and environment concerns increased the demand of biodegradable biopolymer was increased annually. Biopolymers were made from renewable resources materials and it included agro-polymer (e.g, polysaccharides) produced from biomass by fractionation, and

biopolyester obtained by fermentation from biomass or from genetically modified plants (e.g., PHA) or by synthesis from monomers obtained from biomass (e.g., PLA) (John & Thomas, 2008). Biopolymer from agro-polymer was mainly focused on plasticized starch, also called “thermoplastic starch” (TPS), which produced after disruptions and plasticization of native starch with water and plastizer (e.g., glycerol and sobitol) (John & Thomas, 2008). Unfortunately, TPS was shown poor mechanical properties and strong hydrophilic properties compared to conventional polymer.

PLA is one of the polymers from the renewable resources, which polymerized from lactic acid monomers which fermented from corn, sugar cane, and sugar beet (Yu et al., 2006). Otherwise, PLA was eco-friendly polymer as its production also consumed CO₂ by photosynthesis (Rasal et al., 2010). In addition, the biodegradability of PLA was able to reduce the environment pollution, since it can degrades after a period of time in soil and enzymatic environment (Wu, 2009). PLA was not only a low impact materials to environment, but its great properties including good mechanical properties, thermal stability, and processability for injection moulding, extrusion and film-blowing (Carrosco et al., 2010; Schwark, 2009). According to Carrosco et al., (2010), PLA was shown higher tensile modulus and tensile yield modulus as than conventional polymers, like polystyrene (PS) and polypropylene (PP). There was many advantages of PLA compared to conventional polymer, however it was not widely used because of its high cost, high brittleness and poor moisture barrier properties (Carrosco et al., 2010; Madhavan et al., 2010; Wu, 2009). The moisture penetration and hydrolytic degradation of PLA become concerning issues during manufacturing, shipping, storage,

and end-use of PLA products (Madhavan et al., 2010). Furthermore, the lack of the reactive side chain group caused the PLA was chemically inert, that making the surface and bulk modifications much more difficult (Rasal et al., 2010).

PLA biocomposites were a renewable material, which can fully biodegradable, environmental friendly and priceless. PLA biocomposites can be produced by blending PLA biocomposites with low cost, renewable, and fully biodegradable with natural filler. Natural filler was also from renewable source, which were relatively high strength and stiffness and not cause skin irritations (Oksman et al., 2003). However, the PLA/natural filler composites were tend to have moisture sensitive issue as the hydrophilic properties of the natural filler, and it may weaken the mechanical properties of PLA composites. Currently, many research studies of PLA biocomposites involved cassava and pineapple flour (Kim et al., 2011), coconut fiber (Wu, 2009), jute fiber (Bledzki & Jaskiewicz, 2010), bamboo fiber (Okubo et al., 2009; Kang & Kim, 2011), rice husk (Yussuf et al., 2010), olive stone flour (Perinovic et al., 2010), kenaf fiber (Yussuf et al., 2010), banana fiber (Shih & Huang, 2011) and hemp fiber (Pickering et al., 2011). Most of the PLA biocomposites studied shown, the addition of natural fiber was improved the tensile strength of the PLA biocomposites, however the rather than irregular shape natural filler was decreasing in tensile strength (Yussuf et al., 2010). Many of the researchers reported, the modulus of elasticity of PLA biocomposites was increased with the present of natural filler (Yussuf et al., 2010; Shin & Huang, 2011; Kim et al., 2011). Perinovic et al., (2010) also reported that, PLA biocomposites with lignocellulosic filler (olive stone) was shown better thermal stability compared to neat

PLA.

In recent years, the incorporation of lignocellulosic fiber and powder used as reinforcement or fillers in polymer composites was gained great interest among the researchers due to their cheapness, recyclability, and for strength per weight of materials (John & Thomas, 2008). The lignocellulosic filler exhibit some excellent properties compared to mineral filler (e.g., calcium carbonate, kaolin, mica, and talc), as low cost, renewable, high specific strength to weight ratio, minimal health hazard, low density, less abrasion to machine, certainly biodegradability and environmental friendly (Fronaco & Gozalez, 2005; John & Anandjiwala, 2009; Park et al., 2008; Pickering et al., 2003; Tserki et al., 2005; Xie et al., 2010). Some of the lignocellulosic filler are came from agricultural co-product, such as spruce (Tserki et al., 2005), olive husk (Tserki et al., 2005), barley husk, and coconut shell (Bledzki et al., 2010), rice husk (Kim et al., 2006; Yang et al., 2007a), and coconut shell (Pradhan et al., 2004; Sarki et al., 2011).

However, the lignocellulosic fillers naturally are lack of good interfacial adhesion when incorporating with polymer, and resulting in poor mechanical properties of their composites. The present of strong polarized hydroxyl groups on the surface of lignocellulosic fillers make the difficulty in forming a strong interfacial bonding with a non polar polymer matrix, as the hydrogen bonds tend to prevent the wetting of the filler surfaces (John & Anandjiwala, 2009; Tserki et al., 2005). The lignocellulosic filler showed a poor dispersion in the polymer matrix, due to the inherent of strong filler-filler interaction resulting from intermolecular hydrogen bonding (Pickering et al., 2003).

Alternatively, the hydroxyl groups on the surface of lignocellulosic filler could be reduced by reacting with chemical to reduce its hydrophilicity and improve the interfacial adhesion between the lignocelluloses' filler and polymer matrix. Currently, these was many of method to promote the interfacial adhesion between the lignocellulosic filler and polymer matrix, such as alkaline treatment (Fronaco & Gozalez 2005; Gwon et al., 2010; Park et al., 2008), esterification (Danyadi et al., 2010), silane treatment (Fronaco & Gozalez, 2005; Gwon et al., 2010; Ismail et al., 2002; Nachtigall et al., 2007; Park et al., 2008; Pickering et al., 2003), using compatibilizer (Ashori & Nourbakhsh, 2010; Danyadi et al., 2007; Danyadi et al., 2010; Kim et al., 2006; Liu et al., 2008; Nachtigall et al., 2007; Yang et al., 2007a) and treatment with other chemical, such as stearic acid, acetic anhydride, and propionic anhydride (Danyadi et al., 2010; Tserki et al., 2005). Wu (2009) and Kim et al., (2011) reported, PLA grafted maleic anhydride was used as compatabilizer to enhanced the properties of PLA biocomposites. In addition, silane treatment also can enhance the properties of PLA/wood flour/talc composites (Lee et al., 2008).

Coconut (*Cocos nucrifera*) in Malaysia is the fourth important crop in term of acreage, after oil palm, rubber and paddy. Great number of coconut water, coconut milk, coconut oil, and coconut products are processed and consumed in Malaysia. Coconut shell is the lignocellulosic agricultural waste. Coconut shells are the waste left over by the factory which produces coconut milk or oil. Currently, coconut shell powder (CSP) is widely used in produce active carbon, mosquito repellent coil and as a filler in plastic.