

## Comparison of LDPE/Corn Stalk with Eco Degradant and LDPE/Coconut Oil Coupling Agent: Influence on Mechanical Properties

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### ABSTRACT

*This study investigated the effect of corn stalk (CSP) content with different coupling agents, namely eco-degradant and Coconut Oil Coupling Agent (COCA), on tensile properties, as well as the morphology of Low Density Polyethylene/Corn Stalk (LDPE/CSP) biocomposites. It was found that increment of CSP content decreased both tensile strength. The dispersion and interfacial adhesion between CS filler and thermoplastic emerged as significant factors that affected the tensile properties of biocomposites system. In order to improve interfacial adhesion incorporation of eco-degradant and Coconut Oil Coupling Agent (COCA) into LDPE/CS composites is recommended. The Scanning Electron Microscopy (SEM) analysis displayed improvements to the interfacial adhesion between LDPE as matrix and corn stalk powder (CSP) filler with the presence of eco-degradant and Coconut Oil Coupling Agent (COCA).*

### INTRODUCTION

Biocomposites as a material that is made of biodegradable polymer as the matrix material and natural fiber is used as the reinforcing element. Biocomposites using natural fibers and oil derived polymer matrices such as poly olefins and other thermoplastic or thermosets have now existed and have been available commercially as new engineering materials. In automotive industry, Mercedes Benz has forge ahead against the rest in the industry by using jute reinforced plastic for the interior door panels of its E-class vehicles because of lower cost and lower density, apart from automotive industry they are used in many applications such as trucks, homes, offices and factories [1]. Based on this study, plastic matrix which comes from a group of polyethylene thermoplastics has been used broadly in daily life [2]. Polyethylene (PE), the largest volume plastic used in packaging, is the worst offender and is highly resistant to biodegradation. Although starch has been studied as a filler in plastics for about 40 years, degradable starch-plastic composites with good mechanical properties only came into existence in the mid 1970's [3]. However, the degradation of these composites has been of serious concern to environmentalists because of the slow biodegradation of polyethylene. This has prompted the incorporation of starch to serve as a bio degrading and the use of biodegradation aids such as photo oxidants to accelerate the biodegradation process. Despite that, the molecular weight of PE decreases only after a very long period [4]. Corn Stalk (CS), the subject of the present study, is a waste product of corn. Hence, corn stalk can be acquired for industrial purposes without any additional cost. Currently, waste corn stalk is often used as animal food. In addition, corn stalk is also used in pulp and paper industry. In order to make use of harness waste, we also can use corn stalk. Therefore, this study chooses corn stalk as natural filler in biocomposites and indirectly it will increase the value of corn stalk waste [5]. This is to overcome environmental issues and the use of waste product from farming to save production cost. The compatibility setback could probably occur as polyolefin is non-polar and hydrophobic. On the other hand, as the natural polymer, which is also

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a lignocellulosic material and polar due to the -OH groups in cellulose [6]. Cellulose fibers, which are strongly polarized, are naturally incompatible with hydrophobic polymers due to their hydrophilic nature [7]. This is due to the interfacial adhesion between the natural reinforcing filler and matrix polymers, which occurs to be the main factor in associate with these composites. This result has shown adhesion is very poor as well as the effectiveness of the filler reinforcement in the composites is reduced, moisture uptake and inter-fiber aggregation by hydrogen bonding [8]. A coupling agent is used in the studies in order to reduce the hydrophilicity of the filler. This happens because LDPE is made out of petroleum (oil) while corn stalk is from plant, which makes it water absorbent. Thus to overcome this situation, coupling agent is used to improve its compatibility [9].

## EXPERIMENTAL

### Materials

Light Density Polyethylene (LDPE) grade film extrusion was supplied by Titan Chemicals Corp. Bhd, while CS, which is agricultural waste, was obtained from Kodiang. The CS was cleaned, crushed, and ground into powder. After 24 hours, the corn stalk powder (CSP) was dried at 80°C. By using the Malvern Particle Size Analyzer Instrument, 29.96  $\mu\text{m}$  was determined as the average size of the particle for CSP. Behn Meyer Polymer Sdn Bhd supplied the eco-degradant for this study, which refers to PD 04; a Polyolefins that is based on Controlled Degradation Masterbatch. It has been tested by Hong Kong Productivity Council and confirmed to meet the requirement of food grade polyethylene material which is in accordance with FDA 21 CFR 177.1520. Coconut oil acts a new coupling agent which is used in this project. Without using any high heat or chemicals, virgin coconut oil is taken out directly from fresh coconut meat. The reaction between ethylene diamine to acidic oil was like lauric acid from the virgin coconut oil. Figure 1.0 shows the schematic reaction of COCA.

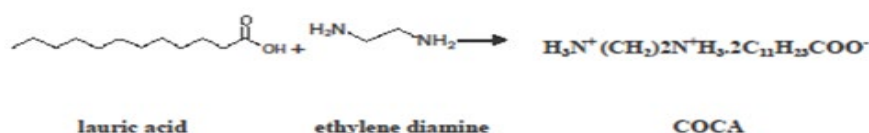


Figure 1. Schematic reaction of COCA.

### Preparation of Biocomposites

Brabender Plastograph mixer Model EC PLUS is used to prepare the LDPE/CS biocomposites at temperature 160°C and rotor speed of 50 rpm LDPE. It takes two minutes for the first LDPE to charge into mixing chamber until it melts entirely. After two minutes added CS powder and continued mixing for another six minutes. The total mixing time was eight minutes. Compression molding machine model GT 7014A is used to compress the biocomposites into tensile bar. According to ASTM D638 tensile bar 1mm thickness type IV has been used. The compression procedure started with 4 minutes for the preheating at 160°C then 1 minute for compressing and later, it took 5 minutes to cool under pressure. The similar procedures were done for preparation LDPE/CS biocomposites with Eco degradant and COCA. At the first, Eco degradant were charged into mixing chamber together with LDPE until completely melt for two minutes and CS was added. For biocomposites treated CS with COCA the addition of filler also at second minutes. The formulation of LDPE/CS biocomposites with COCA and Eco degradant is shown in Table 1.

**Table 1** Formulation of LDPE/CS Biocomposites

Materials	LDPE/CS with Eco Degradant	LDPE/CS with Coconut Oil Coupling Agent (COCA)
LDPE(php)	100	100
CS (php)	0,10,20,30,40	0,10,20,30,40
Eco degradant (php)*	3	3

\*3php from weight LDPE

### Tensile Testing

Instron Machine (Model 5569) was applied to carry out the tensile test by adhering to ASTM D638. The test was conducted with a crosshead speed of 50 mm/min at room temperature. For each composition, five identical samples of tensile properties were measured and the average values for tensile strength, elongation at break, and Young's Modulus had been reported.

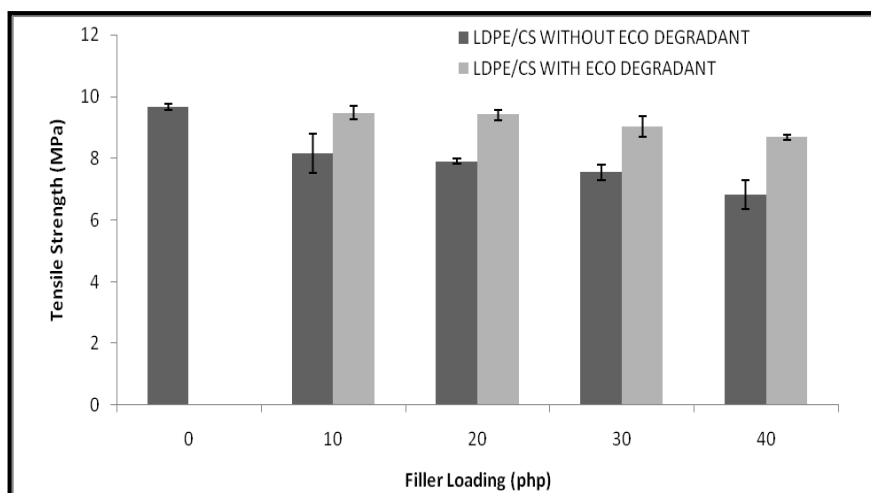
### Morphology Analysis

The instrument applied for morphology study in this experiment was a Scanning Electron Microscope (SEM) (model JEOL JSM-6460LA), in order to observe the dispersion of CSP as filler in the LDPE matrix, as well as the bonding between LDPE as matrix and filler. The fracture ends surface of the specimen was placed on an aluminum stub and the sputter was coated with a thin layer of Palladium (Pd) to avoid electrostatic effect during the examination.

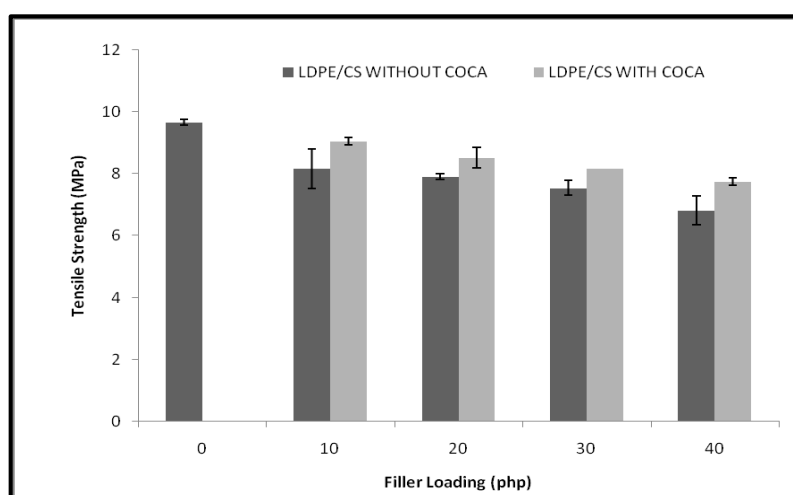
## RESULTS

### Strength

Figure 1(a) illustrates the effect of filler loading on tensile strength of LDPE/CS biocomposites with and without eco-degradant. The results showed that the tensile strength of LDPE/CS biocomposites with and without eco degradant decreased as the CS loading increased. Higher tensile strength was noted for biocomposites with eco degradant than those without eco-degradant. The addition of eco-degradant improved the interfacial interaction between LDPE and CS. The better wettability, dispersion, and orientation of CS in LDPE matrix portray the effectiveness of eco-degradant in enhancing the strength of the biocomposites due to the bonding between matrix and functional group of PE in eco-degradant, which also decreased the hydrophilicity of natural filler. Figure 1(b) displays the tensile strength of with COCA LDPE/CS is higher compared to without COCA LDPE/CS biocomposites. This behavior can be attributed to present of strong interfacial adhesion and better dispersion between filler and polymer matrix with addition COCA. The formation of hydrogen bonding between the COCA and hydroxyl groups of corn stalk have created an interaction of better fillers - matrix. The effectiveness of COCA in increasing the strength of the composites may explain by greater wet ability, dispersion and orientation of the CS and LDPE matrix. This trend is reliable with the previous study, addition of POFA has improved the interaction between bentonite and polypropylene to become more effective thus increasing the tensile strength of the composites [10].



**Figure 1(a).** The effect of filler loading on tensile strength of LDPE/CS biocomposites with and without eco degradant.



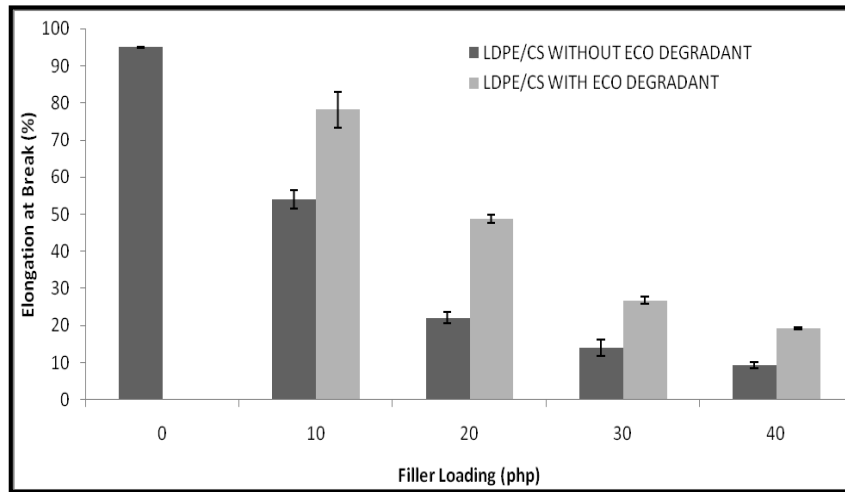
**Figure 1(b).** The effect of filler loading on tensile strength of without COCA and with COCA LDPE/CS biocomposites.

### Elongation at Break

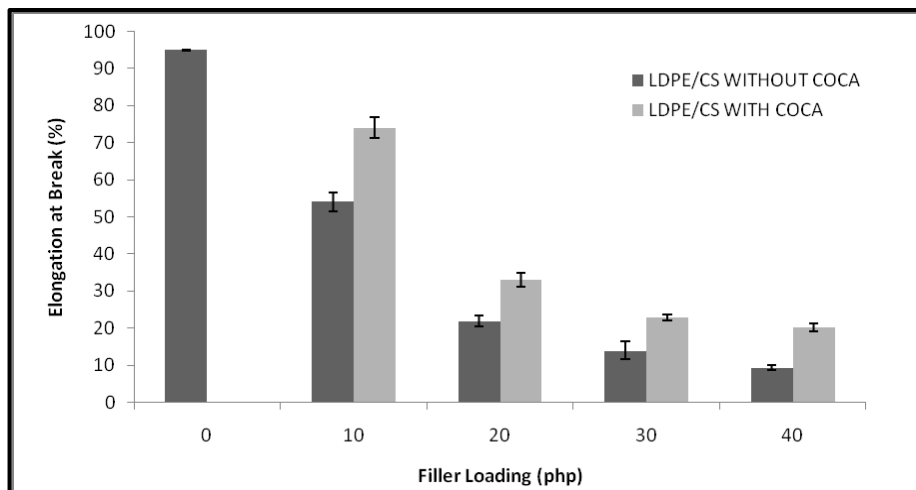
Figure 2(a) presents the effect of filler loading on elongation at break of LDPE/CS biocomposites with and without eco-degradant. The elongation at break has decreased progressively when the filler loading is increased. The decreasing trend on elongation at break could be seen in both biocomposites. The outcomes portrayed in the figure show that elongation at break of biocomposites with eco-degradant was higher than those without eco-degradant. The addition of eco-degradant seemed to increase the ductility of biocomposites. This was clearly visible for biocomposites with eco-degradant because of adhesion between filler and LDPE matrix restricted the deformation capacity of matrix within the elastic zone due to the addition of eco-degradant at the plastic zone.

The increase in elongation at break reveals that eco-degradant had effectively functioned as a degradation additive, while the properties of the eco-degradant promoted the behavior of plasticity in the biocomposites. Figure 2(b) showcases the elongation at break of LDPE/CS biocomposites without COCA and with COCA LDPE/CS decreased in the company of filler loading. The elongation at break of both biocomposites show decreasing trend with CS loading increased, due to the presence of COCA which have good dispersion and interfacial region between filler and

matrix are formed. At similar filler loading, elongations at break of without COCA LDPE/CS biocomposites lower than with COCA biocomposites and presence of COCA shows plasticizer properties of LDPE/CS with COCA biocomposites.



**Figure 2(a).** The effect of filler loading on elongation at break of LDPE/CS biocomposites with and without eco-degradant.

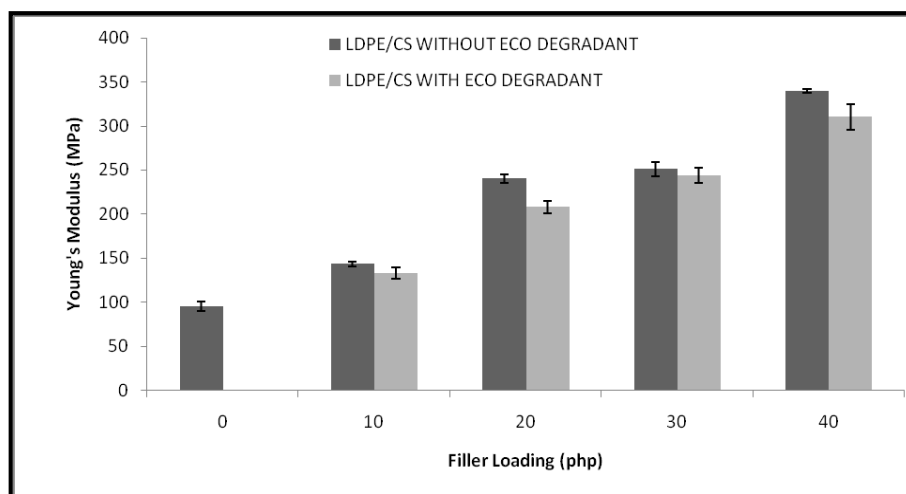


**Figure 2(b).** The effect of filler loading on elongation at break of without COCA and with COCA LDPE/CS biocomposites.

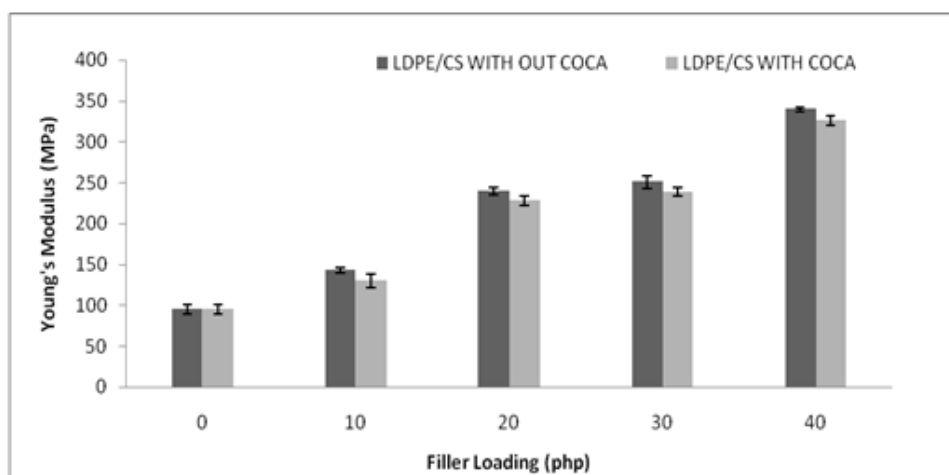
### Young's Modulus

Figure 3(a) illustrates the Young's modulus of LDPE/CS biocomposites with and without eco-degradant. The Young's modulus of biocomposites with and without eco-degradant appeared to increase with increment of CS loading. The increased in Young's modulus with the increasing in CS content is predicted since the addition of filler has increased the stiffness of the composites. The Young's modulus of the biocomposites with eco-degradant was lower than those without eco-degradant. The ductility of LDPE/CS biocomposites with eco-degradant had improved, while the stiffness was reduced due to the presence of eco-degradant. Figure 3(b) shows the Young's modulus of without COCA and with COCA LDPE/CS biocomposites was increasing with the increasing of filler loading. The Young's modulus of the LDPE/CS biocomposites without COCA was higher than those with COCA even though with the similar loading. These results indicate that the efficiency of COCA in improving plasticizers of LDPE/CS composites, while the polymer chain mobility inherent by the better filler-matrix interaction. The application of coupling agent

in polymer composites was used to overcome the dispersion pattern and to enhance the mechanical strength of composites by improving adhesion across the interface [11].



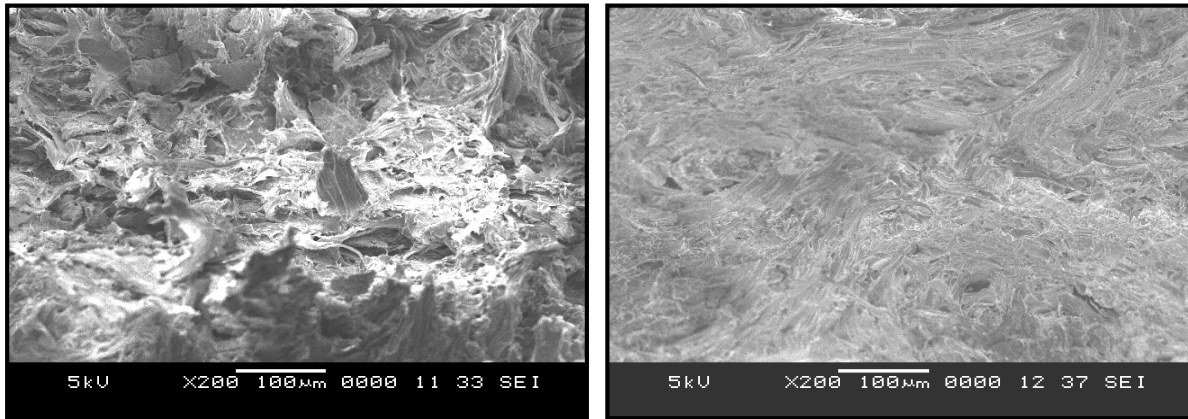
**Figure 3(a).** The effect of filler loading on Young's modulus of LDPE/CS biocomposites with and without eco-degradant.



**Figure 3(b).** The effect of filler loading on Young's modulus of without COCA and with COCA LDPE/CS biocomposites.

### Morphology Study

Figures 4(a) and 4(b) illustrate the micrograph of tensile fractured surface of LDPE/CS biocomposites with eco degradant and COCA LDPE/CS biocomposites at 20 php CS loading, respectively. The surface of the biocomposites became ductile due to the presence of eco-degradant. The CS filler displayed better dispersion and was embedded in the LDPE matrix. This result is in line with the output retrieved from elongation at break for LDPE/CS biocomposites with eco-degradant, which revealed plasticity behavior. The micrograph portrayed in Figure 4 (b) shows that the surface of biocomposites with COCA indicated better interfacial interaction, as well as better adhesion between CS and LDPE matrix. The fiber-matrix adhesion was enhanced due to the chemical connections between fiber and matrix supplied by the COCA. Therefore, these results proved the compatibility between filler and matrix was considerably can be improved when react to the COCA.



**Figure 4(a).** SEM of tensile fracture surface of treated LDPE/CS biocomposites with eco degradant (20 php) at magnification 200X.

**Figure 4(b).** SEM of tensile fracture surface of LDPE/CS biocomposites with COCA (20php) at magnification 200X.

## CONCLUSION

The presence of eco-degradant and COCA enhanced the tensile strength, elongation at break, and crystallinity of biocomposites. The morphology of biocomposites with eco-degradant and COCA showed the plasticity behavior. Nevertheless, the Young's modulus seemed to decrease with the addition of eco-degradant. The morphology of biocomposites with eco-degradant and COCA had revealed plasticity behavior.

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