

# The Influence of Thermoplastic Starch and Banana Fibre Contents on Physical and Thermal Properties of LLDPE

A W M Kahar<sup>1,2\*</sup> and A E Abduati Salem<sup>1</sup>

<sup>1</sup> School of Materials Engineering, Universiti Malaysia Perlis, 02600 Jejawi, Perlis.

<sup>2</sup> Centre of Excellence Geopolymer and Green Technology, Universiti Malaysia Perlis.

E-mail: kaharwahab@unimap.edu.my

**Abstract.** Blending of linear low density polyethylene (LLDPE), thermoplastic starch (TPS) and banana fiber (BF) have been studied. Two types of systems were prepared; the matrix having different ratio of LLDPE/TPS and, the LLDPE/TPS composites having 5 – 30 wt% BF. Morphological changes using scanning electron microscope (SEM) were observed and it showed that TPS particle are homogenously dispersed in LLDPE matrix. On the other hand BF was found to be well embedded in TPS phase, showing the good interaction between BF and TPS phases. This observation show an agreement with the Young's modulus value which is increased with the BF contents. The increment in Young's modulus value was also attributed to the difficulties in LLDPE/TPS chains movement with the presence of BF.

## 1. Introduction

Linear low density polyethylene (LLDPE) has interesting mechanical properties which provide good tensile properties, dimensional stability and high thermal and chemical resistance. Currently there are many intensive study on the biodegradable plastic based on the natural resources due to the many environmental downsides caused by conventional plastics such as LLDPE, starting from the production of plastics until the problem of waste disposal [1-2]. As a response for the environmental issues, blending synthetic polymer with a cheap natural biopolymer from the natural bio-based plant provides new attention to manufacture biodegradable or partial biodegradable products. Most commonly used are cellulose and starch. Incorporation of cellulose and starch into synthetic polymer, which enhances the environmental fragmentation and degradation have been investigated by many researchers [3-5]. This new generation of bio-based plastic can compete in the market which is currently being dominant by the products based on petroleum feedstock.

It is not necessary for producing 100% of biodegradable materials as substitution of petroleum based products immediately, but a partial replacement also could help to save our environment. Economical problem associated with expensive process for producing biodegradable polylactic acid (PLA), polyhydroxyalkanoates (PHAs) polycaprolactone (PCL), polybutylene succinate (PBS) and low mechanical properties of inexpensive thermoplastic starch (TPS) makes the production of fully biodegradable materials difficult. A viable solution is to combine different features and benefits from both petroleum and bio-resources based materials to produce useful products having the requisite combination of cost-performance properties for real world applications. Most of the synthetic polymers are incompatible at molecular level with TPS and leads to poor interfacial adhesion strength. Due to strong intermolecular hydrogen bonding, TPS tend to agglomerate and do not easily disperse in LLDPE matrix. These drawbacks cause the properties of the blends to reduce significantly with



increasing TPS content. In order to overcome incompatibility of these blends, the area of study was concentrated on adding banana fiber that can be compensated the properties deterioration caused by the incorporation of TPS. Therefore, this study proposed a preparation and characterization of linear low density polyethylene LLDPE/thermoplastic starch (TPS) / banana fiber composites for partial biodegradable composites production.

## **2. Materials and methods**

Linear low density polyethylene (LLDPE) granulates with density 0.92 g/cm<sup>3</sup> was supplied by Titan Chemicals, Malaysia. Tapioca starch used was a food grade and obtained from Thye Huat Chan Sdn Bhd., Malaysia. Reagent grade glycerol was obtained from Merck, Darmstadt, Germany and used as received.

### *2.1. Preparation of Thermoplastic Tapioca Starch (TPS)*

Tapioca starch was dried at 80°C in vacuum oven for 24 hours. Tapioca starch was first pre-mixed with 35wt% of glycerol using a kitchen blender (3000 rpm, 2 min) until a homogenous mixture was obtained. Then, the mixture was stored overnight in a dry place. Later the mixture was processed using heated two-roll mill at the temperature of 150°C and the mixing time was 10 min.

### *2.2. Preparations of LLDPE/TPS/Banana Fiber Composites*

The blends were prepared by melt blending LLDPE/TPS/banana fiber in a Haake Rheomix 600 mixer equipped with roller rotors. The mixing was carried out at temperature 150°C and rotor speed was fixed at 55rpm. The LLDPE was first charged into the mixing chamber for 3 min and followed by TPS. Then, the mixing time was continued for 4 minutes before mixed with banana fiber and the process continued until plateau torque was reached. The whole process proceeded for 12 min. The blend was converted into 1mm sheet using a hydraulic hot press Gotech Testing Machine. The hot press procedures involved by preheating at 150°C for 6 min and followed by 2 min compressing at the same temperature, subsequent cooling under pressure for 4 min.

### *2.3 Tensile Test*

Measurements of the tensile properties were performed by using an Instron Universal Testing Machine (model 3366), with crosshead speed 50 mm/min. Dumbbell sample (1mm thick) with 50 mm gauge length were tested according to ASTM D 638. The mean value, out of five samples was reported with standard deviation to show the error range.

### *2.4. Scanning Electron Microscopy (SEM)*

Fracture surface of the samples were performed with a Leo Supra – 3SVP Field Emission Scanning Electron Microscopy at an acceleration rate of 20kV. The fracture surfaces of the specimen were mounted on aluminium stubs and sputter coated with thin layer of gold to avoid electrostatic charge during examination.

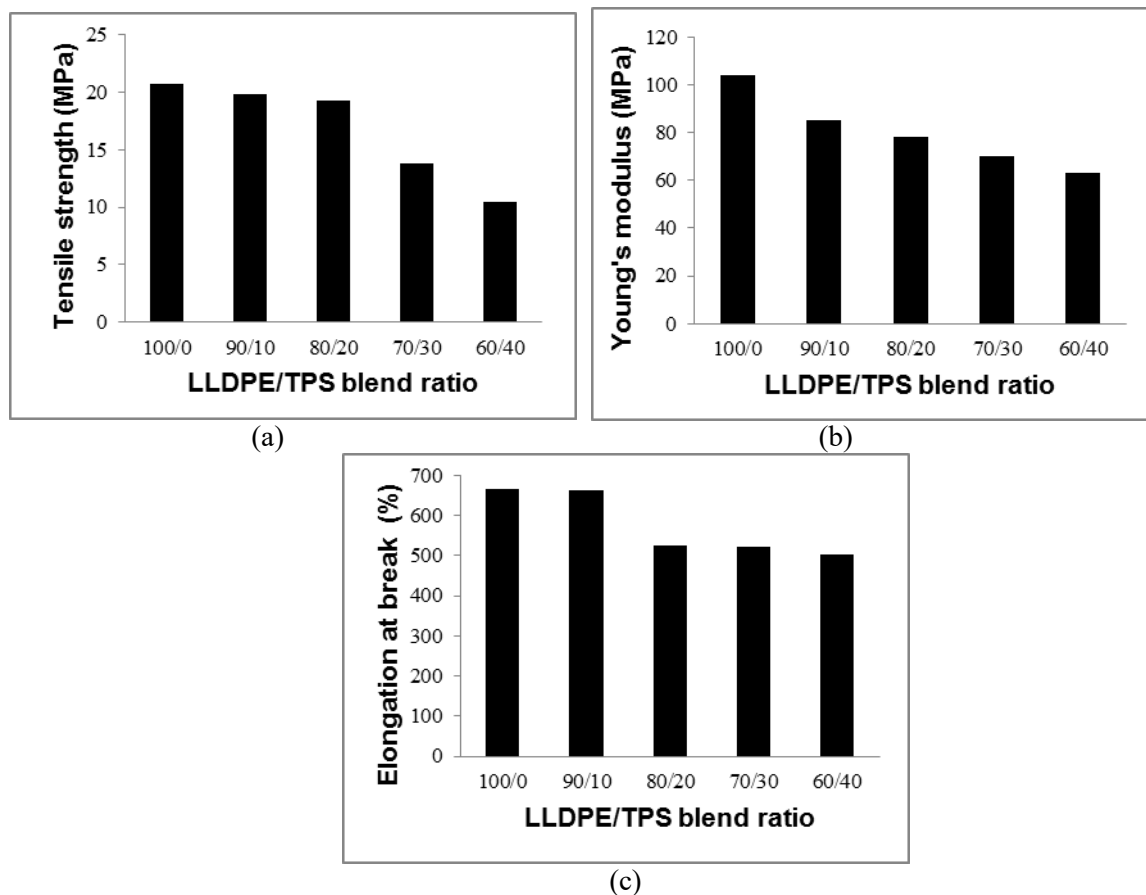
### *2.5. Thermogravimetric Analyses (TGA)*

The thermogravimetric analyses of the blends were operated using a Perkin Elmer Pyris 6 machine. Samples were tested at heating rate 10°C/min from ambient temperature to 600°C. Two types of samples were studied: LLDPE/TPS blends and LLDPE//TPS/banana fiber composites. Some text.

## **3. Results and discussions**

In this TPS based LLDPE blends, LLDPE as a rigid phases poses the excellent tensile properties; whereas, the TPS plays a function as a biodegradation agent of resultant blends. TPS based LLDPE blends offer the combinations between easy processing of thermoplastic with added biodegradable functional of tapioca starch. Besides that, the starch conversion from granule to plasticized form was also considered as an impact modifier to absorb external force applied to the blends and preventing the

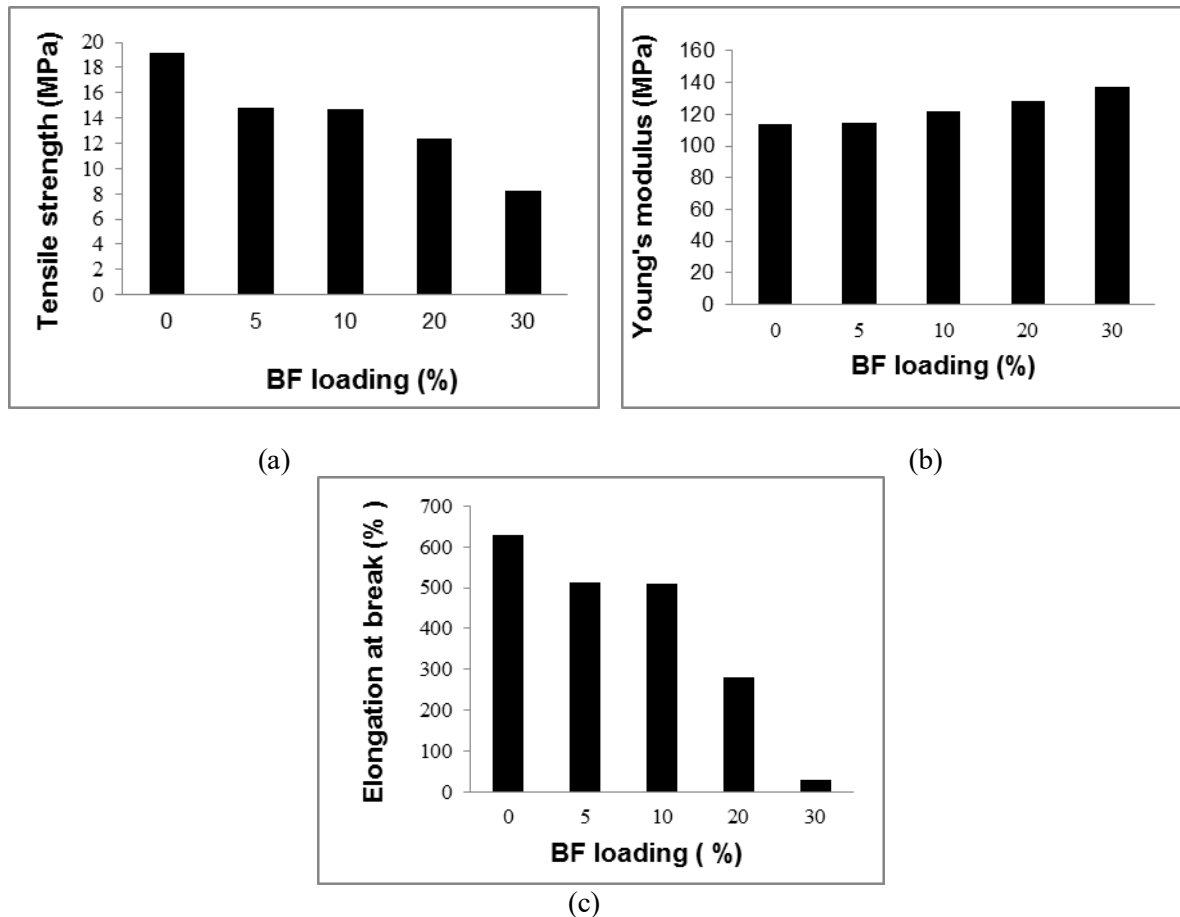
crack propagation. The tensile properties of LLDPE/TPS blends were represented in order to understand the mechanical characteristics of the blends at different LLDPE and TPS ratio (figure 1 (a) – (c)). The blends were varied from 100% of LLDPE to a ratio of LLDPE/TPS 60/40 in order to observe the mechanical behavior of the blends with incorporation of TPS. At low TPS contents, TPS phase existed as a dispersed phase, and thus the deformation behavior of the blends depends on the LLDPE phase. Up to 20% of TPS contents, the tensile strength of the blends almost identical to the virgin LLDPE value. As increasing TPS contents, TPS phase changed from the small size dispersed phase into agglomerate TPS phase, and thus LLDPE plastic response decreased as indicated by decrease in tensile strength and Young's modulus values. Based on these tensile properties results, LLDPE/TPS with a blend ratio of 80/20 was chosen for the further work.



**Figure 1.** (a) – (c) Variation of tensile properties in LLDPE/TPS blends at different blend ratio.

The effect of BF incorporation into tensile strength of LLDPE/TPS (80/20) was shown in figure 2(a). The graph exhibited that the tensile strength decrease sharply at initial addition of BF component and the value remains unchanged up to 10% BF. The possible explanation for this observation was due to the dilution effect as hygroscopic BF component was generally incompatible to the LLDPE phase. Further addition of BF up to 30% showed mark decrease in tensile strength, indicated that the optimum BF loading was achieved. On the other hand, the Young's modulus values were strongly affected by the increment of BF loading in the LLDPE/TPS blends figure 2 (b). The values increased linearly with the BF weight fraction. Its can be explained by the interlocking mechanism between BF surface with LLDPE chains. As expected that the increment in Young's modulus values was proportionally inverse to that elongation at break values figure 2 (c). Insufficient interaction at the BF-LLDPE interphase and high amount of BF contents (30%) might result in early sample failure. It can

be conclude that, the acceptable mechanical strength of LLDPE/TPS-BF composites is only feasible at lower BF content, and from graphs, it can be approximated at around 10-20% of BF portion.

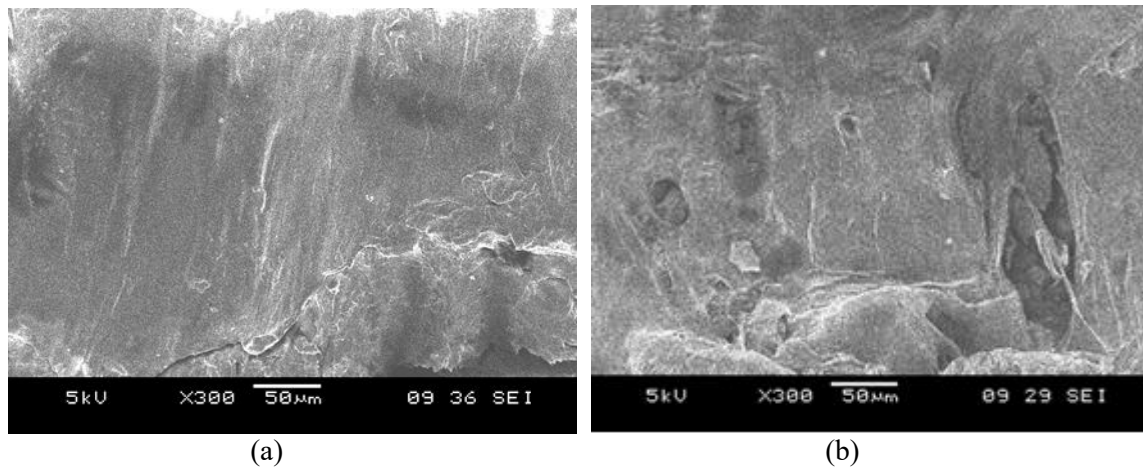


**Figures 2.** (a) – (c) Variation tensile properties in LLDPE/TPS-BF blends at different BF contents.

SEM images of LLDPE/TPS blends containing 10% and 30% of BF were given in figures 3 (a) and (b). The micrographs at 300 X magnification revealed that BF was well embedded in TPS phase. This observation was expected due to the BF and TPS have almost similar characteristics in nature. At the higher BF loading, the fracture surface of the LLDPE/TPS blends become coarser indicating better resistance to the force applied to the blends. This observation can be correlated with Young's modulus value where it increases when BF was introduced to the LLDPE/TPS blends.

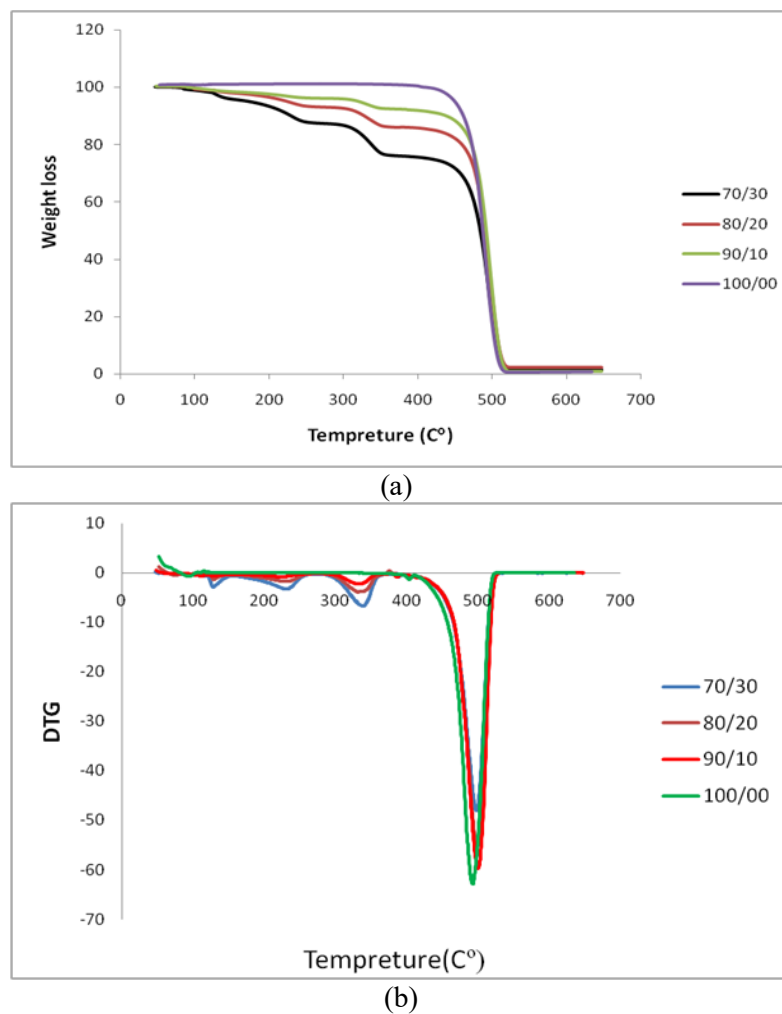
The TGA and DTG experimental results of LLDPE/TPS blends and LLDPE/TPS/banana fiber are shown in figure 4 (a) and figure 4 (b). It can be observed in the TGA curve that the degradation of pure LLDPE starts at temperature of 480°C and for TPS, the degradation start as low as 150°C. This observation indicates the addition of TPS has promoted the degradation of the blends and this might due to volatilization of moisture and glycerol in TPS. Besides that, one significant slope occurred at about 290°C-350°C and it can be attributed to the pyrolytic process in starch chains.

The similar result was reported by Kahar et al. [6] for pyrolytic temperature of starch polymer. For the DTG thermograms of LLDPE/TPS/banana fiber, it's showed that the decomposition point for both 10% and 30% banana loading shifting to higher decomposition temperature. With increasing the amount of banana, the first degradation peak of the TPS shifted from 324°C to 330°C and 335°C with respect to 10% and 30% TPS loading.

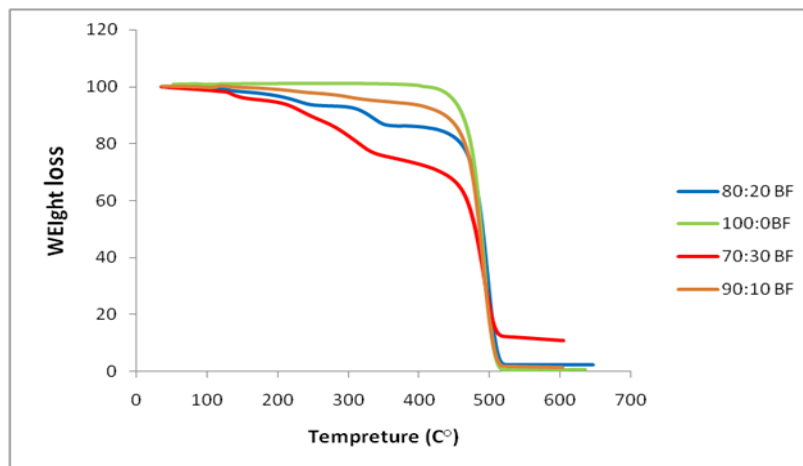


**Figure 3.** (a) – (b) SEM micrograph of LLDPE/TPS at 10% and 30% BF loadings.

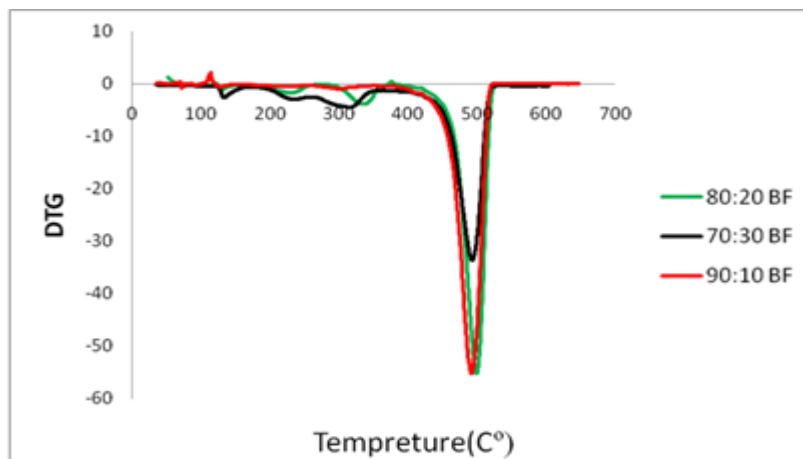
Another peak of the blends also show a similar trend, which is 10 – 15°C higher with the increased in banana fiber loading, showing the significant increase in thermal stability.



**Figure 4.** (a) and (b) Thermogravimetric curves and DTG of LLDPE/TPS blends.



(a)



(b)

**Figures 5.** (a) and (b). Thermogravimetric curves and DTG of LLDPE/TPS/banana fiber.

#### 4. Conclusions

The purpose of this study was aimed to develop partial biodegradable plastics by utilizing TPS and BF. The optimum contents of TPS and BF in LLDPE matrix, and with acceptable mechanical properties were assessed in terms of mechanical properties such as tensile strength and Young's modulus. The mechanical data was also correlated with the blends morphologies in order to understand the interaction behaviour between LLDPE, TPS and BF. In general, the tensile strength was decreased as increased in TPS and BF contents, but the Young's modulus values showed good increment as BF contents was increased. SEM micrographs showed that BF has good adhesion with TPS and also well embedded in TPS phase. The good interaction between BF and TPS resulted in better failure resistance when load was applied to the blends. Banana fiber was introduced into LLDPE/TPS matrix as a reinforcing agent in order to promote better thermal stability of the LLDPE/TPS blends. In the presence of the banana fiber, LLDPE chains are difficult to form crystal due to the interfering chains alignment from the neighboring components. TGA and DTG thermograms displayed the effect of banana fiber in LLDPE/TPS blends. The shift of thermal stability for compatibilized blends towards higher temperature also can be explained by the increase in interfacial interaction between banana fiber and TPS. In conclusion, the composites showed better

thermal properties than the LLDPE/TPS blend, as a result from the stronger interfacial interaction between banana fiber and TPS and the influence of banana fiber on the LLDPE chains alignment.

### References

- [1] Saeoui P, Sirisinha C, Sanguanthammarong P and Thaptong P 2010 *Polymer Testing*. **29** 346-351
- [2] Kahar A W M, Ismail H and Nadras O 2012 *Polymer-Plastics Technology and Engineering* **51** 298–303
- [3] Ning W, Jiugao Y, Xiaofei M and Ying 2007 *Carbohydrate Polymer*. **67** 446–453
- [4] George R N and Carol D S 2008 *Polymer*. **49** 1-173
- [5] Feng S Y, Lauke B and Mai Y W 2008 *Composites Part B: Engineering*. **39** 933-961
- [6] Kahar A W M, Ismail H and Nadras O 2012 *Journal of Applied Polymer Science* **128**(4) 2479–2488

### Acknowledgments

The authors would like to thank Universiti Malaysia Perlis for the supply of raw materials and the equipment provided. The authors also gratefully acknowledge the financial support from the FRGS grant 9003-00601.