MECHANICAL AND PHYSICAL PROPERTIES OF OIL PALM DERIVED CELLULOSE-LDPE BIOCOMPOSITES AS PACKAGING MATERIAL

Chee, P. E.[1], Talib, R. A.[1*], Yusof, Y. A.[1], Chin, N. L.[1],Ratnam, C. T.[2], Khalid, M.[3], and Chuah, T. G.[4]
[1]Department of Process and Food Engineering, Faculty of Engineering, Universiti Putra Malaysia (UPM), 43400 Serdang, Selangor DE, Malaysia.
[2]Radiation Processing Technology Division, Malaysian Nuclear Agency, Bangi, 43000 Kajang, Malaysia.
[3]Department of Biotechnology Engineering, Faculty of Engineering, International Islamic University, Gombak 53100, Kuala Lumpur, Malaysia.
[4]Institute of Tropical Forestry & Forest Products (INTROP), Universiti Putra Malaysia, 43400 Serdang, Selangor DE, Malaysia.
Email: rosnita@eng.upm.edu.my

ABSTRACT

This study focused on processing of various oil palm derived cellulose (OPDC) loading ranging from 0 to 30 wt % into low density polyethylene (LDPE) biocomposites (100 % LDPE polymer act as control) and the material was characterised for mechanical and physical properties. Prior to compounding, biomass oil palm empty fruit bunch fiber (OPEFBF) was derived to cellulose via chlorination and mercerization processes. The cellulose was compounded with LDPE using Brabender twin screw compounder. The processability or mixing characteristics of the biocomposites and pure LDPE polymer during compounding were monitored from their mixing torque curves. The result showed that higher torque value was required to sustain the stable flow of mixture as the cellulose loading was increased. It is also observed that incorporation of 30 % cellulose loading has slightly improved the tensile strength. Whereas, the elongation at break and impact strength were found decreased monotonically with the increasing of cellulose loading. Less than 6 % of water absorption was measured by the composites.

Keywords*: empty fruit bunch fiber, cellulose, LDPE, biocomposite, tensile strength, impact strength.*

INTRODUCTION

In the development for better performance of biodegradable composites, many research has been focused on the utilisation of various types of natural fibers such as wood [1], hemp [2], ramie [3], flax [4], oil palm empty fruit bunch [5] and kenaf [6] treated by irradiation technique namely e-beam [7], chemical treatment [8] and coupling agent [9]. These various approaches were sought with aims to reduce the large consumption of the nonbiodegradable polymer materials by industries in their manufacturing as it causes high level of pollution due to a long process of degradation at landfills and the shortage of landfill space, and the rose of petroleum-based polymer prices due to depletion of it resources which requires minimum utilisation of these materials.

The advancement in biocomposite technology has tremendously increases the processing and production of natural fibers composites. In Malaysia, oil palm is one of the most important commercial crops. Currently, there are about 3.6 million hectares of oil palm plantation producing annually over 10 million tonnes of crude palm oil (CPO), making Malaysia to be one of the major producer of palm oil. However, there are many biomass wastes from the whole process of palm oil. These biomass wastes include oil palm trunks (OPT), fronds (OPF), kernel shell, EFBF, pressed fruit fibers (PFF) and palm oil mill effluent (POME) [10]. From all of these wastes, the majority waste is EFBF, which is estimated around 2.8 – 3.0 million tons per year and about 20 % of the fresh fruit weight [11, 12]. This waste has been converted into reusable products such as particleboard, fiberboard, block board, solid fuel pellets, and pulp and papers. However, the drawback of natural fiber is its high water and moisture uptake [13]. Recently, a study has shown that chemically modified cellulose of oil palm biomasspolymer composite had improved the mechanical properties as opposed to the composite incorporated with untreated biomass composites [8].

In this study, oil palm fiber derivative type of cellulose which compounded with food packaging grade LDPE polymer was produced and the biocomposites were characterised for their properties namely mechanical and physical properties. Thus, the purpose of this study was to investigate the potential of the derived cellulose biocomposites for packaging application. For comparative studies, pure LDPE was employed.

ISBN 1823-1039 ©2010 FEIIC

MATERIALS AND METHODS

Materials

Clean oil palm empty fruit bunch fibers (OPEFBF) were obtained from Malaysia Palm Oil Board (MPOB). Homogenous low density polyethylene (LDPE), Lotrene® FD 0374 was provided by Nuclear Malaysian Agency (MINT). It has a melt flow index of 3.5 g/10min and density of 0.992 g/cm3. Reagent grade sodium hydroxide (NaOH), acetic acid (CH3COOH), and technical grade sodium chlorite (NaClO2) were purchased from Fisher Chemicals Sdn. Bhd. (Malaysia) for cellulose preparation.

Methods

Preparation of cellulose
Preparation of cellulose involved two steps. The first step is the preparation of holocellulose by chlorination method (ASTM D1104). Then, the cellulose was further treated by mercerisation method in order to activate the hydroxyl groups of the cellulose. The detail of the procedure is reported elsewhere [8].

Preparation of biocomposites
Prior to mixing, cellulose fibers were dried for 12 hours in a vacuum oven at 105 °C to remove the moisture. The dried cellulose were cut ground using a Hung Chuan Machinery grinding mill (China) and was passed through 500 m sieve to uniform size. The compounding of LDPE and cellulose was carried out using Brabender Plasticorder PL 2000 at 145 °C for 20 min at roller speed of 50 rpm. The compounding of LDPE only was also done as the control in this study. The composition of composite (wt %) is tabulated in Table 1. Molded sheets with 1 and 2 mm thickness respectively, were prepared at 155 °C and 100 kg/cm3 pressure. This process involved 15 min of preheating and 3 min of complete pressing in hot press followed by cooling for 1 min under pressure.

Table 1: Compositions of cellulose-LDPE biocomposite

Biocomposite	LDPE (wt %)	Cellulose (wt %)
0	100	0
5	95	5
10	90	10
20	80	20
30	70	30

Mechanical testing

Tensile test
All tension testing specimens were cut into dumb-bell shape by using Dumb Bell Cutter (Model SDL-l00). The tests were conducted according to ASTM D-1822-L using Instron Universal Testing Machine (Model 4301) with load cell of 1 kN at crosshead speed of 50 mm/min. Each testing was performed until tensile failure occurred. Seven specimens were tested and at least five replicate specimens were presented as the mean of tested specimens.

Impact test
All notched Izod impact test specimens were cut into rectangular shape with thickness of 3.0 mm. The impact strength was conducted according to ASTM D256 (ASTM 2006) using Impact Pendulum Tester (Model Ceast CE UM-636), performed at 7.5 Joules. Seven specimens were tested and at least five replicate specimens were presented as the mean of tested specimens.

Physical testing

Water absorption test
All water absorption testing specimens were cut into square shaped with dimention of 10 x 10 x 1 mm. Each specimen was oven-dried until a constant weight obtained. The specimens were immersed in distilled water at a room temperature. Next, the specimens were periodically taken out of water, surface dried with absorbent paper

ISBN 1823-1039 ©2010 FEIIC

and reweighed. Immediately, after the measurements were taken, they were placed back into the water. Seven specimens were tested and at least five replicate specimens were presented as the mean of tested specimens.

RESULTS AND DISCUSSIONS

Mixing torque

Figure 1 shows the mixing torque curves obtained from pure LDPE and EFBF derived cellulose-LDPE composites prepared at various loading of cellulose. The curve for pure LDPE demonstrated that highest peak was detected during the first min of the processing time followed by a rapid decrease within the next 2 min before the curve start to decreasing gradually until the mixing torque reaches a constant value which is known as a stabilization zone. For processing the 10 and 20 % of cellulose biocomposites respectively, it is clearly shown that after the torque values were dropped from their maximum peak the curves shown to rise up to their second maximum peak with some fluctuations. Then, the mixing torque gradually reduces to reach their stabilisation zones. The curve for 30 % of cellulose biocomposite demonstrates almost similar trend to the 10 and 20 % cellulose biocomposite, except for a huge drop in torque value before rise up to its maximum peak.

In order to understand this behaviour, the compounding process of 100% LDPE polymer is explained. Initially, the LDPE pellets were loaded into high temperature zone of compounder, this was shown by a rapid increase of the mixing torque to the maximum peak. All of the pellets absorbed the heat inside the compounder and rapidly melted as indicated by a rapid decrease in the torque value before it gradually reduces to a constant value due to the completing of the LDPE melting process. In the case of processing the biocomposites, the second peak was appeared due to the loading of the cellulose fiber into the compounder. This peak indicates that higher torque was recorded to mixing the cellulose with the molten polymer. Once the cellulose has dispersed well in the LDPE, the torque decreased gradually until reached the constant torque value until the end of the processing time. This reflected that the cellulose and matrix LDPE has been completely mixed [14]. Noticed that, a higher mixing torque value at the second maximum peak was observed for the higher % of cellulose loading which indicated that greater amount of energy was required to process the composite.

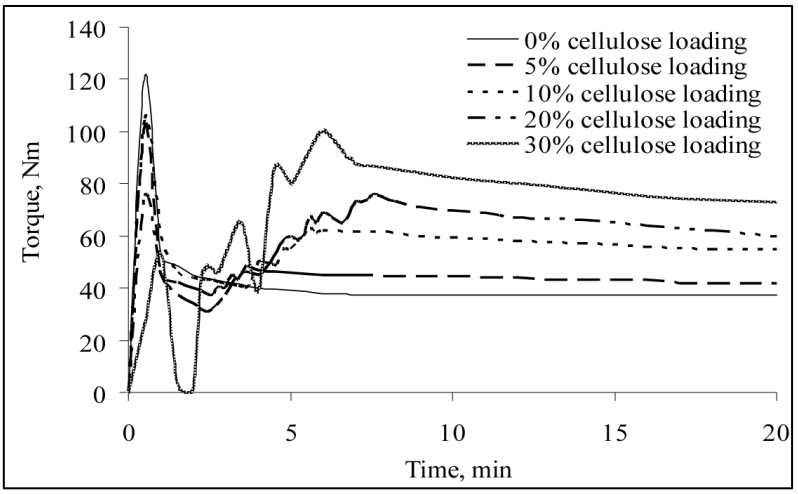

Figure 1: Mixing torque curves of biocomposites and LDPE polymer.

Tensile strength

Figure 2 illustrates the effect of cellulose loading on the tensile strength of the biocomposites. Incorporation 5 % to 10 % of cellulose loading in the biocomposite appeared to show lower tensile strength as compared to pure LDPE. Nevertheless, at 20 % of cellulose loading the tensile strength was equalled to pure LDPE then further increased at 30 % cellulose loading. Incorporation of the natural fibers in the polymer matrix caused an interruption in stress transferring along the applied force. Irregular shape of the ground cellulose fibers were not able to support the stress transferred from the matrix, thus weakened the biocomposite properties [8]. The slight increased shown by 30 % cellulose loading biocomposite could be due to a good interaction of fibers to the matrix. Removal of hemicelluloses during mercerization reduces the cellulose fiber diameter and increases the aspect ratio. In addition, this treatment produces fibers that is less dense and rigid but rougher surface [15], thus allows the fibers to rearranged themselves along the direction of tensile deformation. This was an agreement

ISBN 1823-1039 ©2010 FEIIC

with the previous several studies which have shown that the natural fibres treated with chemical treatment had resulted a significant improvement in tensile strength [8, 16].

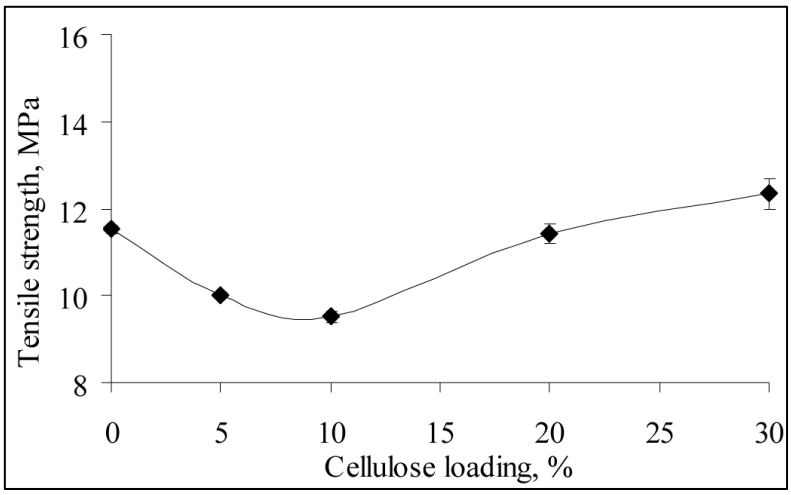

Figure 2: Tensile strength of biocomposites and LDPE polymer

Elongation at break

Figure 3 illustrates the elongation at break for the biocomposites produced at different cellulose loading as compared to pure LDPE. It shows that the elongation at break decreased monotonically as the cellulose loading increases from 0 to 30 %. This is a common observation in biocomposites materials where a similar trend is observed elsewhere [17-19]. The 100 % LDPE matrix has an ability to elongate more due to its elasticity. As the OPEFB derived cellulose loading increased, the elasticity of HDPE has been suppressed by the presence of the derived cellulose. The reduction may be attributed to the decreased deformability of a rigid interface between the fibres and the matrix material [20]. At higher fibre loading, the domination of fibre-matrix interaction can be expected to diminish and being replaced by filler-filler interaction.

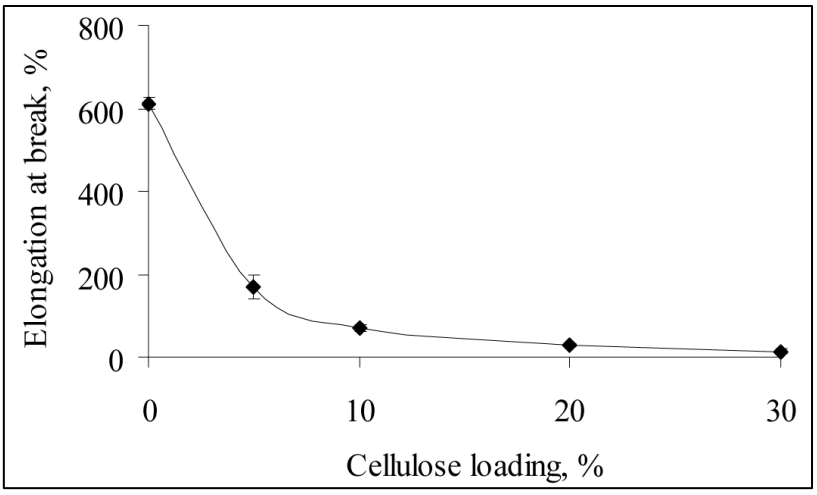

Figure 3: Elongation at break of biocomposites and LDPE polymer

Impact strength

The effect of various cellulose loadings on the impact strength for notched samples is shown in Figure 4. It can be clearly seen that the impact strength decreases monotonically with the increasing of cellulose loading. High impact strength demonstrated by 100 % LDPE is due to the flexible, plasticity and less brittleness of LDPE, which allows it to absorb and distribute the impact energy efficiently [13]. However, increasing the cellulose loading decreases the impact strength. The stiff cellulose fibres will act as stress concentrators in the polymer

ISBN 1823-1039 ©2010 FEIIC

matrix thus reduced the crack initiation energy and consequently the impact strength of the composites [21]. The presence of fibres has also reduced the energy absorbed by the composites during fracture propagation [19].

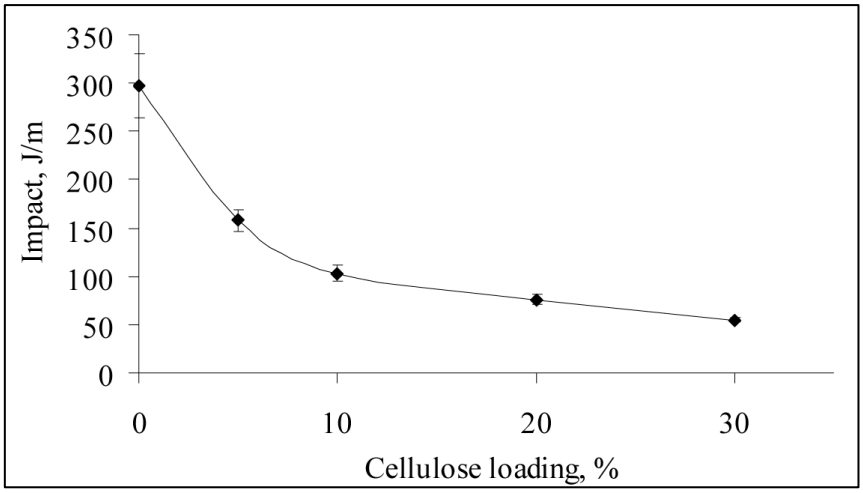

Figure 4: Impact strength of biocomposites and LDPE polymer

Water absorption

Figure 5 shows the water absorption of biocomposites and LDPE polymer. For 100% LDPE polymer, it is clearly shown that less than 0.2 % of water absorption was measured. This is not surprising since the LDPE was compounded prior to pressing into sheet. During this process, it is possible that voids may be trapped within the polymer matrix. Consequently, the water would penetrates through the voids upon immersion in water. This figure also shows that higher water absorption was observed for the higher cellulose fiber loading. Indeed, a sharp increase in their water absorption curves prior to reaching a constant value was also observed as compared to the lower cellulose loading of biocomposites. This type of curve is typical for higher loading of natural fiberpolymer materials [22]. At the beginning of soaking time, the extent of water absorption will take place depending on the amount of cellulose to interact with the water. This process occurs in a short period (rate of water absorption is high) until all cellulose have been interacted with water. At this stage, the biocomposites become saturated and the water absorption has levelled off. Interestingly, the highest loading of cellulose in biocomposite absorbed less than 6 % of water upon immersion in water. It is believed that the mercerization treatment of the derived cellulose from EFBF has reduced the hydrophilicity of cellulose, thus interact less with water [16]. Hence, the water absorption of the biocomposites decreased in their water absorption study on biocomposites.

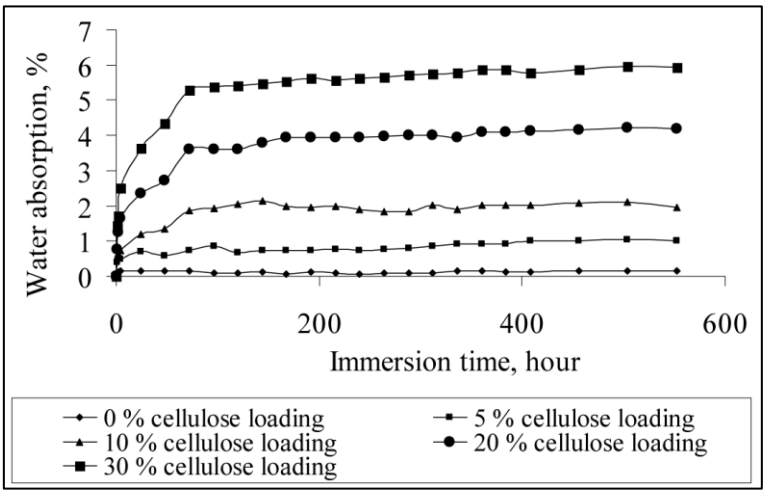

Figure 5: Water absorption of biocomposites and LDPE polymer.

ISBN 1823-1039 ©2010 FEIIC

CONCLUSIONS

This study suggests that incorporating the cellulose that derived from OPEFB has a great potential to reinforce the LDPE polymer and to reduce the amount of polymer in order to obtain a similar tensile strength. The mechanical and physical properties may be further improved by incorporating suitable coupling agent such as maleic anhydride grafted polyethylene (MAPE) to enhance the interfacial adhesion between the cellulose and LDPE matrix.

ACKNOWLEDGEMENT

The authors wish to thank UPM for providing the financial support and also to the staff of MINT for their assistance to carry out this work.

REFERENCES

[1] Gupta, B.S., Reiniati, I., Laborie, M.P.G. and Barun, S. (2007) Surface properties and adhesion of wood fiber reinforced thermoplastic composites. *Colloids and Surfaces A,* 302: 388–395.

[2] Dhakal, H.N., Zhang, Z.Y. and Richardson, M.O.W. (2007) Effect of water absorption on the mechanical properties of hemp fiber reinforced unsaturated polyester composites. *Composites Science and Technology,* 67: 1674-1683.

[3] Goda, K., Sreekala, M.S., Gomes, A., Kaji, T. and Ohgi, J. (2006) Improvement of plant based natural fibers for toughening green composites-effect of load application during mercerization of ramie fibers. *Composites Part A: Applied Science and Manufacturing,* 37(12): 2213-2220.

[4] Oksman K., Skrifvars M., Selin J.F., (2003). Natural fibres as reinforcement in polylactic acid (PLA) composites. *Composite Science Technology,* 63:1324-0237.

[5] Rozman, H.D., Ahmadhilmi, K.R. and Abubakar, A. (2004) Polyurethane (PU) - oil palm empty fruit bunch (EFB) composites: the effect of EFB reinforcement in mat form and isocyanate treatment on the mechanical properties. *Polymer Testing,* 23: 559–565.

[6] Feng, D., Caulfield, D.F. and Sanadi, A.R. (2001) Effect of compatibilizer on the structure-property relationships of kenaf-fiber/polypropylene composites. *Polymer Composite,* 22(4):506-517.

[7] Chantara, T.V., Gunasunderi, R., Wan, W.Y. (2007) Oil palm empty fruit bunch (OPEFB) fiber reinforced PVC/ENR blend-electron beam irradiation. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms,* 265:510-514.

[8] Khalid, M., Ratnam C.T., Chuah, T.G., Ali S. and Choong, S.Y. (2006) Comparative study of polypropylene composites reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose. *Materials & Design,* 29(1):173-178.

[9] Abdelmouleh, M., Boufi, S., Belgacem, M.N. and Dufresne, A. (2007) Short natural-fibre reinforced polyethylene and natural rubber composites: effect of silane coupling agents and fibres loading. *Composites Science and Technology,* 67(7-8):1627-1639.

[10] Ramli R., Shaler S. and Jamaludin M.A. (2002) Properties of medium density fiberboard from oil palm empty fruit bunch fiber. *Journal of Oil Palm Research*, 14: 34-40.

[11] Rozman H.D., Tay G.S. and Abubakar A., Kumar R.N. (2001) Tensile properties of oil palm empty fruit bunch-polyurethane composites. *European Polymer Journa,l* 32: 1759-1765.

[12] Suhaimi M. and Ong H.K. (2001) Compositing empty fruit brunches of oil palm. *Malaysian Agricultural Research and Development Institute (MARDI).*

[13] Yang, H.S., Kim, H.J., Park, H.J., Lee, B.J., Hwang, T.S. (2006) Water absorption and mechanical properties of lignocellulosic filler-polyefin biocomposites. *Composite Structure,* 72: 429-437.

[14] Joseph P. V., Joseph K., Thomas S. (1999) Effect of processing variables on the mechanical properties of sisal-fiber reinforced polypropylene composites. *Composites Science and Technology,* 59: 1625-1640.

[15] Bledzki, A. K. and Gassan, J. (1999) Composite reinforced with cellulose based fibers. *Progress in Polymer Science,* 24: 221-274.

[16] Sreekala M. S., Kumaran M. G., Joseph S. and Jacob M. (2000) Oil palm fiber reinforced phenol formaldehyde composite: influence of fiber surface modifications on the mechanical performance. *Journal of Applied Polymer Science,* 7: 295-329.

[17] Yam, K.L., Gogoi, B.K., Lai, C.C. and Selke, S.E. (1990) Composites from compounding wood fibers with recycled high density polyethylene. *Polymer Engineering and Science,* 30(11): 693-699.

[18] Rozman, H.D., Ismail, H, Jaffri, R.M., Aminullah, A. and Mohd. Ishak, Z.A. (1998) Mechanical properties of polyethylene-oil palm empty fruit bunch composites. *Polymer-Plastic Technology and Engineering,* 37(4): 495-507.

[19] Rozman, D., Mohd Ishak, Z.A. and Ishaiku, U.S. (2005) *Oil palm fiber-thermoplastic composites*. In: Mohanty, A.K., Misra, M. and Drzal, L.T., edi., Natural Fibers, Biopolymers and Biocomposites, CRC Press LLC, USA.

[20] Rozman, H.D., Tan, K.W., Kumar, R.N., Abubakar, A., Mohd. Ishak, Z.A. and Ismail, H. (2000) The effect of lignin as a compatibilizer on the physical properties of coconut fibet-polypropylene composites. *European Polymer Journal,* 36: 1483-1494.

[21] Bengtsson, M., Baillif, M.L. and Oksman, K. (2007) Extrusion and mechanical properties of highly filled cellulose fibre–polypropylene composites. *Composites Part A,* 38: 1922–1931.

[22] Khairiah, B. and Khairul, A. M. A. (2006) Biocomposites from oil palm resources. *Journal of Oil Palm Research*, 103-113.

ISBN 1823-1039 ©2010 FEIIC