



Physico-mechanical Properties of PF Composite Board from EFB Fibres Using Liquefaction Technique

Lee-Lee Chai, Sarani Zakaria*, Chin-Hua Chia, Sharifah Nabihah, and Rozaidi Rasid

School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

Received 26 May 2009; accepted 4 November 2009

ABSTRACT

Phenol formaldehyde (PF) resin was successfully produced from empty fruit bunch (EFB) fibres via liquefaction approach using sulphuric acid as catalyst. The produced PF resin was used to blend with EFB fibres as filler to produce PF-EFB composite boards. The effect of different percentages of EFB fibres on the density, water absorption, impact strength, flexural strength, hardness and thermal properties of the produced PF-EFB composite boards was investigated. The density of the PF-EFB composite boards increased with the increase in EFB fibre content due to the higher density of the EFB fibres relative to the PF resin. The water absorption of the PF-EFB composite boards was also found to be increased with the increase in EFB fibre content. This could be attributed to the hygroscopic nature of the EFB fibres, which had promoted the water absorption process. The mechanical properties (flexural, impact and hardness) of the produced PF-EFB composite boards were increased linearly and reached maximum value with the addition of 20% of EFB fibres. The electron micrographs suggested good compatibility between the PF matrix and the EFB fibres, which had enhanced the mechanical properties of the PF-EFB composite boards. Our results had suggested that the EFB fibres not only could be used to produce PF resin using liquefaction method, but also could be used as reinforcing fillers to enhance the physical and mechanical properties of the composite boards.

Key Words:

liquefaction;
empty fruit bunch;
novolak;
phenol formaldehyde resin;
physico-mechanical properties.

INTRODUCTION

Lignocellulosic wastes mostly originate from deforestation activities, agricultural and wood industries. These industries produce a huge number of lignocellulosic wastes every year, which result in pollution problems without proper managements. Previous studies reported different utilizations of lignocellulosic wastes, including the preparation of novolak PF resins from *Cryptomeria japonica* [1], solvoly-

sis of cellulose using polyethylene glycol and ethylene carbonate [2] and liquefaction of cellulose in phenol using sulphuric acid as a catalyst [3]. Most polymer resins used in industries today are petrochemical based which are non-renewable. As a result of the depletion of crude oil resources, alternative resources should be studied and developed.

In Malaysia, palm oil industries generate a huge amount of wastes,

(*) To whom correspondence to be addressed.
E-mail: sarani@ukm.my

such as empty fruit bunch (EFB) and oil palm mesocarp fibre [4]. A number of studies have been carried out recently to maximize the utilization of palm oil wastes, including conversion of EFB fibre to produce phenolic resins using liquefaction method [5,6].

Liquefaction is one of the promising methods to convert lignocellulosic wastes to phenolic compounds. Our previous study found that the maximum conversion yield (~ 47%) could be achieved by using sulphuric acid as catalyst during the liquefaction of empty fruit bunch fibres [6]. Several relevant studies on the liquefaction of lignocellulosic materials have been reported previously such as the preparation of resole type adhesive from bamboo [7], novolak type phenolic resin from *Cryptomeria japonica* [1] and resole type phenolic resin from bark [8].

Phenolic resins, such as phenol formaldehyde, have been widely used in the manufacturing of paints, insulating and decorative varnishes, adhesive, moulding and casting products [9]. Phenolic resin possesses various advantages over other resins, such as low cost, high rigidity, good dimensional stability, better flame retardancy and chemical reactivity. These can be attributed to its highly cross-linked aromatic structure and greater number of resonance-intensifying hydroxyl groups [9,10].

Natural fibres generated from agricultural wastes, such as palm oil, rice-husks, bagasse and wood chips, are particularly important natural resources in the manufacturing industries. These natural fibres possess low density, low price, great deformability, less abrasiveness to equipments than most inorganic fillers [11]. Natural fibres have been widely used in the preparation of composite boards [12-14]. Cross-linking formed between matrix and fillers provides better reinforcement and excellent final properties [15].

In this study, we have maximized the usage of EFB fibres to produce a totally wood-based composite. Phenol formaldehyde (PF) resin was produced by liquefying EFB fibres via liquefaction technique. It was subsequently mixed with EFB fibres to form fibre-reinforced composite board. We have also investigated the effect of fibre content (EFB fibres) on the physical and mechanical properties of

the composite board. This study is an extension of our previous reported results [16], which we have carried out additional sample characterizations, including density, thermal analysis, infrared analysis and flexural tests.

EXPERIMENTAL

Materials

Empty fruit bunch (EFB) fibres were provided by SzeTech Engineering Sdn. Bhd. Malaysia. The fibre length is ~1-2 mm. The EFB fibres were dried at 105°C for 8 h to remove moisture. Formalin (104003) (37%), methanol (106009), acetone (100014) and analytical grade of phenol (100206) (~99.5%), hexamethylenetetramine (818712) (HMT) and calcium hydroxide (102047) were purchased from Merck, Germany. Magnesium oxide (1304-48-4) and sulphuric acid (7664-93-9) (95-98%) were purchased from System (ChemPur). Zinc stearate (26423) was purchased from Sigma-Aldrich, Germany. All chemicals were used without further purification.

Liquefaction of EFB Fibres

Mixture of EFB fibre and phenol (ratio 1:3) was prepared by mixing 75 g EFB fibres and 225 g phenol in a round bottom flask equipped with a mechanical stirrer, thermometer and reflux condenser. Sulphuric acid (5.0 wt% of phenol) was added into the mixture and followed by heating to 130°C. The mixture was continuously heated and stirred for ~90 min. Afterwards, 75 g formalin was added into the mixture and the temperature was maintained at 100°C for another 60 min. After completion the liquefaction process, methanol was added to dilute the liquefied product and followed by filtration using glass-fibre filter (~3.1 µm) to separate residue. Residue was oven-dried at 105°C for 8 h and weighed for conversion calculation. Magnesium oxide was added into methanol-phenol formaldehyde fraction to neutralize the liquefied product. Methanol and free phenol were then extracted at 80°C and 190°C, respectively, using rotor evaporator. The resulting PF fraction was then dried and grinded into powder for further composite board preparation. The melting point and free phenol of the PF resin is

~180°C and ~39.83 g, respectively [17].

Composite Preparation

Composite board was prepared according to the method we have reported previously with some minor modifications [16]. Briefly, PF resin dissolved in acetone was mixed with HMT (25 wt%), zinc stearate (2.5 wt%), calcium hydroxide (6 wt%) and various amounts of EFB fibre (0, 10, 20, and 30 wt% PF). The mixture was stirred and dried in oven at 70°C for 2 h to remove acetone. After that, the sample was ground into fine powder. The resin powder was then compress moulded in a die with dimension 13 × 10 × 0.3 cm³. The preheating time, ventilation time and curing time are 20 s, 35 s and 350 s, respectively. The press temperature was 185°C and the pressure was 150 kg/cm². The sample was then post cured in oven. The samples were then cut to specific dimensions for further characterizations.

Characterization

The density of PF/EFB composite was measured using a densitometer (MD 300s) under atmospheric condition using water-immersion method. For the water absorption test (ASTM D 570), specimens were dried in an oven at 105°C for 24 h and weighed (W_{dry}). The specimens were immersed in water at room temperature for 24 h, 48 h and 72 h. In time intervals, the specimens were taken out and blotted with filter paper to dry before measuring the sample's weight (W_{wet}). The amount of water absorbed was calculated using equation below:

$$\text{Water absorbed}(\%) = \left(\frac{W_{wet} - W_{dry}}{W_{dry}} \right) \times 100\% \quad (1)$$

Thermogravimetry Analysis

Thermal degradation of the PF board was studied using a Mettler Toledo (TGA/SDTA851) thermogravimetric analyzer. The sample was heated from 30°C to 600°C at a heating rate of 10°C/min.

Flexural Test

Flexural test was performed using a Universal Testometric (Micro 350-10CT) according to ASTM D790. The dimension of the sample was 130.0 × 12.7

× 3.0 mm³. The crosshead speed and span distance were 5.0 mm/min and 100 mm, respectively.

The flexural strength and flexural modulus were calculated by:

$$\text{Flexural strength} = \frac{3PL}{2bd^2} \quad (2)$$

$$\text{Flexural modulus} = \frac{L^3 \Delta P}{4bd^3 \Delta S} \quad (3)$$

where P = ultimate failure load (N), L = support span (mm), b = width of sample (mm), d = thickness of sample (mm), ΔP = increase in load (N), ΔS = increase in deflection (mm).

Impact Test (Izod)

Impact tests were carried out using a Pendulum Tester Ceast Code 6545/000. Unnotched sample with a dimension 65.0 × 12.7 × 3.0 mm was tested according to ASTM D256-73 and five samples were repeated to obtain the average value for each sample.

Hardness Test

Hardness of the samples was obtained using a Shore-D Durometer according to ASTM 2240. Sample was put on a flat surface and a sharp-pointed cone of Shore-D was inserted on the surface of sample. Average value was obtained from at least five areas of each sample.

Scanning Electron Microscope

Morphology for the sample was observed using a Philips XL 30 scanning electron microscope. Sample was coated with a thin layer of gold before the examination.

RESULTS AND DISCUSSION

Density and Water Absorption

Figure 1 shows the effect of EFB fibre content on the density of the PF-EFB board. The density of the control PF board (without fibre) is 0.21 g/cm³ and it increases to 0.52 g/cm³ when the fibre content increases to 30%. This can be attributed to the high density of EFB fibre, i.e. 0.7-1.55 g/cm³ [18]. In

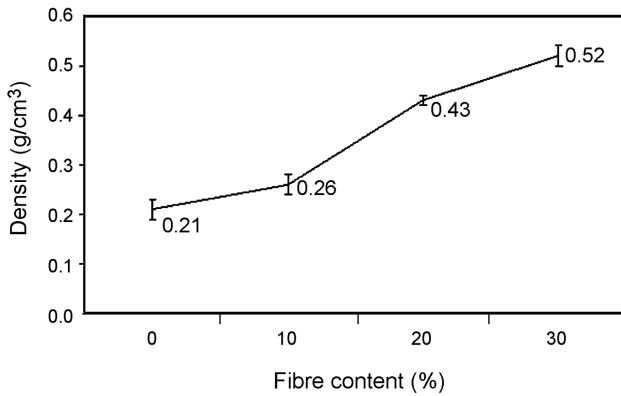


Figure 1. Density of PF board with different fibre content (0-30%).

addition, high pressure during the pressing process may also result in densification of fibre and subsequently increase the density of the composite board. Figure 2 illustrates the water absorption of PF boards with different fibre content. It can be clearly seen that the amount of water absorbed is increased with the fibre content. This is due to the hydrophilic nature and high porosity of the EFB fibres which will enhance the water diffusion process. It should also be noted that the higher the fibre content in the PF board the longer the water absorption takes part to achieve saturation.

Izod Impact and Flexural Tests

The Izod impact of the PF-EFB board with different fibre content is presented in Figure 3. The impact strength of the PF-EFB board increases linearly to

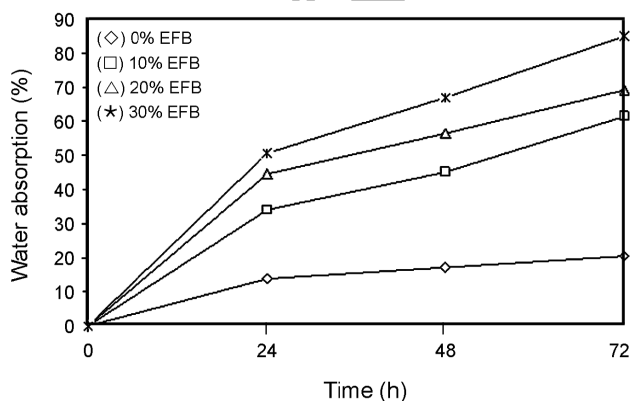


Figure 2. Water absorption of PF board with different fibre content (0-30%).

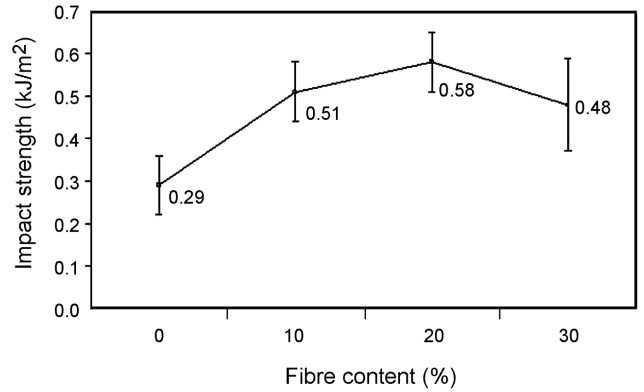


Figure 3. Impact strength of PF board with different fibre content (0-30%).

0.58 kJ/m² with the addition of 20% of EFB fibres. Similar result has been reported in the preparation of kenaf fibre reinforced soy based biocomposites [15]. The presence of EFB fibre in the PF matrix, as fibrous reinforced agent, has strengthened the resulting composite board [14]. The impact strength of the PF-EFB board decreases to 0.48 kJ/m² when the fibre content increases to 30%. According to Sreekala et al. [18], high EFB fibre loading causes difficulties during the processing due to the poor dispersion of fibres in the matrix, leading to weak stress transfer from the matrix to fibres when load is applied [19].

A similar trend can also be observed in the

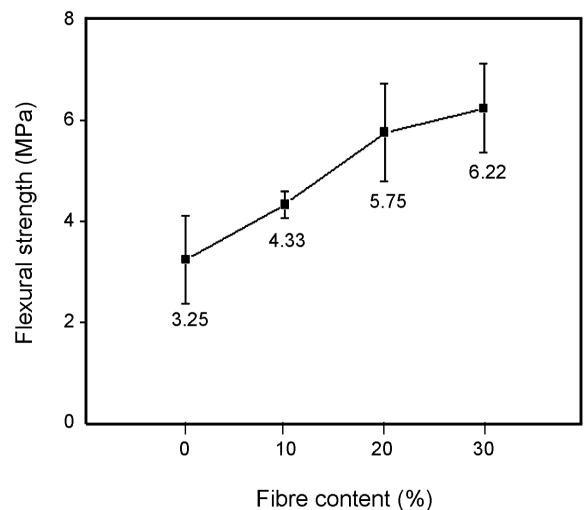


Figure 4. Flexural strength of PF board with different fibre content (0-30%).

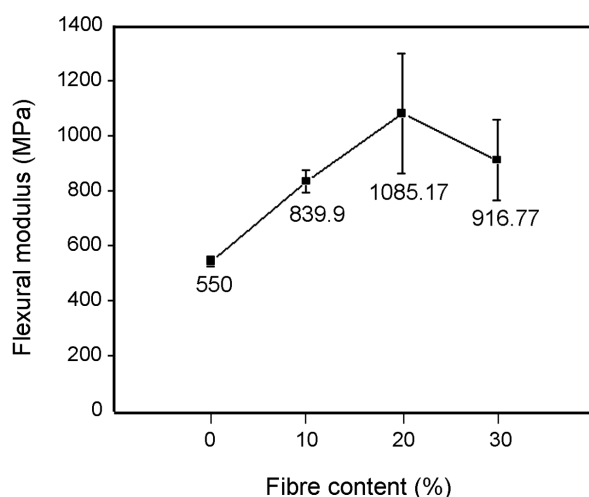


Figure 5. Flexural modulus of PF board with different fibre content (0-30%).

flexural strength and flexural modulus data (Figures 4 and 5). It should be noted that the flexural strength increases linearly with increasing fibre content. While the flexural modulus of the PF-EFB board increases linearly with the addition of EFB fibres and reaches a maximum value of 1085.17 MPa at 20% of EFB fibres, it drops to 916.77 MPa when the EFB fibres content increases to 30%. The reduction may be due to poor dispersion of EFB fibres in the PF matrix. This is consistent with the previous study reported on the preparation of fibre reinforced composite using flax fibres [20].

Hardness Test

The effect of fibre content on the hardness of the PF board is shown in Figure 6. The addition of EFB

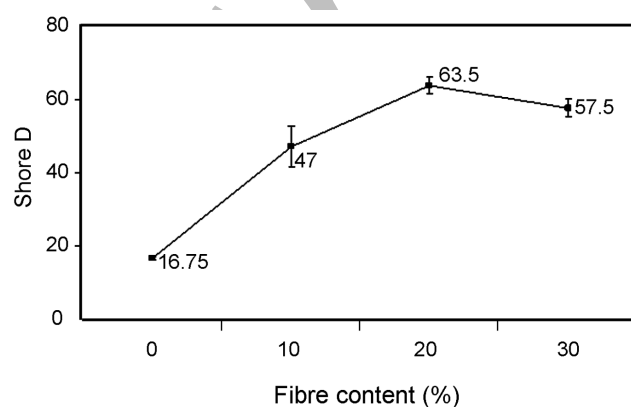


Figure 6. Effect of fibre content on the hardness of PF-EFB boards.

fibres has significantly increased the hardness of the PF board. The hardness of the PF board increases from 16.75 to 63.5 Shore-D when 20% fibre is added. This is because of the strong interfacial bonding strength between fibre and matrix which greatly increases the hardness of PF-EFB board. Due to increase the fibre content, the board becomes stiffer and harder. Thus, an increase in hardness with increasing fibre content was obtained [11]. Beyond 30% fibre content the hardness of the PF board slightly decreases to 54.75 Shore-D.

Thermogravimetric Studies

TG curves of control PF board and PF board with different fibre content (10, 20 and 30%) are presented in Figure 7. It can be seen that all samples undergo three main degradation stages, i.e. around 80-100°C, 200-350°C, 400-600°C. The first stage of degradation is due to the removal of water moisture from the composite boards. The second stage of weight loss of PF and PF-EFB boards with 10, 20 and 30% of EFB fibres are approximately 6.7, 10.8, 14.3 and 18.1%, respectively, which can be attributed to the degradation of EFB fibres, associated to the decomposition of lignocellulosic components. The third stage of degradation of PF and PF-EFB boards with 10, 20 and 30% of EFB fibres are approximately 34.1, 36.1, 42.1 and 42.4%, respectively. This is attributed to the decomposition of the phenol formaldehyde components due to the broken cross-linking network [21].

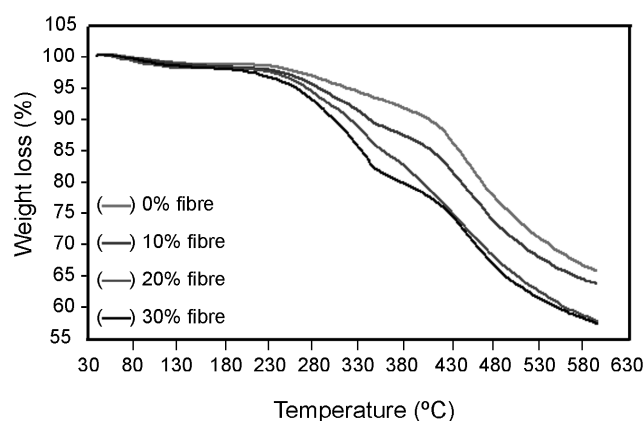
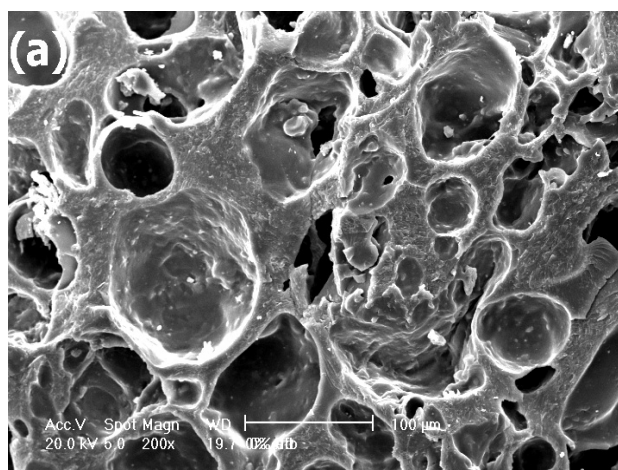


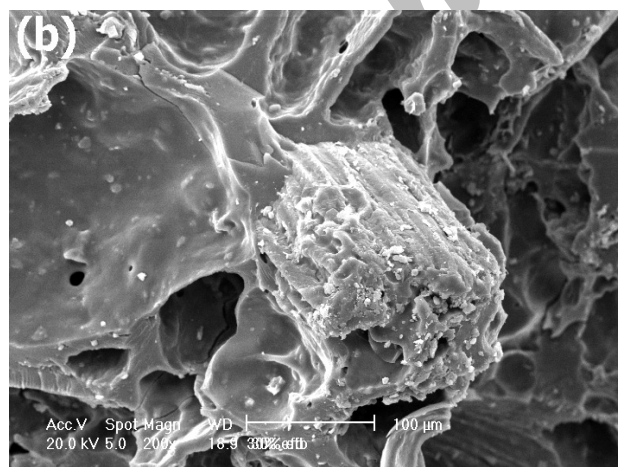
Figure 7. Thermogravimetric curves of PF board with different fibre content (0-30%).

SEM Observations

Figure 8 presents SEM micrographs for the composite board after the impact tests, i.e. (a) control PF board and (b) PF board with 30% of EFB fibre. In Figure 8a, it is obvious that there are numerous voids dispersed throughout the PF matrix, which are associated to water domains formed due to the condensation cure during the formation of PF resin [22]. Therefore, the pure PF board possesses lower impact strength as compared to the PF-EFB boards. It can be clearly seen from Figure 8b, that the EFB fibre is strongly adhered to the PF matrix after the impact test, suggesting good compatibility between the EFB fibres and the PF matrix.



(a)



(b)

Figure 8. SEM micrographs of the PF board (after impact test) (a) without EFB fibre and (b) 30% of EFB fibre.

CONCLUSION

Phenol formaldehyde resin was produced from EFB fibres via liquefaction method using sulphuric acid as catalyst. It was subsequently blended with different contents of EFB fibres as filler to produce PF-EFB composite boards. The results showed that the density and water absorption were increased with increasing fibre content. The mechanical properties, including flexural, impact and hardness of the produced PF-EFB boards were increased linearly and reached maximum with the addition of 20% of EFB fibre. The electron micrographs revealed the good compatibility between the PF matrix and the EFB fibre, which would subsequently enhance the properties of the PF-EFB boards.

ACKNOWLEDGEMENTS

We acknowledge the government of Malaysia for the financial support under project 03-01-02-SF-0030 (MOSTI), Brain Gain (MOSTI), UKM ST-06-FRGS0010-2007 (MOHE) and Research University grant UKM-OUT-BTT-28/2007 (MOHE).

REFERENCES

1. Wen-Jau L, Yi-Chun C, Novolak PF resins prepared from phenol liquefied *Cryptomeria japonica* and used in manufacturing moldings, *Bioresource Technol*, **99**, 7247-7254, 2008.
2. Tatsuhiko Y, Masako A, Satoshi K, Hirokuni O, Chemical analysis of the product in acid-catalyzed solvolysis of cellulose using polyethylene glycol and ethylene carbonate, *J Wood Sci*, **53**, 487-493, 2007.
3. Zhang Y, Atsushi I, Naruhito H, Akio T, Hirokumi O, Tatsuhiko Y, Characterization of liquefied product from cellulose with phenol in the presence of sulfuric acid, *Bioresource Technol*, **97**, 313-321, 2006.
4. Sreekala MS, Kumaran MG, Thomas S, Oil palm fiber: morphology, chemical composition, surface modification and mechanical properties, *J Appl Polym Sci*, **66**, 821-835, 1997.

5. Minowa T, Kundo T, Sudirjot ST, Thermochemical liquefaction of Indonesian biomass residues, *Biom Bioener*, **14**, 517-524, 1998.
6. Ahmadzadeh A, Zakaria S, Preparation of novolak resin by liquefaction of oil palm empty fruit bunches (EFB) and characterization of EFB residue, *Polym-Plast Technol Eng*, **48**, 10-16, 2009.
7. Sun FW, Li XK, Zhang Q, Technologies of liquefaction of bamboo and preparation of adhesive, *Front. For China*, **3**, 493-498, 2008.
8. Wen-jau Lee, Cheng-Tzu Liu, Preparation of liquefied bark-based resol resin and its application to particle board, *J Appl Polym Sci*, **87**, 1837-1841, 2003.
9. Mei-Wen H, Shiao-Wei K, Hew-Der W, Feng-Chih C, Su-Yun F, Miscibility and hydrogen bonding in blends of poly(vinyl acetate) with phenolic resin, *Polymer*, **43**, 2479-2487, 2002.
10. Achary PS, Ramaswamy R, Reactive compatibilization of a nitrile rubber/phenolic resin blend: effect on adhesive and composite properties, *J Appl Polym Sci*, **69**, 1187-2101, 1998.
11. Ismail H, Rozman HD, Jaffri RM, Mohd Ishak ZA, Oil palm wood flour reinforced epoxidized natural rubber composites: the effect of filler content and size, *Eur Polym J*, **33**, 1627-1632, 1997.
12. Hepworth DG, Bruce DM, Vincent JFV, Jeronimidis G, The manufacture and mechanical testing of thermosetting natural fibre composites, *J Mater Sci*, **35**, 293-298, 2000.
13. Singh B, Gupta M, Performance of pultruded jute fibre reinforced phenolic composites as building materials for door frame, *J Polym Environ*, **13**, 127-137, 2005.
14. Sreekala MS, Kumaran MG, Joseph S, Jacob M, Thomas S, Oil palm fibre reinforced phenol-formaldehyde composites: influence of fibre surface modifications on the mechanical performance, *Appl Compos Mater*, **7**, 295-329, 2000.
15. Wanjun L, Lawrence TD, Amar KM, Manjustri M, Influence of processing methods and fiber length on physical properties of kenaf fiber reinforced soy based biocomposites, *Composites Part B: Eng*, **38**, 352-359, 2007.
16. Ahmadzadeh A, Zakaria S, Study on the water absorption and hardness of phenolated oil palm empty fruit bunches (PEFB)-base biocomposite, *Iran Polym J*, **18**, 247-255, 2009.
17. Ahmadzadeh A, Zakaria S, Kinetics of oil palm empty fruit bunch phenolysis in the presence of sulfuric acid as a catalyst, *J Appl Polym Sci*, **106**, 3529-3533, 2007.
18. Sreekala MS, George J, Kumaran MG, Thomas S, The mechanical performance of hybrid phenol-formaldehyde based composites reinforced with glass and oil palm fibres, *Compos Sci Technol*, **62**, 339-353, 2002.
19. Joseph S, Sreekala MS, Oommen Z, Koshy K, Thomas S, A comparison of the mechanical properties of phenol formaldehyde composites reinforced with banana fibres and glass fibres, *Compos Sci Technol*, **62**, 1857-1868, 2002.
20. Arbelaz A, Fernandez B, Valea A, Mondragon I, Mechanical properties of short fibre bundle/poly(ϵ -caprolactone) composites: influence of matrix modification and fiber content, *Carbohydr Polym*, **64**, 224-232, 2006.
21. Wang FY, Ma CCM, Wu WJ, Kinetic parameters of thermal degradation of polyethylene glycol-toughened novolac-type phenolic resin, *J Appl Polym Sci*, **80**, 188-196, 2001.
22. Ku H, Roger D, Davey R, Cardona F, Trada M, Fracture toughness of phenol formaldehyde composites: pilot study, *J Mater Eng Perform*, **17**, 85-90, 2008.