Rigid polyurethane foams from oil palm resources

K. H. BADRI, Z. OTHMAN, S. H. AHMAD
School of Chemical Sciences and Food Technology, Faculty of Science and Technology, Universiti kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

Presently, most polyols used in polyurethane (PU) industry are petrochemical-based where crude oil and coal are used as starting raw materials. However, these materials are expensive, [1] the rate of depletion is high, and they require high technology processing systems. This necessitates a look at utilizing plants that can serve as an alternative feed stocks for monomers in polymer industry. Palmeri oil, vernonia oil, castor oil, and recently carandol oil (extracted from the cashew nut shell), have been synthesized to replace the petrochemical-based polyols [2–4]. Malaysia, a country, which is known globally as the major producer of palm oil, has also participated in researches involving the utilization of palm oil in the oleochemical industry, namely the production of polyurethane polyols. Many works have been published [5–9] regarding this, basically focused on making full use of the oil from the palm tree.

Approximately 22% of the waste from the palm oil industry consists of empty fruit bunch fibers (EFB). These resources, along with a vast quantity of agricultural residues, can be utilized in natural fiber-based composites. The EFB can function as a reinforcing agent or as filler. It is advantageous as compared to the inorganic fillers due to its production cost and higher percentage of filler loading [8, 9]. Moreover, it offers low density, high deformability, no abrasion, and most important, low production cost. In addition, it is obtained from renewable resources and is environmental-friendly. Because the properties of the agro-based fiber result from the chemistry of the cell wall components, modifying the basic chemistry of the cell wall to the polymers can change the basic properties of the fiber. Dimensional stability can be highly improved by bulking the fiber cell wall either with simple bonded chemicals or by impregnation with water-soluble polymers. In most cases the polymer does not enter the cell wall and is located in the cell lumen, so by using this technology the mechanical properties can be greatly enhanced. Coupling agents improve the bonding strength between the fiber and resin. It is the main focus of this study to incorporate organic fillers into the PU to form PU composites in identifying the enhancement properties of filler in refined, bleached, and deodorized, palm kernel oil (RBD PKO) based PU composite. It is also the aim of this study to produce PU foam for insulation purposes. Over the last few years considerable attention has been devoted to these studies: synthesis of the PU polyol from RBD PKO and the production of EFB fiber-filled polyurethane (PU) composites [6–8].

The synthesis was carried out according to the modified procedure of Pourjavadi *et al.* in 1998. The diethanolamine and the potassium acetate were mixed homogeneously in a ratio of 90:10. This reagent and the RBD PKO were mixed to a 20:80 stoichiometric ratio to get the highest functionality value possible. The reaction was carried out in a laboratory scale. The mixture was continuously stirred in a 2-L glass reactor and was reacted at 175–185 °C for 30 min. The samples collected were then analyzed. The EFB fiber was sieved to grade the particles into different sizes. The filler sizes used in this study were of 45–53, 106–150, 300–500 microns. The sieved fibers were then dried in a vacuum oven to a constant weight at 105 °C for approximately 20 hrs [10]. The silane-based coupling agent was mixed with ethanol to form a 10 wt% silanol solution containing silane concentration of 1, 3 and 5% [10]. The surface treatment of the EFB fiber was carried out based on the method suggested by the manufacturer (Dow Corning Sdn Bhd 1995). Mixture of 140 g of crude MDI and 100 g of the resin (100 part by weight (pbw) RBD PKO-based polyol, 2 pbw Tegostab B8408, 0.3 pbw TMHDA, 0.15 pbw PMDETA and 4.5 pbw water) was agitated vigorously using a standard propeller with a speed of 5000 rpm for 10 s at 25 °C. The mixture was poured into a waxed mold, covered, and screwed tightly. The foam was demolded after 10 min. The EFB fiber-filled composites were prepared by using a blend of RBD PKO polyol, surfactant, catalysts, water, and the treated and/or untreated EFB (0, 1.5, 2.5, 3.5, 4.5, 5.5, and 6.5 w/v% of the whole system). Water addition was varied to match the free-rise density to be in the range of 37–39 kg/m^3. Molded foams were characterized for their apparent densities, compressive strength, dimensional stability, and water absorption, following standard method BS4370: Part 1:1988 (Rigid PU foam produced by the press injection method). The insulation value (k-factor or λ-value) of the PU foam and PU composites was determined using the Thermal Conductivity Analyzer model Anacon at a testing temperature of 10 °C and specimen thickness of 20–30 mm. Method BS4370: Part 2: 1993 was followed. A scanning electron microscope (SEM) model Philips, by gold coating ×200 magnification was used to observe the specimens' surfaces.

The derivatized RBD PKO-based polyol is a golden yellow liquid with a cloud point of 13 °C. It has very low moisture content of 0.09% and low viscosity of 374 cps and specific gravity of 0.992 at room temperature. Both FTIR and GC-MS identified the resulting compound as 2-hydroxy-undecanoamide. From the hydroxyl value

0022–2461 © *2004 Kluwer Academic Publishers*

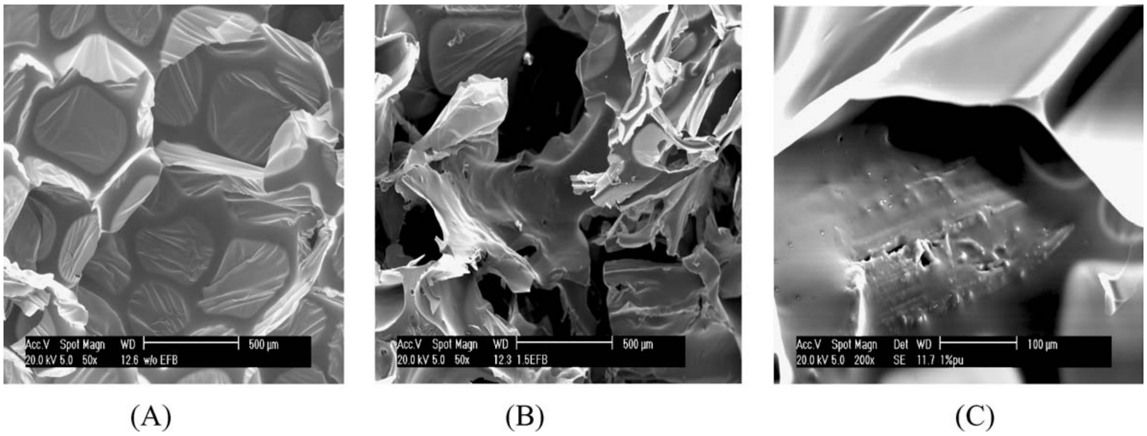

Figure 1 SEM micrographs of (A) the unfilled PU foam, (B) untreated EFB-filled PU foam, and (C) treated EFB-filled PU foam (X200).

obtained and the estimated molecular weight from the GC-MS of 350–370 mg KOH/g sample and 477 respectively, the functionality of the polyesteramide was calculated [11] as 3.0, suitable for rigid foam application. The mechanical properties of the foam produced from the polyol showed good results as compared to its conventional counterpart. For a PU system, the cellular structure played a major role in the determination of good mechanical properties, whereas the untreated EFB-filled PU resulted in ruptured cell walls. This impairment was overcome by the usage of silane as a coupling agent by means of surface modification of the EFB fiber as filler. Maximum stress at 10% deflection of the untreated EFB-filled PU (industrial standard of more than 180 kPa) for 45–53, 106–150 and 300–500 μm was identified at 5.5, 4.5 and 2.5% of fiber loading, respectively. The compressive strength increased to a maximum of 4.5% EFB fiber loading for all concentrations of silane treated PU composites. This increment is almost twice the amount of EFB fiber of the control PU composite, with the untreated EFB fiber (2.5% fiber loading). However, with the addition of 1% silane, the overall strength deteriorated with a maximum value of about 180 kPa at 4.5% fiber loading. Less improvement in strength was observed with the addition of 3% silane to the fiber. The strength dropped below 180 kPa once it reached its maximum value, at about 200 kPa. The optimum values for both the fiber loading and the compressive strength were achieved at 5% silane. The results indicate that EFB fiber has played a role as filler when untreated, and can also be used as a reinforcing agent once treated with silane. This can be observed from the relationship between the compressive strength and the compression modulus. The thermal insulation value, *k*-factor for the unfilled PU, untreated EFB-filled PU, and the treated EFB-filled PU with powder form EFB, are 0.0244, 0.0253, and 0.0229 W/m·K respectively. Inclusion of untreated EFB ruptured the cellular structure due to poor bonding of EFB/matrix, leading to a higher rate of diffusion and increased the k-factor. The tearing shown by the SEM micrograph in Fig. 1B gives a good indication of the occurrence of fiber bundle pull-out, as compared to a uniform cellular structure of the unfilled foam (Fig. 1A). This may explain the poor compressive strength properties of the composites and the low percentage of EFB fiber loading. Fig. 1C of the treated EFB-filled PU gives a good indication of the improvement of interfacial bonding between the constituent phases. The EFB fiber is well bonded to the matrix. This is the possible explanation in describing the improvement in the dimensional stability and the compressive strength of the composites.

Appropriate addition of natural EFB fiber improves the stiffness of the composites but degrades the strength property. This trade off in properties has been encountered with the application of silane coupling agent, which preserves the continuity between the dispersed phase and the matrix. Stress concentration, agglomeration of fibers and dispersion factors can be resolved by employing silane. In terms of application, this composite is most suitable in structures where stiffness and dimensional stability is of prime importance, but is only a secondary choice in areas where structural strength is more vital than component rigidity. Moisture content can lead to poor processability and porous products. Treatment of natural fiber with chemicals can reduce the moisture gain.

References

1. W. I. TAN and S. AHMAD, in Proceedings of the 2001 PIPOC International Palm Oil Congress, organized by Ministry of Primary Industries (Malaysia, 2001) p. 99.
2. A. POURJAVADI, N. REZAI and M. J. ZOHURIAAN-M, *J. Appl. Polym. Sci.* **68** (1998) 173.
3. P. NAYAK, D. K. MISHRA, D. PARIDA, K. C. SAHOO, M. NANDA, S. LENKA and P. L. NAYAK, *ibid.* **63**(5) (1997) 671.
4. A. K. SAHA, S. DAS, D. BHATTA and B.C. MITRA, *ibid.* **71** (1999) 1505.
5. H. D. ROZMAN, B. K. KON, A. ABUSAMAH, R. N. KUMAR and Z. A. MOHD ISHAK, *ibid.* **69** (1998) 1993.
6. K. H. BADRI, S. H. AHMAD and S. ZAKARIA, *ibid.* **81** (2001) 384.
7. *Idem.*, *J. Mater. Sci. Lett.* **19** (2000) 1355.
8. *Idem.*, *Nuclear Sci. J. Malaysia* **18**(2) (2000) 57.
9. H. D. ROZMAN, G. S. TAY, R. N. KUMAR, A. ABUSAMAH, H. ISMAIL and Z. A. MOHD ISHAK, *Eur. Polym. J.* **37**(6) (2001) 1283.
10. Z. A. MOHD ISHAK, A. AMINULLAH, H. ISMAIL and H. D. ROZMAN, *J. Appl. Polym. Sci.* **68** (1999) 2189.
11. G. WOOD, "ICI Handbook" (The ICI PU and John Wiley & Sons Publication, New York, 1991).

Received 6 November 2003
and accepted 1 April 2004