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Effect of Methylene Spacers of Unsaturated Polyester Resins on the Properties of Composites Based on Oil Palm Empty Fruit Bunches and Fiberglass

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Abstract

Monomers of bis-(2-hydroxyethyl) fumarate and bis-(6-hydroxyhexyl) fumarate were synthesized from ethylene glycol and 1,6-hexanediol, respectively, with fumaric acid in the presence of *p*-toluene sulfonic acid as catalyst. The new unsaturated polyester resins were made through condensation reaction between the monomers and terephthaloyl chloride. The chemical structures of these monomers and polymers were confirmed by CHN analyses, FT-IR, ^{1}H-NMR and extended by thermogravimetric analysis (TGA) as well as differential scanning calorimetry (DSC). New oil palm empty fruit bunches (OPEFB) reinforced polyester composites were then derived from the new unsaturated polyester resins containing bis-(2-hydroxyethyl) fumarate and bis-(6-hydroxyhexyl) fumarate. The mechanical properties (tensile, flexural and impact) and water absorption of the new oil palm empty fruit bunches reinforced polyester composites were studied and compared with fiberglass reinforced polyester composites.

Keywords

Composite, methylene spacer, unsaturated polyester, fiberglass

1. Introduction

Unsaturated polyesters have recently been discovered to be an important class of high performance and engineering polymers [1–3]. Unsaturated polyesters are extremely versatile in properties and applications and have been used as a popular matrix thermoset in polymer composites such as fiber-reinforced plastics and polymer concretes. They are widely produced industrially as they possess many advantages compared to other thermosetting resins including good balance of properties, room temperature cure capability and they are relatively inexpensive [4, 5]. However, unsaturated polyesters are generally difficult to process because of their

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high melting temperature or high glass-transition temperature by virtue of their rigid structure [6]. Hence, flexible aliphatic units (–CH_2–) such as ethylene and hexamethylene were introduced in the backbone of the polymers. These methylene spacers enhance the processing and fabrication of polyesters. In addition, fumaric acid as the trans-diacid was used because it is relatively cheap, less corrosive and gives lighter-colored products, whereas the simplest diols increase the tendency of the polyester to crystallize [7]. In this article, we report the synthesis of new unsaturated polyester resins containing methylene groups as a matrix in OPEFB reinforced composites and the effect on the physical and mechanical properties of the composites. We also extended the studies by using fiberglass as the reinforcing material for comparison.

Wide interests exist in the usage of natural fibers as reinforcing materials because of their renewability, easy processibility, biodegradability and environmental friendliness [8]. The reinforcement of polyesters with oil palm empty fruit bunch fibers has been widely reported [9–12]. In this study, oil palm empty fruit bunches (OPEFB) have been chosen as a fiber material because of their low cost, abundance in nature, low density and biodegradability [13, 14]. These have been found to be an important source of fiber for composites and other industrial applications. OPEFB also gives good fiber alignment during composite manufacture [15]. Oil palm is a native commercial crop in Malaysia and over 30 million tonnes of its residues are generated annually [16].

2. Experimental

2.1. Materials

Fumaric acid, 1,6-hexanediol, terephthaloyl chloride, sodium bicarbonate, styrene and *p*-toluene sulfonic acid (all purchased from Fluka, Germany) were used without further purification. Ethylene glycol, ethanol, methylene chloride, toluene and tetrahydrofuran (THF) (all produced by Sigma-Aldrich, USA) were purified by distillation under reduced pressure. Methyl ethyl ketone (MEKP) (supplied from Euro Chemo-Pharma Sdn. Bhd, Malaysia) was used as initiator. Triethylamine (TEA) (from Fisher Scientific, UK) was distilled over calcium hydride (CaH_2) and stored in a septum sealed bottle until use.

2.2. Instrumentation

FT-IR spectra were measured in a Nicolet spectrophotometer, Avatar Model 360 instrument, using KBr pellets and the range studied was from 4000 to 400 cm^{-1}. ^1H-NMR spectra were obtained using a Bruker 400 MHz NMR spectrometer with DMSO-d_6 as the solvent and tetramethylsilane (TMS) as the internal reference. Thermogravimetric analyses (TGA) were carried out in nitrogen with Perkin-Elmer TGA7 series at 10°C/min. Differential scanning calorimetry (DSC) measurements were carried out with a Perkin-Elmer DSC7 series at a heating rate of 10°C/min in nitrogen.

2.3. Preparation of Reactants and Composites

2.3.1. Preparation of Bis-(2-hydroxyethyl) Fumarate (III)

Fumaric acid (0.25 mol) was added to ethylene glycol (1.5 mol) (I) in the presence of *p*-toluene sulfonic acid as catalyst. The mixture was refluxed for 24 h at 120°C with stirring in a 500 ml round bottom flask. After reaction, the solvent was eliminated and the ester was brought to ice water and filtered off. The ester was washed with sodium bicarbonate solution and finally with deionized water. Then, it was dried under vacuum at room temperature and the final purification was carried out by re-crystallization from tetrahydrofuran (THF) to give white crystals. Yield was 70%. Elemental analysis: Found: C, 58.09; H, 4.40; $C_{16}H_{14}O_8$ Calc.: C, 57.93; H, 4.19.

2.3.2. Preparation of Bis-(6-hydroxyhexyl) Fumarate (IV)

This compound was prepared by following the procedure of the preparation of (III) with the fact that ethylene glycol was substituted by 1,6-hexanediol (II). The yield was 68%. Elemental analysis: Found: C, 47.38; H, 6.12; $C_{24}H_{30}O_8$ Calc.: C, 47.06; H, 6.19.

2.3.3. Preparation of Polymers (V, VI)

The general polymerization scheme is illustrated in Fig. 1. Triethylamine (TEA) (0.01 mol) in 25 ml of dichloromethane was added dropwise to the monomer mixtures, III and IV (0.005 mol) with terephthaloyl chloride (0.005 mol) and 25 ml of dichloromethane at 20°C for 5 min. Subsequently, the temperature of the reaction mixture was maintained with stirring for 3 h. The unsaturated polyesters V and VI were precipitated in absolute ethanol, filtered, and washed several times with ethanol then dried for 24 h in a vacuum oven at 75°C. The results are summarized in Table 1.

2.3.4. Preparation of Composites (A, B and C)

In general, composite types A and B are based on new unsaturated polyester resins V and VI, respectively, with OPEFB, while composite type C is based on commercial unsaturated polyester resin produced by Euro Chemo-Pharma Sdn. Bhd with OPEFB. Composite types D and E are based on new unsaturated polyester resins V and VI, respectively, with fiberglass, whereas composite type F is based on the commercial unsaturated polyester resin produced by Euro Chemo-Pharma Sdn. Bhd with fiberglass. The formulation of the unsaturated polyester resins consist of 100 parts resin by weight to 1.5 parts methyl ethyl ketone (MEKP) initiator.

Firstly, fibre bundles from loose oil palm empty fruit bunches (OPEFB) were washed and cleaned to remove unrelated impurities, air-dried for 24 h and oven-dried at 80°C overnight [17, 18]. Then, the OPEFB fibers were cut into 3 cm long strips and dispersed in a sieve which was placed in a tub of water. After an even dispersion was obtained, the sieve was taken out from the tub and water was drained out by pressing the mat against a flat plate. The non-woven OPEFB fiber mat was subsequently dried in an oven for 24 h at 80°C. After that, it was pressed under

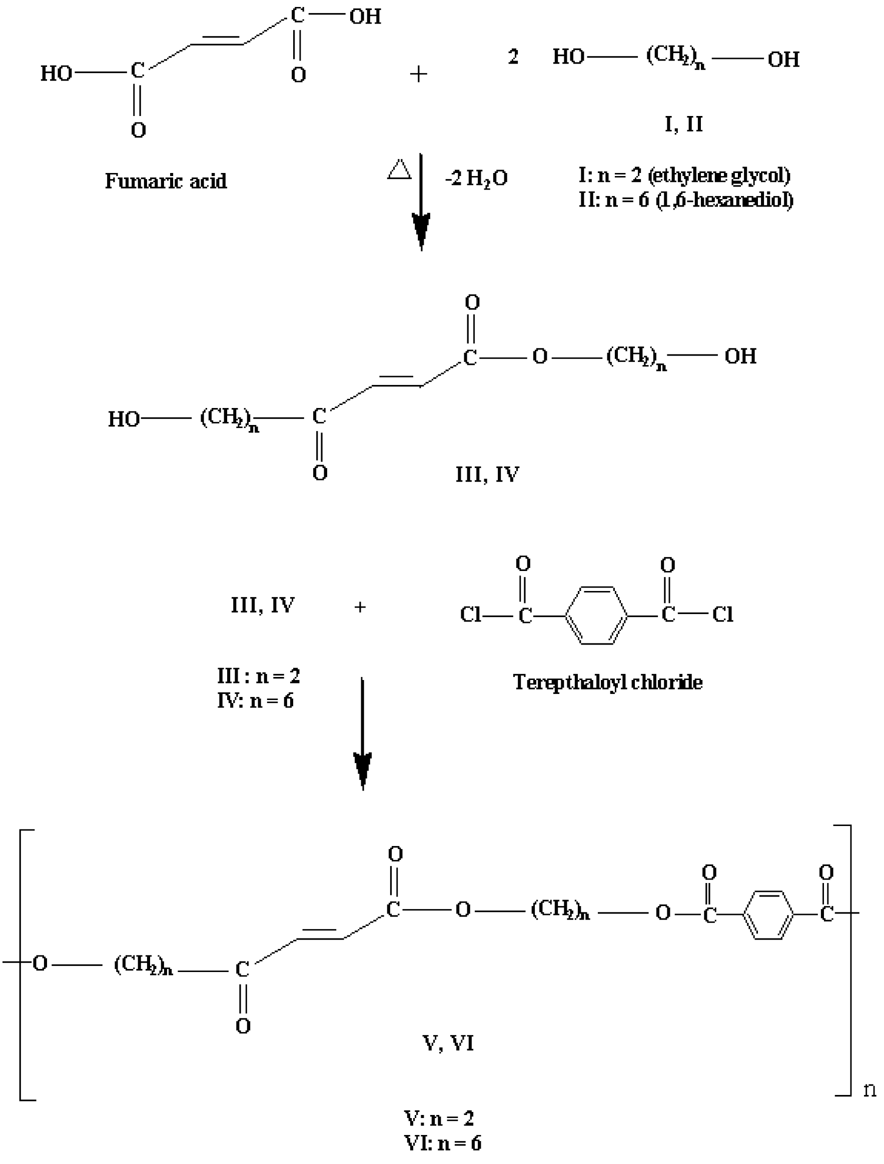

Figure 1. General polymerization scheme of new unsaturated polyester resins.

pressure of 500 psi, followed by the trimming of the uneven edges in order to obtain a uniform shape.

The solid unsaturated polyester resins synthesized were dissolved in hot petroleum ether (60–80°C) to obtain liquid resins and the resins V and VI were mixed well separately with required amounts of MEKP as initiator. Then, the resins were poured onto separate fiber mats by vacuum impregnation for 10 min to ensure good delivery of the resins. The wetted fiber mats in the mould were then placed be-

Table 1.
Yield, elemental analysis and thermal properties of new unsaturated polyesters

Polymer	Yield (%)	C%	H%	${}^{a}T_g$	${}^{b}T_m$
V	67	(57.49) 56.33	(4.19) 3.97	210°C	255°C
VI	68	(47.06) 46.38	(5.88) 5.55	75°C	94°C

[*] The data in the bracket were calculated.
[a] Glass-transition temperature.
[b] Melting point temperature.

tween electrically heated platens of hot press at 100°C. The mould was heated for 25 min at a moulding pressure of 8 MPa. The composites were post-cured at 80°C overnight in an oven.

2.4. Mechanical Tests

The tensile, flexural and impact tests of unsaturated polyester composites were studied. The tensile tests were performed using an Instron Universal Testing Machine Model 1114. The tensile test was carried out by using rectangular strips of 150 × 20 × 10 mm. The width and thickness of the specimens were measured and recorded. Specimens were tested at a cross-head speed of 3 mm/min and gauge length of 60 mm. The tests were carried out in accordance with ASTM D638 [19]. Tensile strength, tensile modulus and elongation at break were then calculated from these test results.

Flexural tests were performed according to ASTM D790 by using the same machine [19]. The width and thickness of the specimens were measured and recorded. Specimens were tested at a cross-head speed of 3 mm/min. Rectangular strips of 160 mm × 20 mm × 10 mm were carefully sanded using a fine graded sandpaper to remove small cracks. The flexural strength and flexural modulus were calculated from this test.

Charpy impact tests were performed using the Impact Pendulum Tester (Zwick) Model CS-1370. The width and thickness of the unnotched specimen were measured and recorded. The tests were carried out in accordance with ASTM D256 [19]. The impact test was carried out on polished samples with dimensions of 70 mm × 15 mm × 10 mm. The samples were rigidly mounted on a vertical position and were struck using a pendulum with a force of 10 J at the center of the samples. From these data the fracture values were calculated. All the specimens were conditioned at ambient temperature (25 ± 3°C) and relative humidity (30% ± 2) before testing.

2.5. Physical Properties

The physical properties basically measure the bulk properties of the composite. Water absorption tests were carried out according to ASTM D570 at ambient temperature (25 ± 3°C) by using Mettler balance type AJ150. The water absorption was determined by weighing the samples at weekly intervals for a period up to 8 weeks [19]. The samples were weighed and immersed in water. They were removed at specified intervals, gently blotted with tissue paper to remove the excess water on the surface, and then the weight was recorded. The water absorption was calculated according to the equation below:

$$\text{Water absorption, WA (\%)} = \frac{M_2 - M_1}{M_1} \times 100,$$

where M_2 is the mass of the sample after immersion (in grams) and M_1 is the mass of the same sample before immersion (in grams).

3. Results and Discussion

3.1. Structural Elucidation

The FT-IR spectra of monomers III and IV showed bands centered at 3400–3439 cm^{-1} characteristic of the hydroxyl groups (–OH) and at 1704–1717 cm^{-1} due to the carbonyl stretching (C=O). In addition, there were absorption bands due to the CH alkene (–HC=CH–) at 3069–3210 cm^{-1}.

The ^{1}H-NMR spectra of the monomers III and IV exhibited characteristic singlet peaks for protons of a hydroxyl group in the range of 4–5 ppm. Two characteristic triplet peaks in the range of 3.6–3.9 ppm and 3.5–4.1 ppm were due to the protons in the $–C\underline{H}_2–OH–$ and $–CH_2–O–CO–$ for monomers III and IV, respectively. The results of elemental and spectroscopic analysis confirmed the structure of the monomers III and IV.

The identity and purity of unsaturated polyesters were confirmed by elemental analysis, FT-IR and ^{1}H-NMR spectroscopy. The FT-IR spectrum of polymer V in Fig. 2 shows the characteristic absorption bands of the carbonyl groups at 1793 cm^{-1} and 1727 cm^{-1}, respectively. This apparently confirmed the ester linkage. Apart from these absorption bands, the presence of the –C=C– aromatic and C–H alkene (–CH=CH–) groups was also observed by absorption bands at 1576 cm^{-1} and at 3067 cm^{-1}, respectively.

The ^{1}H-NMR spectrum of the polymer VI is shown in Fig. 3. Multiplet peaks at 1.3 and 1.6 ppm were due to the protons of $C\underline{H}_2–C\underline{H}_2$ and $C\underline{H}_2–CH_2O$, respectively. The spectrum also showed two triplet peaks at 3.35 and 4.15 ppm assigned to the protons in the ester $–C\underline{H}_2–COO$ linkage and $–CH_2–OCO–Ph–$. In addition, two characteristic peaks centered at 6.2 ppm and 7.3 ppm were assigned to the protons of the alkene and aromatic ring, respectively.

Thermal stability and degradation of unsaturated polyesters V and VI were analyzed based on thermograms obtained from TGA at constant heating rate of 10°C/min in the temperature ranges of 40 to 800°C under nitrogen. Equal weights were used in all experiments to eliminate the mass effect. From the thermogram data, one step at 380°C for 10% degradation (T_{10}) was observed in polymer V. This was due to the degradation of methylene $–(CH_2)_n–$ and ester groups. The char

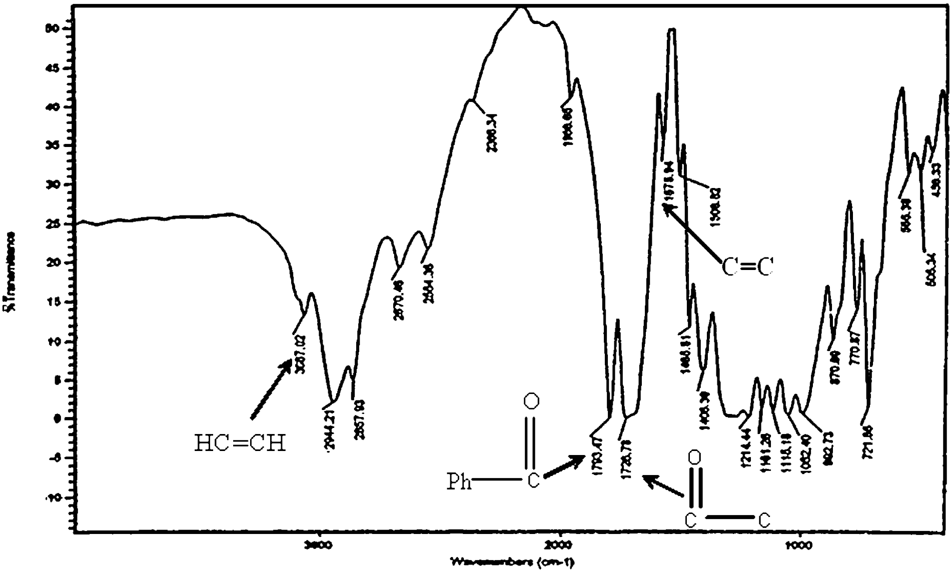

Figure 2. FT-IR spectrum of polymer V.

yield was about 8%. For polymer VI, two steps were observed at 250°C and 380°C due to the cleavage of the aliphatic group and aromatic ester degradation, respectively. The char yield was about 8%. From DSC curves, a single endothermic peak was observed during first heating in the polymers V and VI as illustrated in Figs 4 and 5, respectively. No transition was observed during cooling or second heating. For polymer V, the glass-transition temperature, T_g was 210°C and melting point temperature, T_m was 255°C. While for polymer VI, T_g was 75°C and T_m was 95°C. The results, thus, implied that the presence of methylene spacers in the backbone of the unsaturated polyester decreases the melting point, thus, making it easier to be processed. This is similar to what has been reported by earlier study [7] on the effect of these groups conferring thermal stability in an aromatic polyester.

3.2. Mechanical Properties of Composites

As shown in Table 2, the mechanical properties of the new composites improved with the presence of methylene spacers in the unsaturated polyester matrix.

For tensile properties, composite B gave superior values, followed by composites A and C. Similar results were found for glass fiber as the raw material whereby composite E exhibited the highest tensile values, followed by composites D and F. This is attributed to the structure of unsaturated polyester in composites B and E that consist of six methylene spacers as compared to only two methylene spacers in unsaturated polyester of composites A and D. The increase of methylene spacers in the composites, therefore, increases the flexibility of the main structures in the matrix [7]. The compatibility in the interfacial region between the fibers and matrix

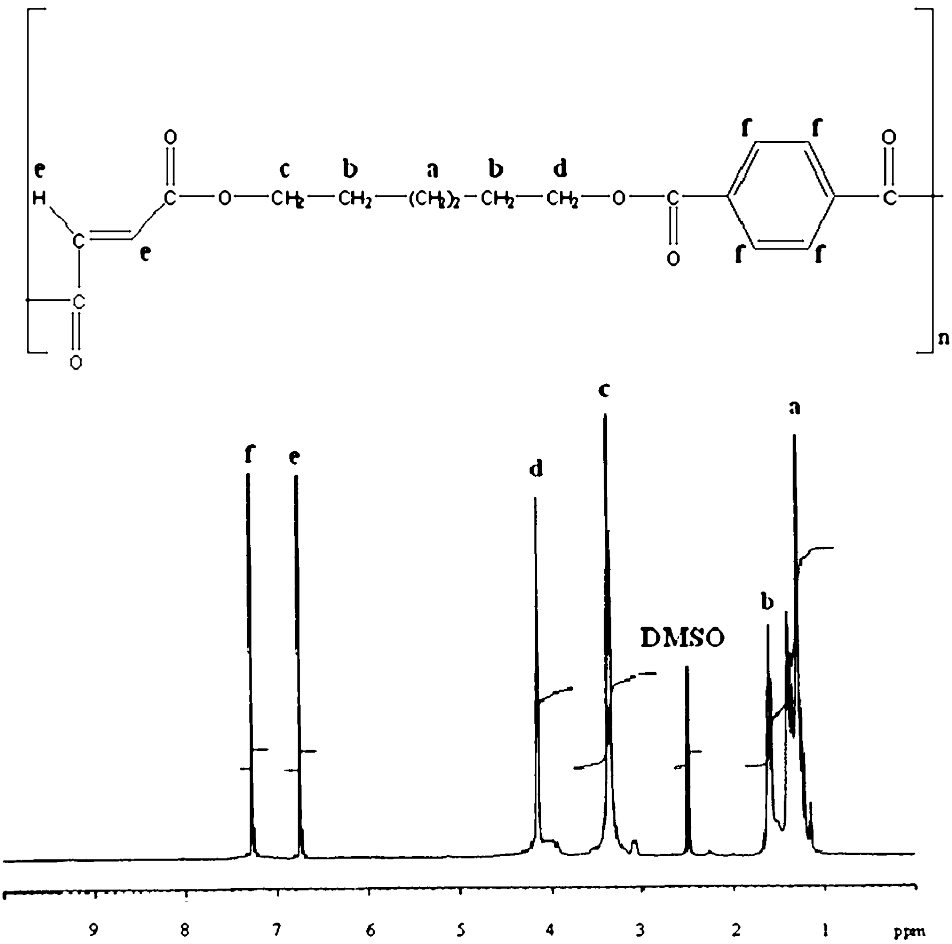

Figure 3. ^{1}H-NMR spectroscopy of polymer VI in DMSO-d_6.

was also enhanced due to the increased dipole interaction of the methylene spacers in the main chain of the matrix.

For flexural properties, composite A gave higher values than composites B and C, whereas composite D was better than composites E and F. The improvements in the flexural properties of the new unsaturated polyester composites were caused by the modifications at the molecular level of the matrix, which led to better fiber–matrix bonds, increased adhesion and more effective stress transfer.

In addition, composites A and D also gave the highest work of fracture values as compared to the other respective composites. It has been reported by Alsina *et al.* [5], that the Charpy impact strength decreases with enhanced fiber–matrix adhesion. Table 2 clearly shows that the impact strength of the new reinforced composites improved significantly by the presence of methylene spacers. In general, composite fracture toughness is influenced by interlaminar and interfacial strength

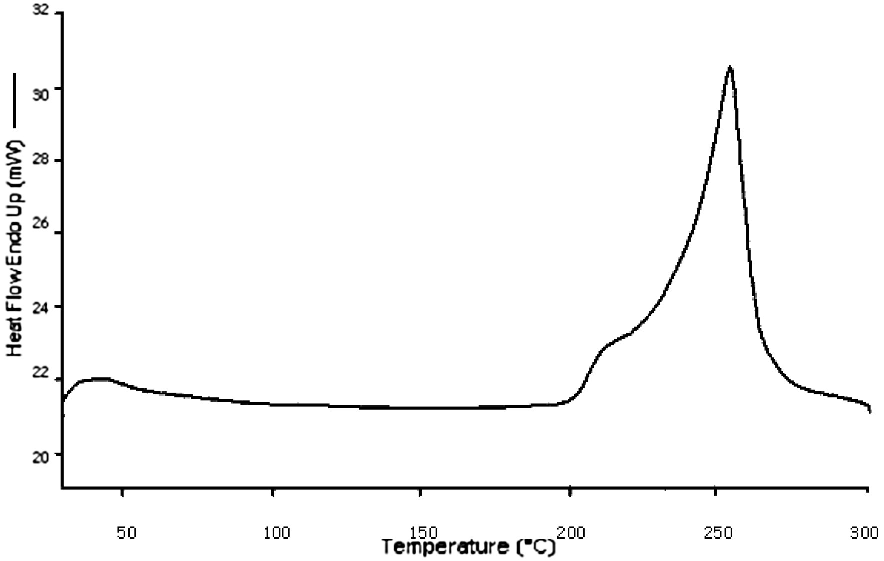

Figure 4. DSC measurement of polymer V.

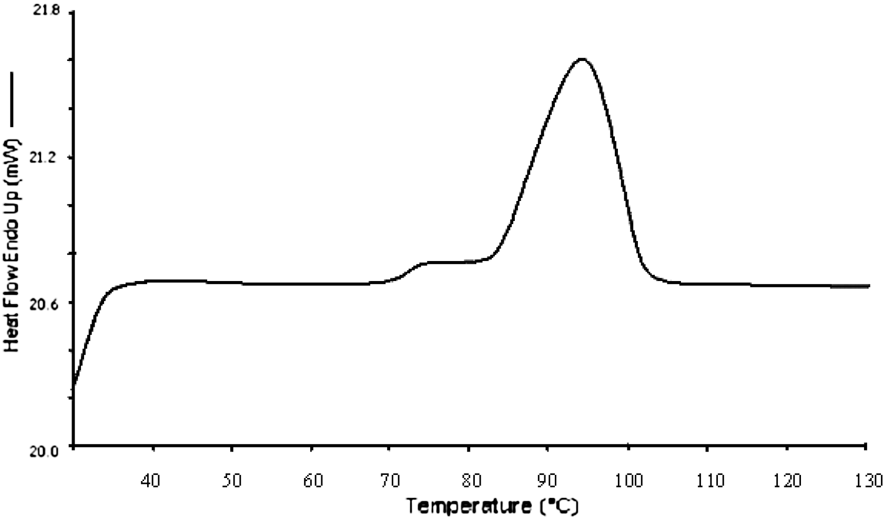

Figure 5. DSC measurement of polymer VI.

parameters. Furthermore, the fibers used simultaneously play an important role in the impact resistance of the composites as they interact with the crack formation in the matrix and response as a medium for transferring stress. In this study, the impact strength increased due to the enhanced flexibility of the interface molecular chains which thereby results in greater energy absorption in the composites. For example, when the cracks move forward, the chain motions also change due to their increased flexibility, thus, creating a barrier to prevent the crack growth.

Table 2.
Mechanical properties of reinforced composites

Composite	Tensile strength (MPa) ± SD	Tensile modulus (GPa) ± SD	Elongation at break (%) ± SD	Flexural strength (MPa) ± SD	Flexural modulus (GPa) ± SD	Impact strength (kJ/m^2) ± SD
Polyester A	43 ± 1.1	3.6 ± 0.7	4.0 ± 0.8	50.29 ± 1.3	4.1 ± 0.7	11.5 ± 0.8
Polyester B	46 ± 0.9	4.1 ± 0.3	4.8 ± 0.5	46.72 ± 1.1	3.8 ± 0.8	10.2 ± 0.9
Polyester C*	35 ± 1.2	2.8 ± 0.6	2.7 ± 0.5	38.86 ± 1.0	3.4 ± 0.5	9.1 ± 0.6
Polyester D	96 ± 1.1	5.76 ± 0.1	2.11 ± 0.6	187.73 ± 1.0	6.4 ± 0.6	75.3 ± 0.2
Polyester E	101 ± 0.8	6.01 ± 0.5	2.5 ± 0.4	182.89 ± 1.1	6.2 ± 0.3	72.2 ± 0.4
Polyester F*	94 ± 1.3	5.52 ± 0.2	2.10 ± 0.2	180.00 ± 1.3	6.1 ± 0.1	68.1 ± 0.6

SD = Standard derivation.
* = commercial resin.

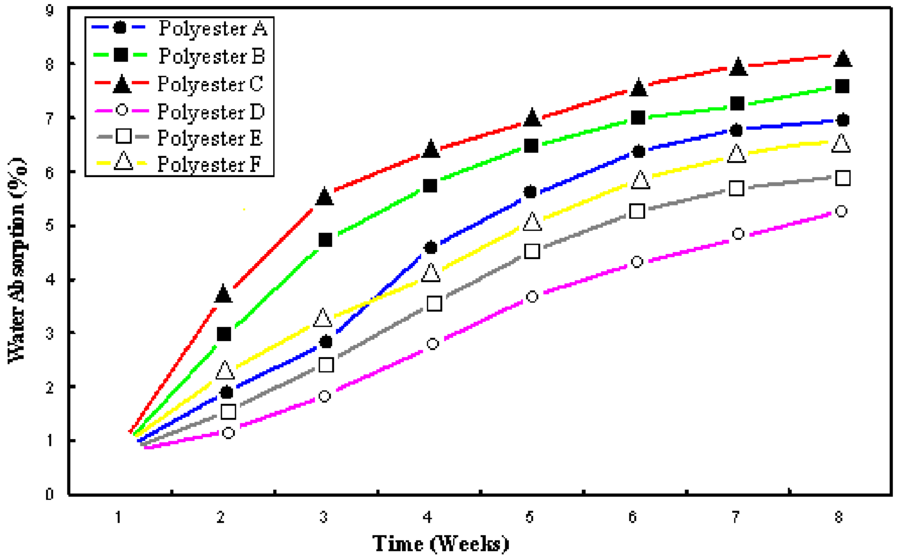

Figure 6. Water absorption of the reinforced composites. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm

3.3. Physical Properties of Composites

The water absorption behavior of composites A, B, C, D, E and F are depicted in Fig. 6. The water absorption curves of the composites displayed a typical Fickian behavior where the mass of water absorbed by the composites increased linearly with the square root of time and then gradually slow down until the equilibrium plateau or complete saturation was reached.

Figure 6 shows that composite C gave the highest percentage of water absorption as compared to composites A and B, whereas for composite E it was higher than for composites D and F. It had been reported by T. Bahman *et al.* [6] that the water absorption characteristic of a composite material is influenced by many factors, such as temperature, fiber volume fraction, orientation of reinforcement, fiber and matrix characteristics; such as polarity, extent of cross-linking or crystallinity of the matrix and presence of residual hardeners, area of exposed surface, duration of immersion in water, and processing techniques as well as surface protection. The presence of a methylene spacer in the unsaturated polyester matrix used decreases the water absorption capability of the composites as compared to commercial unsaturated polyester resin used in composites C and F.

The high water absorption behavior of the oil palm fiber-reinforced composite was mainly due to the highly hydrophilic nature of the oil palm fibers. The hydrophilic nature of the oil palm fibers was caused by the presence of polar groups such as hydroxyl, acetal and ether linkages in the cellulose structure of the fibers. The free hydroxyl groups from the cellulose and lignin structure of the oil palm fiber were able to interact with the water molecules *via* hydrogen bonding [16]. Porous surface structure of the oil palm fiber surface also facilitated the penetration of water into the fiber by capillary action especially when the fiber was exposed to water [11].

High water absorption behavior of the OPEFB composites was attributed to swelling of the oil palm fibers (when exposed to water) leading to crack formation in the composites. Micro-crack formation then created pathways for the water molecules to diffuse into the composite, hence increasing the water uptake. In contrast, the fiberglass composites exhibited poor water absorption characteristics because of the hydrophobic nature of fiberglass.

4. Conclusion

New unsaturated polyesters have been successfully synthesized and characterized by C, H and N elemental analysis, FT-IR and ^{1}H-NMR spectroscopy. The mechanical properties of the unsaturated polyester composites based on six methylene spacers (composites B and E) showed the highest strength as compared to the unsaturated polyester composites based on two methylene spacers (composites A and D). From the overall results, it can be concluded that the incorporation of methylene spacers in the backbone of the unsaturated polyesters enhanced the strength of the OPEFB composites as well as the fiberglass composites. In water absorption, composites based on commercial unsaturated polyester resin (composites C and F) exhibited highest value as compared to composites based on new unsaturated polyester resins (composites A, B, D and E) due to the fact of enhanced bonding and interfacial adhesion between the fiber and matrix.

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