Photo-Fabricated Unsaturated Polyester Resin Composites Reinforced by Kenaf Fibers, Synthesis and Characterization

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ABSTRACT: New bio-fiber composites (UPRC) cured by ultraviolet radiation were produced using kenaf fiber as reinforcing agent and unsaturated polyester resins as matrix in the presence of styrene and IRGACURE 1800 as photoinitiator. Unsaturated polyester resins based on palm oil were prepared from various ratios of monoglyceride (MG)/maleic anhydride (MA) by the interaction of the corresponding MG monomer, with different equivalents of MA, in the presence of 2-methylimidazole as catalyst. The various characteristics of the obtained bio-fiber composites, including mechanical, gel content, water absorption and thickness swelling test, thermal analysis, were determined and the data were discussed. Bio-fiber composite with MG: MA ratio (1 : 4 eq./eq.) showed better mechanical properties (tensile, flexural, and impact strength) than

INTRODUCTION

Natural fiber is renewable and can be used in fiber reinforced composites to replace glass and other nonrenewable fibers. These fibers are cheaper and less abrasive to tools. The utilization of natural fiber reinforcement in synthetic polymer composites has gained importance over the years.¹ Palm oil is a very common cooking ingredient in Southeast Asia. Along with its use in food, it is also used in a wide array of cosmetics and pharmaceuticals and increasingly for biodiesel production. Despite a short-term economic dip, diesel fuel prices are likely to continue to increase over the next decades and so will the demand for alternative sources.² The oil palm industry in Malaysia generates large quantities of biomass. Recent investigations show that various products can be produced from the by-products of the palm oil industry, such as oil palm component plastic composite,³ oil palm component-rubber composite,⁴ sheet-molding compound,⁵ and pulp-paper.⁶

other formulations. Gel content increased as the amount of MA was increased up to the MG: MA ratio was 1 : 4 (eq./ eq.) then slightly decreased at the higher ratio formulation. Bio-fiber composite (UPRC_c) was considered the best prepared bio-fiber composite which contained higher degree double bond, cross-linking and thermal stability. Moreover, morphological study of selected examples of the formed bio-fiber composites was also carried out and showed the evidence of the enhancement of the compatibility between fiber and polymer matrix. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 123: 968–976, 2012

Key words: composites; unsaturated polyester resins; palm oil; ultraviolet radiation

Besides low cost, natural fibers in composites have lower specific gravity, higher specific tensile strength, biodegradable, and are less energy intensive to process.⁷ Ultraviolet curable systems are used in surface coatings and have several distinct advantages such as solvent free, environmentally friendly, and energy saving.⁸ The entire curing process occurs at room temperature and hence does not require expensive high temperature equipment for the curing.⁹ The thermoset fiber reinforced polymer matrix composites are currently produced from unsaturated polyester resins by thermally induced initiators. Examples are polyester prepregs, sheet-molding compounds, bulk molding compounds etc.¹⁰ The production of these composites involves high temperatures and requires expensive molds and hydraulic presses. Unsaturated polyester resins (UPR) is one of the widely used thermoset polymers in polymeric composites, due to their good mechanical properties and relatively inexpensive prices. The curing reaction is a free radical polymerization of C=C double bonds present in the UPR with the presence of an unsaturated solvent which is usually styrene. The resins are processed over a wide temperature range to form sheet-molding compounds. They possess a wide range of properties and are

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used in marine, automobile, construction, sport, and furniture applications.¹¹ Room temperature curing of such systems takes a long time. Ultraviolet radiation are widely used throughout the world and also in Malaysia in surface coatings especially in wood industry and followed by printing, electronic, and adhesive industry.¹² In recent times, photo-curing process has been adopted for making composites that may prove superior to the thermal curing systems. This process employs ultraviolet radiation to cure the composites. In view of the advantages of ultraviolet curing system and the success of the photo-fabrication of polymer matrix composites by the use of ultraviolet radiation,¹³ attempts have been made to apply the radiation curing technology for the production of bio-fiber composites. Based on the above advantages and the success of the photo-fabrication of polymer matrix composites, the work described in this article involved the production of composites with unsaturated polyester as matrix reinforced with kenaf fibers with the employment of ultraviolet radiation to cure the polymers matrix. Various testing were carried out to study the composites. Moreover, a comparative study was conducted on the obtained UPR/composites of various formulations.

EXPERIMENTAL

Materials and solvents

Kenaf fiber obtained from National Tobacco Board, Malaysia. Palm oil was purchased from local market in Malaysia, maleic anhydride (MA) (Fluka, Analytical \geq 98.00% mp = 52–54°C), glycerol reagent plus $(Sigma, \geq 99.00\% FW = 92.09, FP = 160^{\circ}C, bp =$ $182^{\circ}C/20 \text{ mm Hg}, d = 1.262), 2-methylimidazole$ (Fluka, \geq 98%, mp = 144–146°C) sodium hydroxide (System company \geq 99.00%), molecular sieve (Fluka, type 3A) styrene (ς -Aldrich, \geq 99.00% d = 0.909), pyridine (Aldrich, \geq 99%, bp = 115.2°C, d = 0.9819), hydroquinone (System company, >99%, mp = 171-175°C) and photoinitiator IRGACURE 1800 was supplied by (Ciba Specialty, Singapore) Pte. Ltd. All the materials mentioned before were used without purification. Solvents with different solubility parameters and of analytical grade, such as toluene [8.9 (cal/ $(cm^3)^{1/2}$,¹⁴ were used. All the other solvents was purified as reported in the literature.¹⁵

Methods

Preparation of ultraviolet-cured UPR

*Monomer preparation*¹⁶. In a 500 mL reaction flask equipped with a mechanical stirrer (2000 rpm/min) with dry nitrogen inlet and outlet. A mixture of palm oil (162.26 g) and 0.1% sodium hydroxide were

placed in the reaction flask, 27.74 mL of glycerol (within the molar ratio 1 : 2.2) was put inside the dropping funnel which was fitted to the reaction flask and was added in a drop-wise manner. For the first 5 min, nitrogen gas was allowed to flow swiftly. It was then adjusted to slow rate throughout the experiment. The reaction mixture was heated to 220–240°C for 3–4 h. After that, a small amount of sample was taken out by means of glass rod to test the solubility in ethanol. The test continued until solution was obtained where no emulsion or white spots were observed. Then the mixture was cooled in ice water, and then poured into sealed beaker for the next experiment.

Polymers matrices preparation¹⁶. In a round-bottomed reaction flask equipped with a mechanical stirrer (2000 rpm/min), 1 eq. of monoglyceride (MG) monomer was heated up to melt, then a mixture of 0.15% of hydroquinone was added to retard any polymerization reaction that may occur. Variable amounts of MA (2-5 eq.), 1-1.5% of 2-methylimidazole as catalyst were added to the reaction mixture with constant stirring. The mixture was subsequently stirred at temperature not more than 90°C for 5 h, and then it was cooled down giving a dark viscous liquid. The resulting UPR were dried and poured into sealed beaker for the next experiment. All the symbols and the chemical compositions for the synthesized UPR/composites were listed in Table I.

Photo-fabrication of UPR composites

Fiber mat preparation. Kenaf fibers have been combed using laboratory Carding Machine-337A, (MESDAN, Italy), then the fibers were formed into mat (20×20 cm²) in a deckle box by the procedure reported earlier¹³ and dried in an oven at 105°C for 24 h. After that, mats were passed through needle punch Testing Machine model (SNP-50), Shoou Shyng Machinery Co. with speed 200 strokes/min. Finally, the needle punched mats were pressed using hot press Gotech Testing Machine Inc model GT-7014-100 (2 min, 500 kg/cm², 100°C) and the thickness of the mats were controlled to ~ 1 mm.

Preparation of composites. The previously prepared UPR polymers matrices (100 g) were mixed with styrene (30 mL, 30%) and photoinitiator (IRGACURE 1800, 3 g, 3%). The mixture was stirred for 10 min with mechanical stirrer, evacuated under vacuum (30 mmHg, 20 min) to remove the air bubbles if any. The whole mixture was then poured onto kenaf fiber mat. The mat was impregnated with resin mixture and the air bubbles if any were removed by means of a hand roller. The mat was then sandwiched between two glass panels. After impregnation, mats were passed through ¹ST UV machine (model M20-

Tensile Properties (Strength, Modulus, Strain, Toughness) for UPRC _{a-d}						
Tensile properties				operties		
Composite symbol	MG : MA (eq./eq.)	Strength (MPa)	Modulus (GPa)	Strain (%)	Toughness (kJ/m ²)	
UPRC _a UPRC _b UPRC _c UPRC _d	1:2 1:3 1:4 1:5	$3.99 (\pm 0.52)$ 11.57 (±0.50) 12.07 (±1.20) 11.59 (±2.08)	$\begin{array}{c} 173.10 (\pm 1.78) \\ 451.14 (\pm 2.05) \\ 738.79 (\pm 1.79) \\ 866.42 (\pm 1.07) \end{array}$	$\begin{array}{c} 1.82 \ (\pm 0.14) \\ 1.78 \ (\pm 0.12) \\ 1.54 \ (\pm 0.22) \\ 1.27 \ (\pm 0.20) \end{array}$	$\begin{array}{c} 8.89 \ (\pm 1.48) \\ 14.99 \ (\pm 1.86) \\ 15.38 \ (\pm 3.78) \\ 6.87 \ (\pm 2.04) \end{array}$	

TABLE I

1-Tr-SLC) for 20 passes (each pass $28.3 \times 10 \text{ uW}/$ cm²) at conveyor speed of 5 m/min. The machine consists of a medium pressure mercury arc lamp with UV radiation wavenumber of 180-450 nm.

Composite characterizations

Mechanical properties (tensile, flexural, and impact)

The obtained UPR/composites were cut into three types of test sample: i.e., flexural, tensile, and impact tests. Tensile tests were carried out on samples with dimension of $10 \times 1.5 \times 0.2$ cm³ (length \times width \times thickness), using Universal Testing Machine model GT-TGS-2000 at a cross-head speed of 1.0 mm/min, according to ASTM D-3039. The properties were reported based on the average of five measurements for each UPR/composite. Properties such as strength, modulus, strain, and toughness were obtained from this test. Flexural tests were conducted according to ASTM D-790, i.e., a three-point bending method using Universal Testing machine model GT-TGS-2000. The samples, with dimension of 8 \times 1.2 \times 0.2 cm³ (length \times width \times thickness) were tested at a cross-head speed of 1.0 mm/min. Five replicates were prepared for this test for each UPR/composite composition. Properties such as strength, modulus, and toughness were computed.

The properties were reported after five measurements for each were taken and properties such as strength, modulus, and toughness were computed. Impact test was carried out using Pendulum Tester (Zwick) Model 5101 (charpy method) with sample dimension of 6.5 \times 1.2 \times 0.2 cm³ (length \times width \times thickness). The pendulum energy of 2 J was used for all the samples. The test was conducted according to ASTM D252, a minimum of five samples were tested in each case. The impact strength of the sample was calculated.

Gel content

The degree of curing for the formed UPR/composites was determined by Gel Content test according to ASTM D2765. Gel content test was determined by Soxhlet extraction using toluene as solvent. The cured sheets were weighed and placed in a cellulose extraction thimble in the Soxhlet's extractor. Four sets of Soxhlet's extractor have been used. The solvent extraction was carried out with 250 mL toluene for 3 h. After that, the samples were taken out followed by vacuum dried and re-weighed until a constant weight was achieved. Gel content of the coating was calculated according to the following equation:

Gel content
$$\% = \{w_2/w_1\} \times 100$$

where, w_1 and w_2 represent the weight before and after extraction, respectively.

Thermal analyses

Thermogravimetric analysis (TGA) and differential thermogravimetric analysis (DrTGA) for the resulting composites were carried out as the following: The predried UPR/composites were tested using Perkin-Elmer thermogravimetric analyzer Pyris, 1 TGA. Sample weight in the range of 15–20 mg was heated at 10°C/min in the range of 50-800°C with nitrogen atmosphere; one measurement was made for each sample.

Scanning electron microscopy (SEM) study

The fracture surfaces of test UPR/composite samples were investigated with a Leica Cambridge S-360 scanning electron microscope (Leica, Milton Keynes, Buckinghamshire, UK). The specimens were mounted on aluminum stub and sputter coated with a thin layer of gold to avoid electrostatic charging during examination.

Water absorption and thickness swelling test

Samples (30 mm \times 12 mm \times 2 mm) were used for the measurement of the water absorption and thickness swelling. After being oven-dried at 100°C to a constant weight, the specimens were immersed in boiled out distilled water. The specimens were periodically taken out of the water, surface-dried with absorbent paper, and reweighed; their thickness was re-measured, and they were immediately put back

into the water. The water absorption and thickness swelling were calculated according to the following formulas:

Water absorption (%) =
$$M_2 - M_1/M_1 \times 100$$

where M_1 and M_2 are the masses (g) of the sample before and after immersion.

Thickness swelling $(\%) = t_w - t_0/t_0 \times 100$

where t_0 and t_w are the thicknesses (mm) of the sample before and after immersion.

RESULTS AND DISCUSSION

The main target of this study is to synthesize a new UPRC_{a-d} based on palm oil and ultraviolet curing and make a comparative study to select the best composite composition based on variable ratios of MG: MA. The various characteristics of the resulting UPRC_{a-d} including mechanical properties (tensile, flexural, and impact), gel content, water absorption, and thickness swelling test and thermal analysis were determined and the data were discussed. In addition, scanning electron microscopy (SEM) study of selected examples of UPRC was also investigated as shown below.

However, these new bio-fibers composites necessitate the preparation of monomer as well as polymer matrix as precursors.

Photo-fabrication of UPRC_{a-d}

Before polymerization process, MG monomer was prepared by the interaction of palm oil and glycerol in the presence of sodium hydroxide as basic catalyst. UPR based on palm oil were prepared with various MG : MA ratios as reported in our work¹⁶ by the interaction of the corresponding MG monomer, with different equivalents of MA, in the presence of 2-methylimidazole as catalyst. Finally, bio-fiber composites UPRC_{a-d} of various formulations were synthesized using ultraviolet curing technique in the presence of kenaf fiber mat, styrene and free radical initiator IRGACURE 1800. The general schematic diagram for the resulting UPRC_{a-d} is illustrated in Figure 1.

UPRC characterizations

The mechanical behavior of variable bio-fiber UPRC compositions was achieved by tensile, flexural, and impact tests. Table I shows the tensile properties (strength, strain, modulus and toughness) of all UPRC formulations using variable MA content. It is generally believed that the increase in the MA con-



UPR / composites UPRC_{ad}

Figure 1 Schematic diagram of UPRC_{a-d} preparation.

tent as a source of double bond in the structure enhances the properties of the bio-fiber composite as mentioned in previous work.¹⁶ All the prepared UPRC_{a-d} has the same composition except the amount of MA presented in each form. From the data presented in Table I, it can be seen that the tensile strength increases as the MA content within the bio-fiber composite is increased and reach a threshold value in $UPRC_c$ (MA 4 eq.), followed by a slight decrease in $UPRC_d$ (MA 5 eq.). Tensile toughness showed nearly the same behavior as tensile strength in the first three formulations UPRC_{a-c} and reaches a threshold value in UPRC_c. Then sharp decrease is observed in the fourth formulation $UPRC_d$. Moreover, the tensile modulus for the UPRC shows a sharp increase as the MA content is increased starting from 173.10 (±1.78) for UPRC_a up to 866.42 (± 1.07) for UPRC_d. This may be attributed to the higher rigidity within the bio-fiber composite as the MA content is increased due to more cross-linking. No significant trend is observed for the tensile strain % where the results are in the range 1.82 (± 0.14)– 1.27 (± 0.20). Thus, the results indicate that in the composite $UPRC_c$ with higher MA content, the stress distribution is more efficient than in composite UPRC_a with lower content. This is probably due to the ability of the C=C groups in styrene polymerize through a radical process with C=C sites along polyester chains which was supplied by MA. These would create a better and stronger bridging in the composite. The resulting composites display an increasing trend of tensile strength and modulus as the maleic content increases within the polymer structure till UPRC_c with a MG : MA ratio 1 : 4 (eq./ eq.). The distribution of stress transfer from the matrix to the fiber is somewhat improved. This may be attributed to the increased compatibility at the interfacial region between fiber and the polyester matrix. The increased compatibility may affect the mobility of the resin polymer, thus, resulting in the increased modulus. This phenomenon is further supported by the SEM study which will be discussed later. Table II shows the flexural properties (strength, modulus, and toughness) of the UPRC_{a-d}. A similar

Unsaturated Polyester Resins + Styrene + Kenaf fiber mat

9	1	2	

Flexural and Impact Properties for UPRC _{a-d}							
			Impact				
Composite symbol	MG : MA (eq./eq.)	Strength (MPa)	Modulus (GPa)	Toughness (kJ/m ²)	strength (KJ/m ²)		
UPRC _a UPRC _b UPRC _c UPRC _d	1:2 1:3 1:4 1:5	$\begin{array}{c} 3.73 \ (\pm 0.54) \\ 7.00 \ (\pm 1.69) \\ 9.20 \ (\pm 0.49) \\ 6.53 \ (\pm 1.12) \end{array}$	$\begin{array}{c} 116.18 \ (\pm 0.72) \\ 382.26 \ (\pm 0.90) \\ 545.19 \ (\pm 0.45) \\ 632.65 \ (\pm 0.76) \end{array}$	$\begin{array}{c} 0.88 \ (\pm 0.24) \\ 1.94 \ (\pm 0.39) \\ 2.14 \ (\pm 0.12) \\ 0.93 \ (\pm 0.26) \end{array}$	1.05 (±0.33) 1.70 (±0.20) 1.92 (±0.27) 1.40 (±0.22)		

TABLE II

trend as shown in tensile properties is observed except for flexural strength and toughness for UPRC $_d$ which shows sharp decrease, which may be due to the brittle structure of these UPRC formulation. For the impact strength which is depicted in Table II, UPRC formulations show the same trend as for both tensile and flexural strength. Hence, the same explanation probably holds in this regard.

Gel content refers to the degree of curing that occurs for each UPRC formulation. From the results given in Table III, it can be clarified that, all the UPRC formulations have high gel content of more than 93% except UPRC_a which shows low gel content value 89.23% (± 0.32). UPRC_c shows higher gel content value 94.78% (±0.22). The order of gel content values for all UPRC formulations is in the following manner: $UPRC_c > UPRC_d > UPRC_b >$ UPRC_a. The higher the gel content the higher the degree of curing in the bio-fiber composite formulation which is due to the higher equivalent of MA content and the higher degree of cross-linking. Gel fraction increases as the amount of MA is increased up to the ratio of MG to MA was 1 : 4 (eq./eq.) for UPRC_c, while decreases for the last formulation 1:5(eq./eq.), which may be attributed to the prereaction of the unsaturated groups during the preparation of the resin. It is believed that there is unreacted styrene which can be easily extracted using soxhlet extraction due to insufficient of unsaturated group in the resin. Hence, these (unreacted styrene) can act as stress concentration point and subsequently lower the strength of the composite. As the quantity of MA increases within the polymer matrix the extent of UV radiation being absorbed is also increased. This would leave more light energy to cure the styrene monomer to produce cross-linked polymer matrix. During the curing of UPRC by UV radiation,

TABLE III Gel Content Test for UPRC_{a-d}

Composite symbol	MG : MA (eq./eq.)	Gel content %
UPRC _a	1:2	89.23% (±0.32)
UPRC _b	1:3	93.15% (±0.18)
UPRC _c	1:4	94.78% (±0.22)
UPRC _d	1:5	94.18% (±0.15)

the more photoinitiator would be able to absorb the radiation and disintegrate to produce free radicals. Hence, more cross-links would be produced through the unsaturation sites along the polymer matrix molecular chain. From all the above results, it was found that, bio-fiber composite UPRC_c was considered the best prepared bio-fiber composite which contain higher degree of double bond and cross-linking. It can be explained that more energy is needed to fail UPRC_c. This phenomenon is resulted from a better cross-linking network that has been introduced into the bio-fiber composite structure when such specific MA content is employed.

The thermal behavior of UPRC_{a-d} was evaluated by TGA and DTG nitrogen atmosphere at a heating rate of 10°C min⁻¹. TGA curves show a small weight loss in the range 2-3% starting at 82°C until 120°C which may be attributed to loss of moisture and entrapped solvents. The thermographs of the prepared bio-fiber composites UPRC_a as a selected example are given in (Fig. 2), while Table IV gives the temperatures for various % weight losses. The thermographs also indicated that, all the UPRC_{a-d} decomposition three stages and depend upon the nature of the unsaturated polyesters,^{17,18} the first stage between 195 and 290°C. The second and the third stages of degradation of bio-fiber composites are usually overlapped and occurred between 310 and 430°C. The rate of degradation in the second and third stages is somewhat faster than in the first stage. The first degradation step involves the scission of cross-links/weak links with the liberation of free linear chains. The second step involves a random scission of the free linear chains into smaller fragments. This observation is in agreement with that observed by Bansal et al.¹⁹ The initial decomposition temperature (IDT)²⁰ corresponds to the temperature, at which the initial degradation may occur. IDT of all the UPRC_{a-d} appeared at T₁₀ which is considered the same time for the polymer decomposition temperature (PDT)^{21,22}; it occurred in the range 213-258°C. Therefore, the data in Table IV indicate that the thermal stabilities of UPRC_{a-d} (at 10%) are in the order $UPRC_c > UPRC_d > UPRC_b > UPRC_a$. On comparison the T_{40} and T_{50} values of UPRC_c with other formulations, it was found that the later



Figure 2 The TGA and DrTGA trace of UPRC_a. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

formulations are less stable than the former. This may be attributed to higher degree of double bond and cross-linking which presented in such composite formulation. The final decomposition temperature (FDT) corresponds to the temperature at which the rate of degradation that may occur is nearly completed. It can be clarified from the thermograms that the FDT for all UPRC_{a-d} is nearly completed at around 430°C. This indicates that the UPRC formulations are somewhat have high thermal stability, i.e., over 400°C.

Figures 3 and 4 show the results of water absorption and thickness swelling for $UPRC_{a-d}$ as a function of immersion time in days. Each data point represents the average of four specimens. In general, lignocellulosic material is hydrophilic with the presence of OH groups in the cell wall which comes

TABLE IVThermal Properties of UPRC_{a-d}

Composite	Tem	°) perature dec	C) for vari compositio	ious percei ns ^a	ntage
symbol	10%	20%	30%	40%	50%
UPRC _a UPRC _b UPRC _c UPRC _d	214 227 258 244	291 318 332 326	339 354 360 357	364 371 375 374	378 383 388 387

^a The values were determined by TGA at heating rate of 10° C min⁻¹.

from cellulose and hemicellulose. These groups absorb water through the formation of hydrogen bonding. Like other wood fibers, kenaf are also hydrophilic. This is because they have the same cell wall polymers that can interact with water molecules via hydrogen bonding. The fiber cell wall polymers have hydroxyl- and oxygen containing groups that can interact with water molecules via hydrogen bonding. Fiber-water interaction can lead to a build up of moisture in the fiber cell wall that would result in fiber swelling and adversely affect dimensional stability of such composites. It can be clarified from given data that, all UPRC have significant values from water uptake and thickness swelling ranging between 9.66% (±1.01)–16.80% (±0.88) and 5.39% (±0.58)–10.63% (±0.86), respectively. This is explained by the hydrophilic nature of the material. As for UPRC composites, appreciable amount of water uptake is observed. $UPRC_d$ shows the highest ability to absorb water and thickness swelling up to 16.80% (±0.88) and 10.63% (±0.86), respectively, as illustrated in Figures 3 and 4. It is then reasonable to assume that all the water was absorbed by fiber and also voids that may present supplied by the weak interaction between the fiber the matrix. Like other wood fibers, kenaf embedded in the UPR polymer matrix is also hydrophilic. This is because they have the same cell wall polymers that can interact with water molecules via hydrogen bonding. Voids can exist particularly at the fiber-matrix interface when



Figure 3 Thickness swelling % of UPRC_{a-d}.

the fiber is poorly wet by the matrix polymer due to inferior fiber–matrix interaction. UPRC_d composites have inferior fiber–matrix interface. This is evident from the SEM micrographs of the controls as shown later in Figure 7 which display fiber pull-outs during failure and the existence of moderate voids between the fibers and the polymer matrix. As a result, transportation of water molecules via capillary action to voids as well as flaws at the fiber–matrix interface can be expected. Water molecules accumulated at the interfacial voids contribute to the total amount of water uptake by the composite.

The morphological studies of UV-cured bio-fiber composites UPRC_c, UPRC_a, and UPRC_d were examined by SEM micrographs as shown in Figures 5 and 6 and 7, respectively. Figure 5 shows the SEM micrograph of UPRC_c composite. It is obvious that the fibers are tightly embedded in the matrix with evidence of fiber breakage during failure. This indicates good wetting between the fiber and matrix.

Without good wetting, a strong interfacial adhesion cannot be achieved.^{23,24} According to Cautinho et al.,²⁵ interfacial adhesion is crucial in getting optimum strength properties of composites. In addition, the micrograph shows a good distribution of fiber within the polymer matrix, while the fiber distribution is vital in allowing more light to penetrate and causes complete matrix cure. Moreover, high compatibility between the kenaf fiber and polymer matrix is observed. However, for the SEM micrograph of UPRC_a composite (Fig. 6), the fiber is loosely embedded in the polymer matrix and bad fiber distribution. There is also evidence of fiber pull-outs during failure. For UPRC_d composite (Fig. 7), the fiber compatibility with the matrix is better than UPRC_a.

CONCLUSIONS

New bio-fiber composites UPRC_{a-d} of variable formulations according to the amount of MA were



Figure 4 Water absorption % of UPRC_{a-d}.



Figure 5 SEM micrograph of UPRC_c with magnification X = 1.00 K. The arrow shows the quality of fiber–matrix interaction.



Figure 6 SEM micrograph of UPRC_{*a*} with magnification (a) X = 300 and (b) X = 500. The arrow shows the quality of fiber–matrix interaction. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]





Figure 7 SEM micrograph of UPRC_{*d*} with magnification (a) X = 500 and (b) X = 1.00 K. The arrow shows the quality of fiber–matrix interaction. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]

prepared using solution polycondensation technique. UPRC_{a-d} were synthesized using ultraviolet curing technique in the presence of kenaf fiber mat, styrene and free radical initiator IRGACURE 1800. The tensile strength as well as the tensile toughness increased as the MA content within the bio-fiber composite were increased and reached a threshold value in $UPRC_c$ (MA 4 eq.). $UPRC_c$ showed higher gel content value more than 93%. Gel fraction increased as the amount of MA was increased up to the ratio of MG to MA was 1 : 4 (eq./eq.). The thermal behavior of UPRC_{a-d} indicated that, all the UPRC_{a-d} decomposed in two stages depended upon the nature of the unsaturated polyesters. The thermal stabilities of UPRC_{a-d} (at 10%) were in the order $UPRC_c > UPRC_d > UPRC_b > UPRC_a$. All the UPRC_{a-d} formulations had high thermal stabilities which were over 400°C. All UPRC had significant values from water uptake and thickness swelling

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ranging between 9.66% (±1.01) – 16.80% (±0.88) and 5.39% (±0.58) – 10.63% (±0.86), respectively.

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