Mechanical properties of kenaf fiber thermoplastic polyurethane-natural rubber composites

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Abstract: Thermoplastic polyurethane-natural rubber TPUR-NR composites filled with treated and untreated kenaf fiber as filler were prepared at different TPUR and NR contents. The content of kenaf fiber was maintained at 12.5 wt % and the fiber was treated with 6 % solution of sodium hydroxide (NaOH), then dried for 24 hours in 100 °C, hot blended with polymer components, pulverized and pressed. The mechanical properties of the composites such as tensile, flexural and impact strength were determined, and their dependence on NaOH treatment of kenaf fibers was investigated. The analysis using scanning electron microscope (SEM) was implemented to identify the effect of alkali treatment on the microstructure of kenaf fiber and TPUR-NR composites. An improvement of fiber surface roughness and bonding between the fiber and polymer as well as an increase in impact energy and elongation at break of the composites was observed.

Keyword: kenaf fiber treatment, NaOH, tensile, flexural, impact, morphology, fiber and matrices surface.

Właściwości mechaniczne kompozytów na osnowie termoplastycznego poliuretanu i naturalnego kauczuku z włóknami kenafu

Streszczenie: Otrzymano kompozyty na osnowie termoplastycznego poliuretanu (TPUR) i naturalnego kauczuku (NR) (o różnej zawartości TPUR i NR), napełnionej włóknami kenafu KF (12,5 % mas.). Włókna poddawano wstępnej obróbce przy użyciu 6 % roztworu wodorotlenku sodu, suszono w temp. 100 °C przez 24 h, mieszano na gorąco z komponentami polimerowymi, pulweryzowano i prasowano. Zbadano właściwości mechaniczne przy ściskaniu i rozciąganiu oraz odporność na uderzenie kompozytów zawierających modyfikowane lub niemodyfikowane włókna kenafu, a także wpływ alkalicznej obróbki włókien na właściwości kompozytu. Obserwacje mikrostruktury kompozytów prowadzono metodą skaningowej mikroskopii elektronowej. Stwierdzono, że obróbka włókien KF roztworem NaOH wpłynęła na zwiększenie chropowatości ich powierzchni i zdolności wiązania z osnową polimerową, a także na poprawę odporności na uderzenie i zwiększenie wydłużenia przy zerwaniu kompozytów zawierających modyfikowane włókna.

Słowa kluczowe: obróbka włókien kenafu, NaOH, rozciąganie, zginanie, uderzenie, morfologia, powierzchnia włókna i osnowy.

The main disadvantages of synthetic fibers compared to the natural ones are due to their non-environmentally friendly waste and cost, what creates new opportunities for researchers to develop the applications of biodegradable and inexpensive natural fibers. They can act as reinforcement or filler in the composites and are able to improve their mechanical characteristics [1–4] in certain product applications.

Some researches on strengthening composites using kenaf fiber have been done [5–8]. Thanks to specific mechanical properties and biodegradability of kenaf fibers, after adequate treatment, they can be used in combination with synthetic polymers, such as polyester or rubber, which can help to greatly reduce the fiber hydrophilicity, in the production of composite materials [9]. Kenaf short fibers were compounded with maleated polypropylene/maleated polyethylene MAPP/MAPE showing a potential as reinforcement in thermosets and thermoplastics composites [10–14]. Kenaf fibers have been treated by 6 % of sodium hydroxide to improve the tensile properties or the bonding between the fibers and rubber [10].

Natural rubber was used in engine mountings because of its combination of valuable properties, including high

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strength, outstanding fatigue resistance, high resilience, low sensitivity to strain effects in dynamic applications and good resistance to creep [15]. It was one of the elastomers that are strain crystallizing, and as such possesses inherent strength without the addition of particulate reinforcement. The features of natural rubber allow for fabrication of the materials having very low modulus, low damping and high strength [16, 17]. The rigidity of rubber mount's supporting structure, along with mount system strength and geometry, are very important considerations [18].

This study focused on determining the effect of fiber treatment using sodium hydroxide to the kenaf fiber characteristics in the TPUR-NR composites. The investigation was focused on the characterization of mechanical properties and microstructure of the composites to evaluate the interfacial adhesion between the treated/untreated kenaf fiber and polymer matrix.

EXPERIMENTAL PART

Materials

Latex rubber GIVUL MR (Table 1) was provided by Getahindus (M) Sdn. Bhd. (Tangkak, Johor, Malaysia). Kenaf fiber (KF), thermoplastic polyurethane (TPUR) Estane 58311 (Table 2) and sodium hydroxide (NaOH) were supplied by Innovative Pultrusion Sdn. Bhd. (Seremban, Malaysia). The kenaf fiber was initially sieved and only the fraction 200–300 μm was used in the study. The latex rubber was solidified first into thinned rectangle slab and cut into small cubic shape.

Sample preparation

The kenaf was soaked for 24 hours in the 6 % of sodium hydroxide solution. Then, the alkali-treated kenaf fibers were washed and dried at 100 °C for 24 hours. The mass fractions of the components used for the preparation of composite samples are shown in Table 3.

The components were blended using the hot melt blending Brabender machine [19–23]. The mixed TPUR/NR composites were pulverized into small cubic shape. The pulverized TPUR/NR composites were pressed using 40-tonne hot press machine and shaped into a sample board with a size of 3 mm × 15 cm × 15 cm. The sample board was then cut according to the dimensions of testing specimens.

Methods of testing

– The tensile tests were conducted according to ASTM D-638 [9, 23, 24] at the temperature of 22 °C and relative humidity of 59 %. The tests were carried out on four samples with four replications using a Universal Testing Machine (INSTRON 5556) with a 5 kN load cell; the crosshead speed was maintained at 50 mm/min.

T a b l e 1. Latex natural rubber (NR) – GIVUL MR specification

Properties	Unit	Specification
Total solid content	%	60.5 ± 0.50
Dry rubber content	%	NA
Non-rubber content	%	NA
Alkalinity	%	min. 0.60
Magnesium content	ppm	NA
Volatile fatty acid		NA
MST	s	min. 1100
Coagulum content	ppm	max. 100
KOH		NA
pH		10.0–11.0
Viscosity B/F LVT	cps	max. 120
Toluene swell	%	82–85

The mechanical stability is represented by the time *(MST*; s*)* from the start of agitation to the end point where a visually distinguishable aggregate begins to form; NA – not available.

T a b l e 2. TPUR – Estane 58311 specification

Physical properties	Unit	Value
Hardness (5 s)	Shore A	85 ± 3
Specific gravity		1.13
Tensile strength	MPa	45.5
Ultimate elongation	%	520
Tensile stress at:		
100 % elongation	MPa	6.2
300 % elongation	MPa	11.7
Tear strength:		
graves	kg/mm	7.0
trouser	kg/mm	2.1
T_m (by DSC)	°C	135
T_g (by DSC)	°C	-49

– Flexural tests were conducted according to ASTM D-790 [9, 24, 25] at a temperature of 22.5 °C and relative humidity of 49 %. The samples were prepared with dimensions of 130 × 13 × 3 (mm^3). The tests were carried out on five replications using a Universal Testing Machine (INSTRON 5556) with a 5 kN load cell; the crosshead speed was maintained at 50 mm/min. The support span length was set at a ratio of 16 : 1 to the thickness of samples.

– Izod impact tests were conducted according to ASTM D256 [9, 24, 26] at a temperature of 26 ± 1 °C and relative humidity of 50 ± 5 %. The notched samples were prepared with dimensions of 60 × 13 × 3 (mm^3). The tests were performed on five replications using a digital INSTRON CEAST 9050 pendulum impact tester. The impact strength (J/mm^2) was calculated based on the impact energy (J) and cross-section area (mm^2) of the specimen.

– The morphology of tensile fractured surfaces was observed using a Phenom Pro Scanning Electron Microscope model, with an acceleration voltage of 5 kV and 10 kV. These activities were conducted at UniKL-MFI

T a b l e 3. Composition of fibers and matrices

Fiber condition	Sample code	Kenaf wt %	NR wt %	TPUR wt %
Untreated fiber	1KF1NR6TPUR	12.5	12.5	75.0
Treated fiber	T1KF1NR6TPUR			
Untreated fiber	1KF2NR5TPUR	12.5	25.0	62.5
Treated fiber	T1KF2NR5TPUR			
Untreated fiber	1KF3NR4TPUR	12.5	37.5	50.0
Treated fiber	T1KF3NR4TPUR			
Untreated fiber	1KF4NR3TPUR	12.5	50.0	37.5
Treated fiber	T1KF4NR3TPUR			

laboratory, Teras Jernang, Selangor, Malaysia. The samples were divided into the treated and untreated kenaf fiber-matrix composites. Each of the samples was examined using the Phenom ProX desktop SEM available at UniKL-MFI laboratory as well.

RESULTS AND DISCUSSION

Tensile properties of treated kenaf fiber TPUR-NR composites

The dimension of the samples prepared for the testing was in good condition where the size tolerance of the sample was ± 0.5 mm. There was no obvious large bubbles or porosity in the sample board before it was cut into the required specimens. The observation of bubbles was carried out using a bright light placed under the sample board. Figure 1 shows the increase in tensile strength with increasing the content of TPUR in kenaf-TPUR/NR composites. There is a slight difference in tensile strength between the untreated and treated kenaf fiber for the high content of TPUR in composites. On the other hand, Fig. 2 shows that the elongation at break increases almost double for the composite with the highest content of TPUR and NaOH-treated kenaf fibers when compared to that containing untreated fibers. This finding is similar to that reported in [27] where the treated kenaf fibers gave higher elongation with NR compounds.

The composites with the highest rubber content show the lowest tensile strength values for both treated and untreated kenaf fibers (Fig. 1). In other study, the NaOH used as fiber treatment reduces intermolecular hydrogen bonding in kenaf cellulose to give better interfacial bonding of the fibers and rubber matrix [10]. On the other hand, despite of decrease in cellulose hydrogen bonding with increasing NaOH concentration, the composites of TPUR with alkali-treated kenaf shown a decline in mechanical properties [12].

The tensile modulus shown in Fig. 3 increases with increasing TPUR content in TPUR/NR composites, both containing untreated and alkali-treated kenaf fibers. Further increase in modulus, observed for the compos-

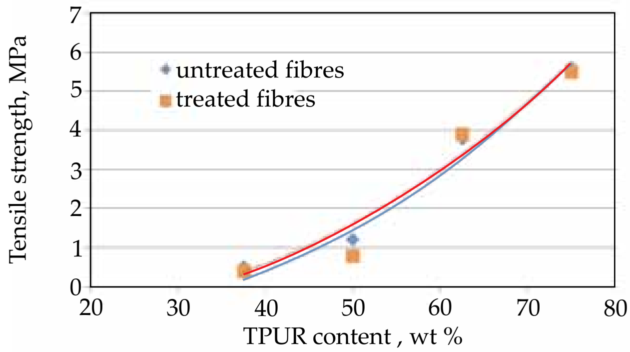

Fig. 1. Tensile properties of treated and untreated kenaf fiber TPUR-NR composites

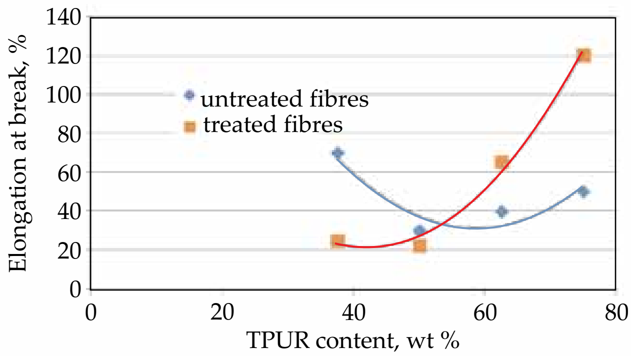

Fig. 2. Elongation properties of treated and untreated kenaf fiber TPUR-NR composites

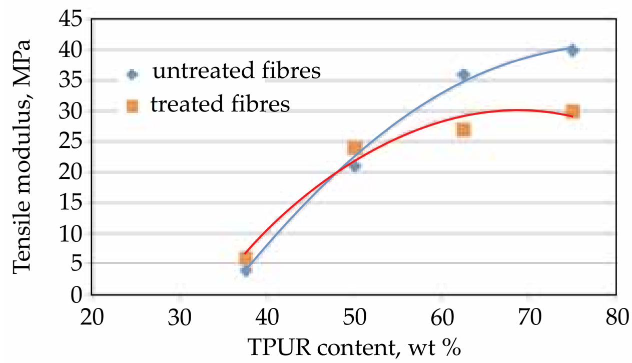

Fig. 3. Tensile modulus properties of treated and untreated kenaf fiber TPUR-NR composites

ites with NaOH-treated kenaf fibers indicates that alkali treatment results in the increased bonding strength between the fiber and polymer matrix. This finding is almost the same as in [27], where the tensile modulus increases for the treated kenaf fiber in NR compounds.

During the stretching test, at 62.5 % and 75 % of TPUR content, the specimens exhibited large measurement errors which may reflect non-hydrogen bonding between the treated kenaf fibers in the TPUR region.

Flexural properties of treated kenaf TPUR-NR composites

As shown in Fig. 4, slight difference in flexural modulus between the composites with NaOH-treated and untreated kenaf fibers is observed, especially for the specimens with higher contents of rubber. The composites with untreated fibers have higher modulus values. Meanwhile, at the highest content of TPUR in the composite, the modulus values determined for the specimens with untreated and treated fibers are almost equal.

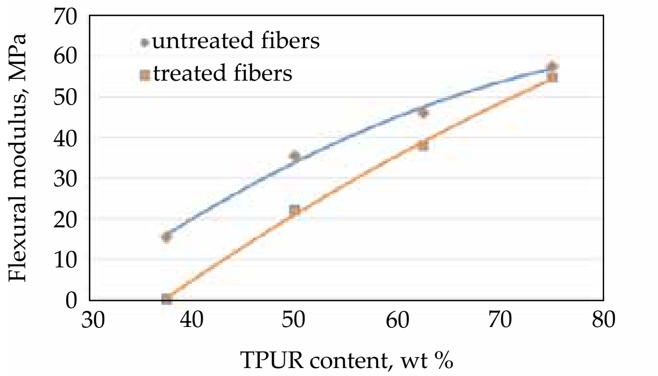

Fig. 4. Flexural modulus properties of treated and untreated kenaf fiber TPUR-NR composites

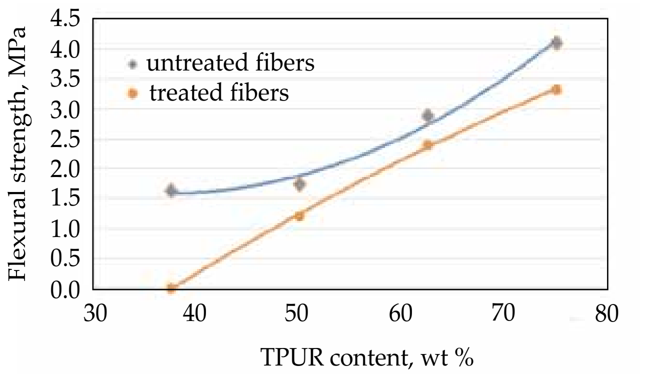

Fig. 5. Flexural strength properties of treated and untreated kenaf fiber TPUR-NR composites

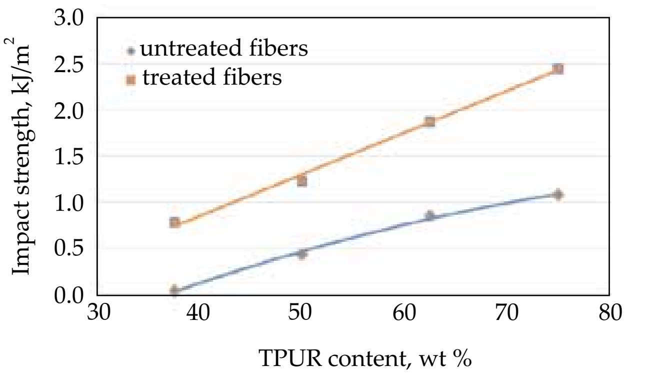

Fig. 6. Impact properties of treated and untreated kenaf fiber TPUR-NR composites

In the case of flexural strength, the higher values were obtained for the composites with untreated kenaf fibers. As it is shown in Fig. 5, a decrease in the flexural strength is observed with increasing rubber content in the composite. The similar experiment shows the inter-fibrillar matrix became soften after NaOH treatment [11].

Impact properties of treated kenaf TPUR/NR composites

The treatment of kenaf fibers with NaOH gives an excellent result of increasing the impact strength of TPUR/NR kenaf composites by up to 127 %. An increase in impact strength is more pronounced with increasing TPUR content in the composite.

Several studies reported the improved impact properties of kenaf polymer composites [28–30]. The results of flexural tests of TPUR/NR composites indicate that the treated kenaf fiber has become softer. In effect, it would give a cushion effect in TPUR/NR composites and act as energy absorbent. This gives better impact results compared to the composites with untreated fibers as shown in Fig. 6.

SEM observation of treated kenaf TPUR/NR composites

The morphology study was concentrated on the samples of treated and untreated kenaf fiber TPUR/NR composites with highest results of tensile and impact tests. In this study, the 1KF1NR6TPUR and T1KF1NR6TPUR kenaf composites were chosen. The purpose of this observation was to investigate the effect of fiber interfacial bonding on the composite properties. The cross-sections of broken samples after the tensile tests were examined in a scanning electron microscope. The content of the kenaf fiber was fixed at 12.5 wt % for every TPUR/NR composite.

As is illustrated in Fig. 7, the kenaf fiber are equally distributed in the TPUR/NR composites. No agglomeration of fibers is observed because in this study only short kenaf fibers were used. This indicates that the mixing process using an internal mixer and hot press machine was carried out at a suitable temperature and pressure [9]. In Fig. 8 it is clearly shown that the fiber break obviously occurred in the treated kenaf fiber polymer composites. For the untreated kenaf fibers many fiber pull-outs can be observed. This shows that the NaOH treatment of the kenaf fibers has a positive influence on the bonding between the fiber and the polymer matrix. According to other studies [27, 31] alkali treatment cleans kenaf fiber surface and may improve fiber-matrix adhesion.

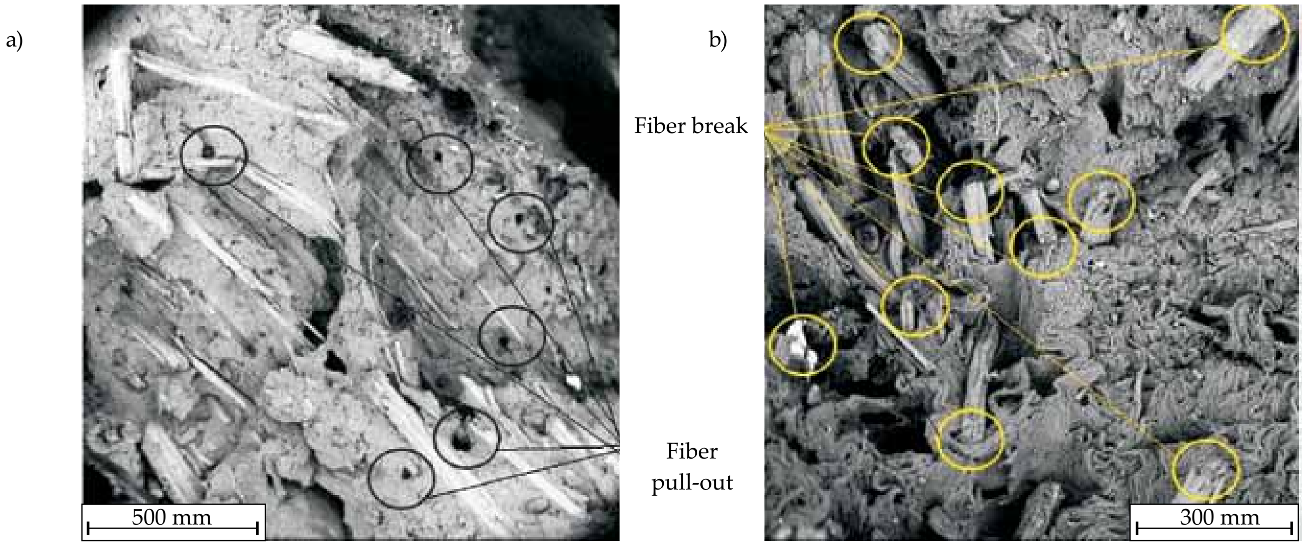

Fig. 7. Fiber distribution and surface fracture of TPUR/NR composites: a) untreated kenaf fiber, b) treated kenaf fiber

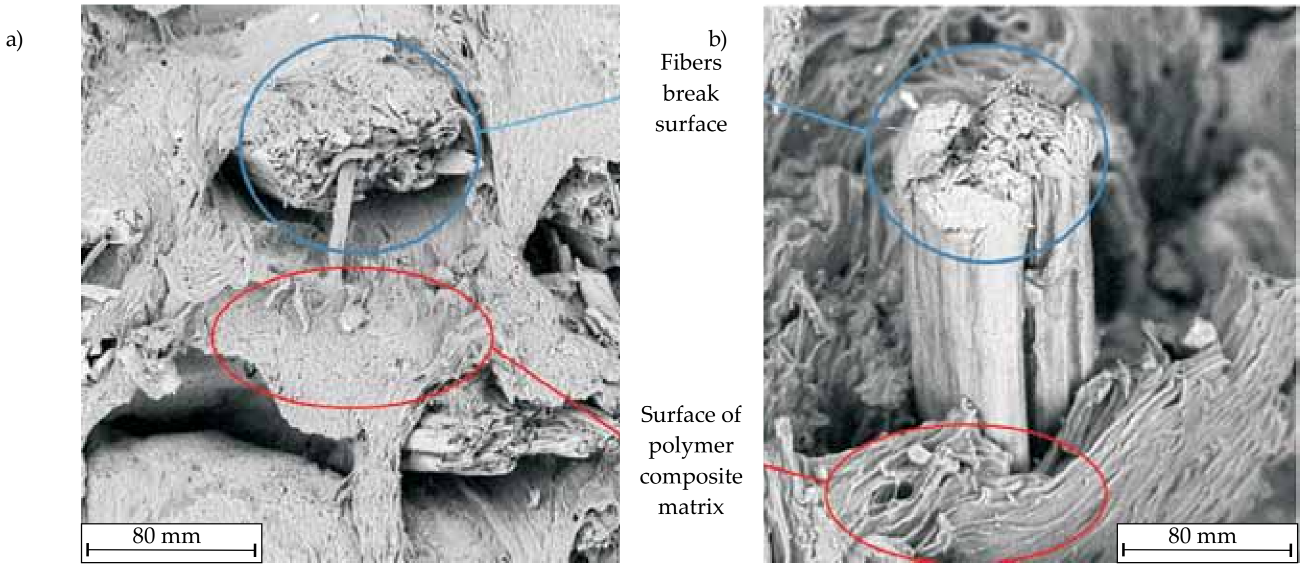

Fig. 8. Fiber and composite matrix surface: a) untreated kenaf fiber – 1KF1NR6TPUR, b) treated kenaf fiber – T1KF1NR6TPUR

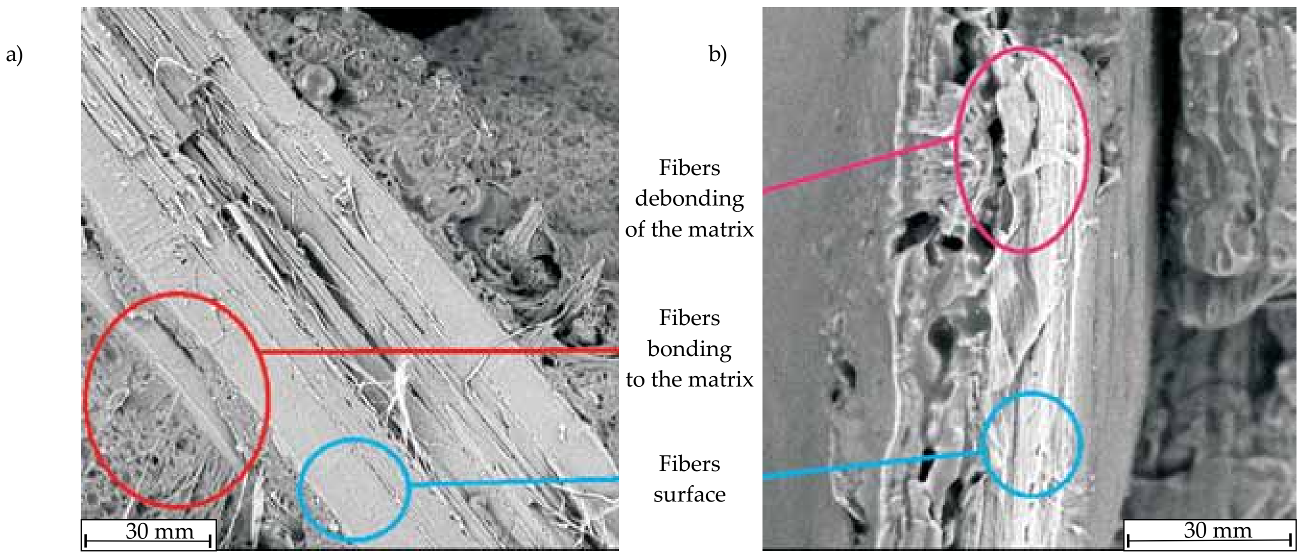

Fig. 9. Interfacial fiber adhesion to the polymer matrix: a) untreated kenaf fiber-matrix – 1KF1NR6TPUR, b) treated kenaf fiber--matrix – T1KF1NR6TPUR

Examples of fiber bonding and debonding in the TPU/NR composites are presented in Fig. 9. The untreated fibers show flat surface, while those subjected to NaOH treatment are roughly surfaced, what is clearly shown in Fig. 8, illustrating the surface of broken fibers. In works of other researchers this increased roughness results in a better wetting and impregnation of the fibers thus increasing their bonding with matrix [32, 33].

The surface of the TPUR/NR matrix compound is smooth in the specimens with untreated kenaf fibers (Fig. 8a), comparing to the rough matrix surface in the composites with a alkali-treated kenaf. The increased bonding between the fibers and matrix in kenaf-filled TPUR/NR compounds also contributes to their improved impact properties.

CONCLUSIONS

The effect of NaOH as a treatment on kenaf fiber used in TPUR/NR composites on their mechanical properties was investigated. A significant improvement in the impact characteristics (increase in impact strength by up to 127 %) was observed for the composites with alkali-treated kenaf fibers in comparison with those containing untreated fibers.

The elongation at break increases with the increasing content of TPUR in TPUR/NR composites with the treated kenaf fiber. On the other hand, the treatment of kenaf fiber contributes to a decrease in flexural modulus of TPUR/NR composite. Using SEM observation, it was verified that NaOH treatment applied to the kenaf fibers may, in some conditions, improve the interfacial bonding between fiber and TPUR-NR matrices.

SEM analysis showed that surfaces of alkali-treated kenaf fibers were rough, which might result in improved wettability and bonding properties.

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