IMPROVEMENT OF MECHANICAL PROPERTIES OF RECYCLED HIGH DENSITY POLYETHYLENE/NATURAL RUBBER/KENAF POWDER BIOCOMPOSITES BY USING MALEATED NATURAL RUBBER AS A COUPLING AGENT

X. Viet Cao[1], H. Ismail[1,*], A. A. Rashid[1], T. Takeichi[2], T. Vo-Huu[3]

[1]*School of Materials & Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, Malaysia*
[2]*Department of Environmental and Life Sciences, Toyohashi University of Technology, Japan*
[3]*Department of Polymer Materials, Ho Chi Minh University of Technology, Viet Nam*

Kenaf powder was incorporated into recycled high density polyethylene (rHDPE)/natural rubber (NR) blend using an internal mixer at 165 °C and rotor speed of 50 rpm. The rHDPE/NR ratio was fixed at 70/30. The tensile strength and elongation at break of the composites decreased, while the tensile modulus increased with increasing filler content. The maleic anhydride grafted natural rubber (MANR) was prepared and used as an effective coupling agent. The addition of MANR improved the tensile properties of rHDPE/NR/KP biocomposites. Scanning electron microscopy of the tensile fracture surface of the composites showed a better filler-matrix adhesion which was an evidence for the enhanced tensile properties in the presence of MANR.

Keywords: Kenaf; Recycled High Density Polyethylene; Natural Rubber

INTRODUCTION

The development of polymer composites using recycled or recyclable polymers and natural organic fillers is very imperative due to threats of uncertain petroleum supply in the near future and environmental concerns.

Thermoplastic elastomers (TPEs) are promoted for their attractive elastic properties at room temperature, their flowability at high temperature, and remelting for recycling use [1]. Recently, kenaf is gaining a lot of attention in the composite industry, since they can be applied as filler in polymer composites. It offers several advantages including low density, low cost, relative non-abrasiveness, high filling levels, recyclability, biodegradability, and renewable nature [2].

The mechanical properties of TPEs composites are dependent strongly upon dispersion of the filler in the matrices and the adhesion at the filler/matrix interface. Because of the hydrophilic nature, the filler does not wet or interact with hydrophobic polymers due to the difference in surface energies [3]. It is necessary to use coupling agent to enhance the compatibility between fiber and matrix.

In the present work, maleated natural rubber (MANR) was prepared and used as a coupling agent for kenaf core powder filled rHDPE/NR biocomposites. Mechanical improvement of the treated bicomposites was presented.

MATERIALS AND METHODS

Materials

Recycled high density polyethylene (rHDPE) was obtained from Zarm Scientific and Supplies Sdn Bhd, Penang. Natural rubber used was SMR L grade from the Rubber Research Institute of Malaysia (RRIM). Maleic anhydride (MAH) was supplied by Sigma Aldrich. Kenaf powder was produced by grinding kenaf core in a table-type pulverizing machine and sieved to obtain the powder size in range of 32 to 150 μm.

Preparation and Characterization of rHDPE/NR/KP Biocomposites

Maleic anhydride grafted natural rubber (MANR) was prepared in an internal mixer at a temperature of 135 °C for 10 min and a rotor speed of 60 rpm. The maleic anhydride content used in this study was 6 phr.

Formulation of rHDPE/NR/KP biocomposites is given in Table 1. Mixing process was carried out at 165 °C and a rotor speed of 50 rpm for 12 min. The blends were then compression molded in a hydraulic hot press into 1mm sheets for preparing test samples. FTIR (Perkin Elmer System 2000) was used to confirm the grafting reaction between NR and MAH. Sample was extracted the unreated MAH by Soxhlet extraction in acetone for 24 hrs, and further dried in a vacuum oven at 40 °C for 24 hrs prior to FTIR measurement.

* *Corresponding author: Tel. + 6 (04) 59961136*
Email: hanafi@eng.usm.my

The tensile properties were measured using an Instron 3366 machine at a cross-head speed of 50 mm/min at 25 ± 3°C according to ASTM D 412. Tensile strength, tensile modulus, and elongation at break of the each sample were obtained from the average of five specimens.

The tensile fractured surface of the composites was also analyzed with a Supra-35VP field emission scanning electron microscope (SEM). The samples were coated with gold/palladium by a sputter coating instrument (Bio-Rad Polaron Division) for 45 minutes to prevent electrostatic charging during evaluation.

Table 1. Formation of rHDPE/NR/KP biocomposites*

Materials	Composition (phr)
Recycled high density polyethylene (rHDPE)	70
Natural rubber (SMR L)	30
Kenaf powder (KP)	0, 10, 20, 30, 40
MANR[a]	5

Note: (phr)-part per hundred resin

[a] 5% of KP

* Similar biocomposites but without MANR were also prepared.

RESULTS AND DISCUSSION

FTIR Analysis

The FTIR spectra of NR and MANR are shown in Fig.1. The peaks at 1662 cm^{-1} and 833 cm^{-1} were corresponded to C=C of NR and observed for both cases. For the MANR spectrum, the absence of the absorption peak at 698 cm^{-1} suggested the unreacted MAH has been completely removed from the MANR. A broad and intense characteristic peak at 1779 cm^{-1} and a weak absorption peak at ca 1862 cm^{-1} were observed. These peaks were assigned to grafted anhydride, which are due to symmetric (strong) and asymmetric (weak) C=O stretching vibrations of succinic anhydride rings, respectively[4, 5]. Therefore, it can be confirmed that the MAH was successfully grafted onto NR backbone.

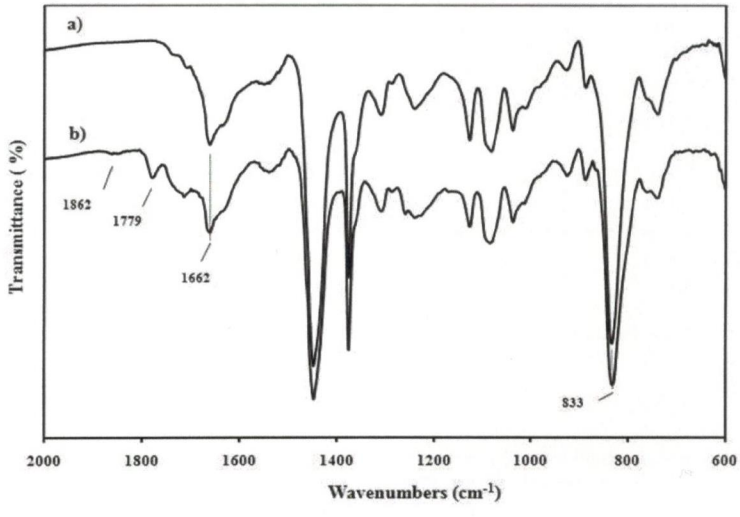

Fig. 1. FTIR spectra of NR (a) and MANR (b).

Tensile Properties

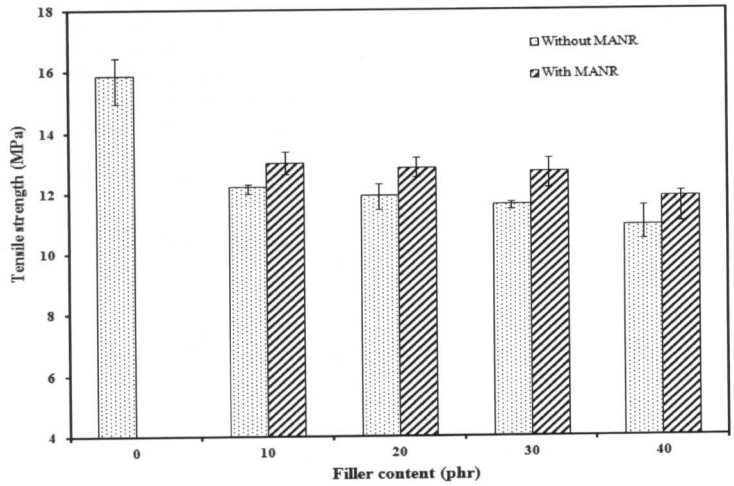

Fig. 2. Tensile strength of rHDPE/NR/KP biocomposites at various filler content.

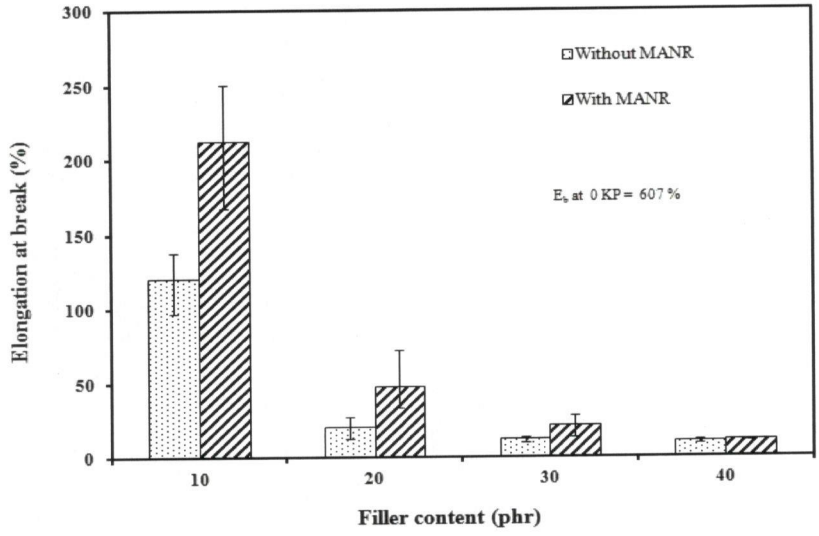

Fig. 3. Elongation at break of rHDPE/NR/KP biocomposites at various filler content.

As shown in Fig. 2 and Fig. 3, tensile strength and elongation at break of rHDPE/NR/KP biocomposites decreased gradually with increasing filler content due to the weak interfacial bonding between the hydrophilic filler and the hydrophobic polymer matrixes [3]. However, the addition of MANR as coupling agent enhanced the composites properties. MANR improved interfacial adhesion between KP and matrix by forming hydrogen bonding between KP and MANR. The better interfacial bonding also prevented fiber-fiber contact, hence gave the better filler dispersion. As a result, tensile strength and elongation at break increased with the addition of MANR. This was also responsible for the higher tensile modulus for composites with MANR (Fig. 4).

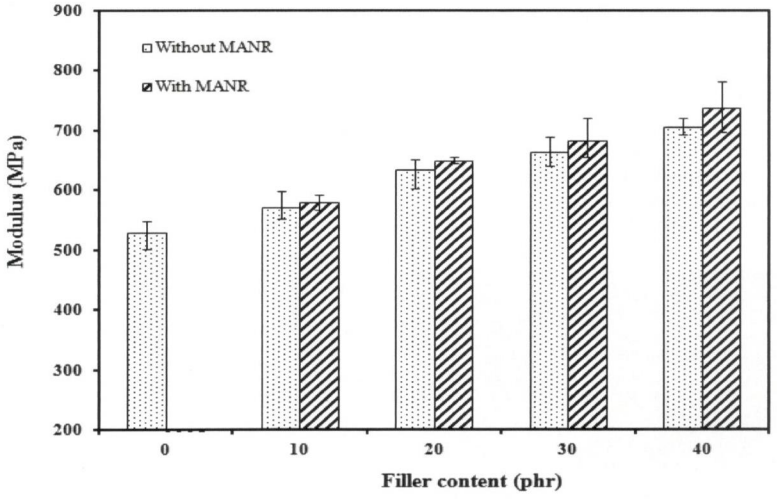

Fig. 4. Tensile modulus of rHDPE/NR/KP biocomposites at various filler content

Morphology

Tensile fracture surfaces of the biocomposites with and without MANR at 10 and 40 phr of KP are shown in Fig. 5. In the case of composites without MANR, the fibers were not well oriented and the poor adhesion at the interface can be deduced from the clean surface of the fibers. This poor adhesion is clearly visible at 40 phr, where some fibers were pulled out or remained loosely with the matrix. Voids were also presented in the samples.

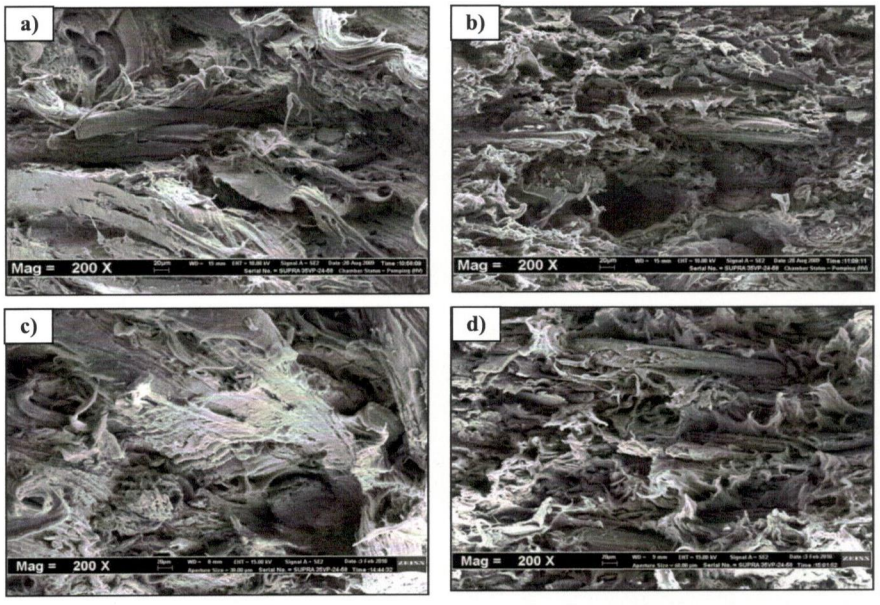

Fig. 5. Tensile fracture surfaces of rHDPE/NR/KP biocomposites at 200X: (a) 10 phr, (b) 40 phr without MANR, (c) 10 phr and (d) 40 phr with MANR.

The addition of MANR significantly improved adhesion to the fiber. Fig. 5 (c) and (d) shows the microstructure of KP filled rHDPE/NR composites with MANR at 10 phr and 40phr, respectively. The morphology was clearly different compared to the composites without MANR. All the micrographs of the composites with MANR also showed better dispersed fillers compared to the composites without MANR. The better fiber-matrix adhesion can be measured by the fact that more matrix fibrillation, rougher fracture surfaces, and less fiber pull out were observed. Interestingly, the improvement of the adhesion at the interface was still obvious at 30 phr by looking at a good bonding between KP fiber and matrix (Fig. 6). The KP fiber was coated and there were NR matrix tearing bridges between fiber and matrices.

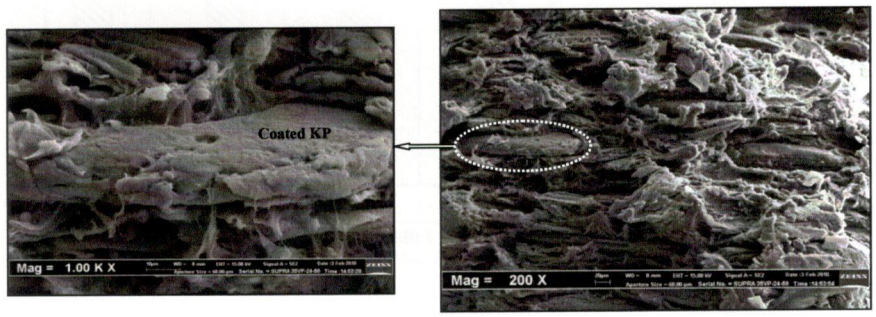

Fig. 6. Tensile fracture surfaces of rHDPE/NR/KP biocomposites at 30 phr of KP with MANR.

CONCLUSIONS

Maleated natural rubber was prepared and used in this study to improve the interfacial adhesion between hydrophilic kenaf powder and the rHDPE/NR hydrophobic matrices. The SEM micrographs showed the better adhesion at the fiber-matrix interfaces as MANR was added to the composites. rHDPE/NR/KP biocomposites with MANR provided an enhancement in tensile strength, tensile modulus, and elongation at break compared to the biocomposites without MANR.

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