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SHORT COMMUNICATIONS

Effects of Clay and LNR on Mechanical Properties and Morphology of NR/HDPE-Aramid Composites

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Various fibers have been incorporated with natural and synthetic rubbers.[1–3] However, few studies on fiber reinforced thermoplastic natural rubber (TPNR) were found. Therefore this study tried to ascertain the advantages of reinforced TPNR blends. Aramid fibers have become important materials due to the fibers' excellent mechanical, physical and chemical properties, *i.e.* high strength and modulus, high-energy absorption and good chemical resistance. In order to improve aramid-rubber interaction, these fibers have been treated with resorcinol formaldehyde latex.[4] Although the composite materials based on aramid-reinforced polymer produced excellent mechanical properties, the price concerned still has the challenge in mass production.

Hybrid composites are materials made by combining two or more different types of fillers in a common matrix. By careful selection of fillers, the material costs can be substantially reduced.[5] To get the optimum balance of technical properties and to reduce the price of the product, aramid fiber is partially replaced with clay or kaolinite. Clay is readily available and a cost-effective mineral filler. In the process of mixing the clay in the molten polymer, the nature of the clay (*i.e.* the filler-matrix interactions) affects the structure of the obtained polymer-clay composite: either a micro-composite, an intercalated nanocomposite or an exfoliated nanocomposite. When particle fillers are incorporated into polymers, stiffness is generally improved but strength, elongation and impact properties normally deteriorate. However, it has been found that clay can also be acted as reinforcing filler in polymer matrix if the particle size is small especially at low filler loading. Previous research by Sheng *et al.*[6] found that the incorporation of clay nanofiller enhances the tensile properties of nylon 66 matrix. Their research is consistent with Kojima's finding on nylon 6/clay nanocomposites.[7] They reported that the improvement of mechanical properties was due to the higher parity between the matrix surface and clay leads to platelet exfoliation of clay among the polymer matrix. In addition, the mechanical properties improved could also be due to the presence of Si–O and Si–OH groups on its surface.[8,9]

In this study, the optimum fiber loading in NR:HDPE (50:50) blend was determined. The effect of partial replacement of aramid fiber by clay on the mechanical properties of the composites with and without liquid natural rubber (LNR) as compatibilizer was also investigated.

EXPERIMENTAL

Materials

The materials used in this study were SMR-L grade natural rubber (NR) from the Rubber Research Institute of Malaysia (RRIM), high-density polyethylene (HDPE) from Polyethylene Malaysia with density 0.94 g cm^{-3}. Aramid short fibers (3 mm long) were supplied by Teijin-Twaron, The Nethelands under the trade name of Twaron 1588 (Twaron). The fiber had already been treated with resorcinol formaldehyde latex (RFL) for adhering to rubber. The refined clay (kaolin) used is a hydrated aluminosilicate with average particle size of 10 μm and density of 2.5 g cm^{-3} was supplied by Kaolin (M) Sdn. Bhd. The LNR was generated by photochemical oxidation of NR in our laboratory.[10]

Composite Preparation

Composites were prepared in an internal mixer (Haake Rheomix CTW 100p) at 155 °C, 50 r.p.m. for 12 min. The compositions of composites were 50/50 NR/HDPE ratios (v/v%) with the variation of fiber

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Table I. Composition of the studied materials

NR/HDPE/LNR	Twaron (%)	Clay (%)
50/50/0	20.0	0.0
	17.5	2.5
	15.0	5.0
	12.5	7.5
	10.0	10.0
40/50/10	20.0	0.0
	17.5	2.5
	15.0	5.0
	12.5	7.5
	10.0	10.0

between 0 to 30% fiber loading. For the preparation of hybrid composites, clay and liquid natural rubber were added in the composites. Table I shows the formulation for hybrid composites used throughout in this study.

Tensile properties were measured using Testometrix M350 according to ASTM D412. The gauge was kept at 20 mm with crosshead speed of 50 mm min^{-1}. The average stress and tensile modulus were calculated from at least 8 samples. The impact strength was measured by an Izod impact tester at room temperature using a digital Universal Fractoscope CEAST 6545/000 model for the average of 8 samples. All measurements were performed at room temperature (27 °C).

RESULTS AND DISCUSSION

Mechanical Properties

The effect of Twaron loading on the tensile strength and tensile modulus as well as the impact strength of NR/HDPE blends is shown in Table II. It can be seen that the tensile strength of the composites increases with increasing volume fraction of Twaron and the optimum fiber loading in the system is 20%. Tensile strength increased is probably due to the uniform fiber cross-section and to relatively high aspect ratio (length to diameter ratio l/d). The capability to support transmitted stress from the matrix is high. It is also because of the fibers itself have a high strength. It can also be deduced from Table II that the Young's modulus increased rapidly with fiber content in the composites. The increase in tensile modulus also indicates the ability of Twaron to impart greater stiffness to the NR/HDPE matrix. A similar observation can be seen in Table II for the effects of fiber loading on impact strength. Since Twaron is ductile fiber and TPNR has high impact strength, NR/HDPE reinforced by Twaron required more strength and energy to break. The increase of impact strength also showed that the adhesion between the fiber and the matrix is significant.

Table II. Effect of Twaron loading on mechanical properties of 50/50 NR/HDPE

Twaron loading (%)	Tensile strength (MPa)	Tensile modulus (MPa)	Impact Strength (kJ mm^{-2})
0	4.9	7.3	13.2
5	5.6	13.0	15.1
10	7.2	21.6	18.3
15	9.2	28.0	20.0
20	10.7	32.9	23.6
25	10.0	29.4	21.5
30	8.4	23.6	17.8

Figures 1 to 3 show the effect of partial replacement of Twaron by clay in the NR/HDPE blend with and without LNR on the mechanical properties. It can be seen at 0% clay in Figure 1 that the tensile strength increases with the addition of LNR in the blend. An improvement of tensile strength strongly indicated the existence of an interaction between NR and HDPE phase in the presence of LNR. It is believed the active groups present on LNR like –OH, –OOH and –OR

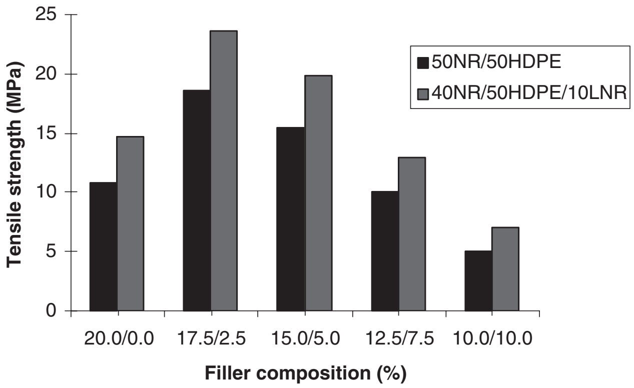

Figure 1. Effect of filler loading on tensile strength of partial Twaron/clay composites.

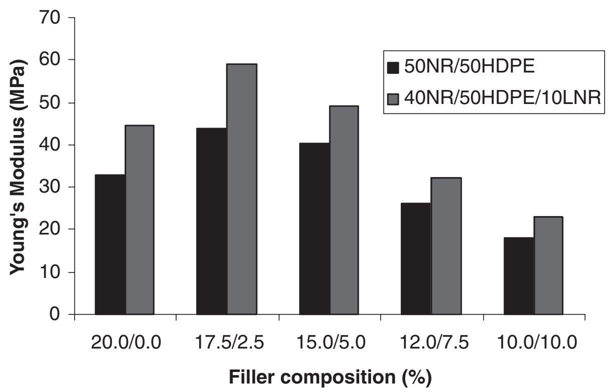

Figure 2. Effect of filler loading on Young's modulus of partial Twaron/clay composites.

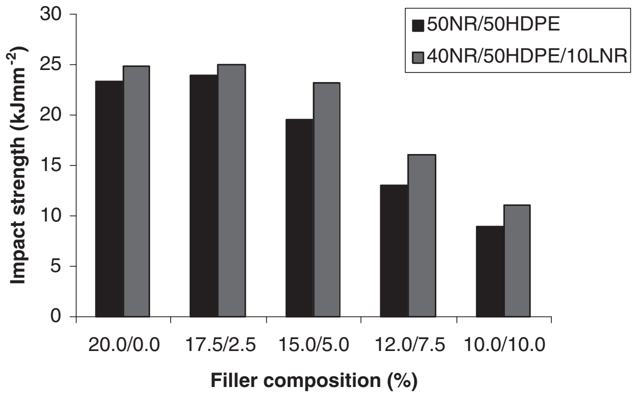

Figure 3. Effect of filler loading on impact strength of partial Twaron/clay composites.

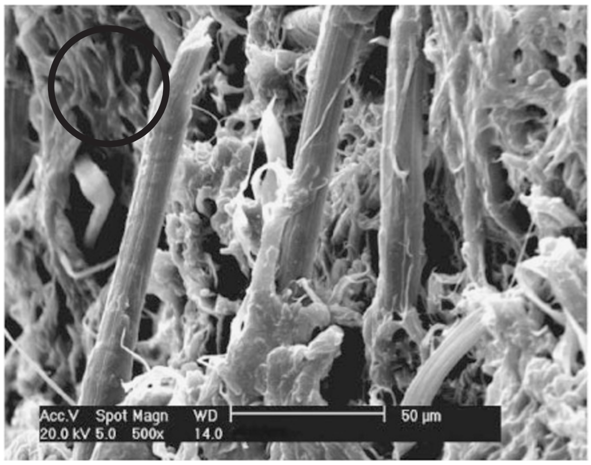

Figure 4. Scanning electron micrograph of tensile fracture surfaces of 20% Twaron filled NR/HDPE.

improve the matrix-matrix and fiber-matrix interactions.[11] Figure 1 also shows that the tensile strength increases at the small amount of clay especially in the system with LNR. An improvement can be clearly seen from 0% clay to 2.5% clay. The tensile strength increased from 10.7 to 18.6 MPa for the system without LNR and from 14.7 to 23.6 MPa for the system with LNR. The increase of tensile strength could be due to the only small amount of clay particles in the system, which led to enhance the adhesion between filler and matrix. The role of LNR in the system is as an interphase binder between the matrix and the fillers. By the introduction of LNR, the surface interaction and the particle distribution are improved. This indicates that clay in NR/HDPE/Twaron composites produce a good filler particle distribution and better particle wettability. Nevertheless, higher filler loading increase the agglomeration of clay particles causing a reduction in tensile strength.

The effect of partially replacement of Twaron by clay on Young's modulus of the composites is illustrated in Figure 2. As can be seen, Young's modulus at 0 to 2.5% clay increases especially in the system with LNR. The observation indicates that the incorporation of filler will improve the stiffness of the composites. At higher clay loading in the composites, the existence of filler agglomerates and its pull-out from matrix have reduced the Young's modulus.[12] Figure 2 also showed that 10%/10% Twaron/clay loading has lower Young's modulus than the composite at 10% Twaron loading without clay. The reduction of Young's modulus is also due to large particle size and low interaction between inert filler and the matrix.

Figure 3 shows that there is no significant difference on impact strength at the small amount of clay then dropped sharply as the filler content was further increased in both systems. When the crack propagation is initiated through filler particle in the composite, filler can absorb energy if filler-matrix interaction is strong enough and debonding process occurs at the interface. Clay is a layered particle and therefore will promote the rigidity of the composite. This led to the reduction of impact strength with clay loading. The impact strength reduced with further increase of clay also due to their highly porous structure or agglomeration. The result therefore indicates that although better distribution of clay particle in the matrix occurred with the introducing of LNR, the interaction of clay-matrix is weaker than Twaron-matrix.

SEM Micrograph

Figure 4 shows the SEM micrograph of Twaron in NR/HDPE blend. It can be seen that the matrix adhering to the fiber surface after the fiber was extracted from the matrix. The appearance of the matrix at the fiber surface suggests that an improvement in adhesion was obtained. It can also be seen very clear in Figure 4 that the microvoid formation in the matrix. It is believed that the mechanical properties can be improved by reducing the microvoid in the composites. Figure 5 shows the fractured surface at 5% clay in the composite with 10% LNR. It is clear that fibrillation and cavitation is operative. Moreover, there is some evidence of polymer-clay particle interaction in its central part. Figure 5 also showed that the domain size of the dispersed phase and the microvoid formation reduced by the introducing of clay especially for the composite with 10% LNR.

In general, as the load is applied to the composite, the stress is transfer from the fiber to the matrix *via* interface. It is often neglected the point of stress concentration *i.e.* air bubbles or microvoid especially if the current level of stress is not high enough compared to the fiber strength. In fact, this factor is also important in obtaining the actual strength. By introducing the small amount of clay, the microvoid formation

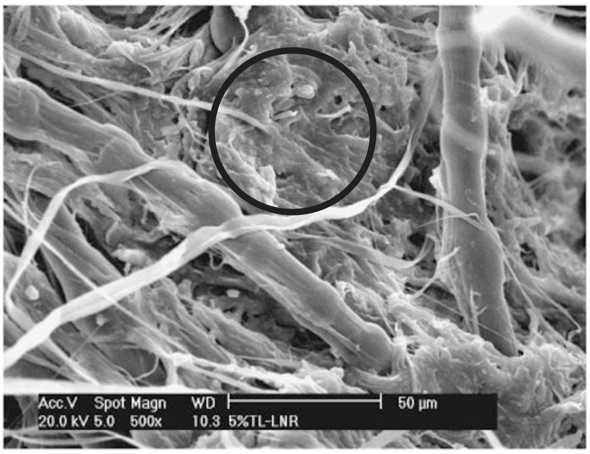

Figure 5. Scanning electron micrograph of tensile fracture surfaces of partial Twaron/clay composites with 10% LNR (15% Twaron/5% clay).

can be reduced especially if the size of clay is very small and the dispersion is good. This shows the role of LNR in the system. Nevertheless if the amount of clay is too high the function of the filler as reinforcing agent is less important due to agglomeration and low aspect ratio compared to Twaron.

CONCLUSIONS

Partial replacement of Twaron by clay at certain percentage, have improved the mechanical properties and better surface morphology of the composites especially with the 10% LNR. This enables to use Twaron partially with clay in the composites in order to improve the mechanical properties and reduce the cost of composites material.

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