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## International Journal of Polymeric Materials

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713647664>

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**To cite this Article** Ismail, Hanafi , Nizam, J. M. and Khalil, H. P. S. Abdul(2011) 'White Rice Husk Ash Filled Natural Rubber/Linear Low Density Polyethylene Blends', International Journal of Polymeric Materials, 48: 4, 461 – 475

**To link to this Article:** DOI: 10.1080/00914030108048842

**URL:** <http://dx.doi.org/10.1080/00914030108048842>

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# White Rice Husk Ash Filled Natural Rubber/Linear Low Density Polyethylene Blends

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*(Received 22 November 1999; in final form 1 December 1999)*

Natural rubber (NR)/Linear low density polyethylene (LLDPE) blends were prepared using an internal mixer at 150°C and a rotor speed of 55 rpm. The tensile strength, tensile modulus and hardness increase with increasing LLDPE content whereas elongation at break and mass swell show decreasing trend. With 30/70 (W:W) NR/LLDPE blends, the increasing white rice husk ash (WRHA) loading also increases the tensile modulus and hardness but reduced the tensile strength, elongation at break, and mass swell.

**Keywords:** Natural rubber; Linear low density polyethylene; Blend; Mechanical properties; Mass swell

## 1. INTRODUCTION

In recent years, elastomeric rubber – plastic blends have become technologically important for use as thermoplastic elastomers [1]. Thermoplastic elastomers are materials which can be moulded like plastics at appropriate processing temperatures, but which retain some of the flexibility and resilience of rubbers at normal temperatures [2]. The study on thermoplastic elastomer with various types of elastomer and polyolefins have been reported by many researchers [3–6].

Burnt rice husk ash yield two grades of fillers, namely white rice husk ash (WRHA) and black rice husk ash (BRHA). WRHA has been

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analysed to have about 96% silica content, while the BRHA has a lower silica content, typically about 54% and a substantial carbon content, *i.e.*, about 44%. It has been shown that with WRHA [7], apart from exhibiting better physical properties than BRHA, the properties approach those of commercial carbon black. In our previous works [8–10], we have reported the use of WRHA as a filler for natural rubber compounds. The mechanical properties *viz* tensile and tear strengths were enhanced by the addition of WRHA. The optimum level of WRHA to obtain maximum mechanical properties was achieved at 20 phr after which there was deterioration in properties. The presence of silane coupling agent,  $\gamma$ -mercaptopropyl trimethoxy silane (A-189) and Si69 has increased the effectiveness of WRHA as a filler for natural rubber compounds.

In this paper the use of WRHA in natural rubber (NR)/linear low density polyethylene (LLDPE) blends were carried out. The first part of this work was focussed on the effect of LLDPE content on mechanical properties of NR/LLDPE blends. The second part was carried out to determine the effect of WRHA loading on mechanical properties of LLDPE blends. Oil resistance of this blends was also examined.

## 2. EXPERIMENTAL

### 2.1. Materials

SMR-L grade natural rubber was obtained from Rubber Research Institute of Malaysia (RRIM). The filler used was WRHA, obtained from Plastic Technology Centre, SIRIM (M) Ltd. Its characteristics are as follows: Mean particle size 5.4  $\mu\text{m}$ ; surface area 1.4  $\text{m}^2/\text{g}$  and density 2.2  $\text{g}/\text{m}^3$ . Linear low density polyethylene (LLDPE), Etilinas LL0209SA grade was obtained from Polyethylene (M) Sdn Bhd.

### 2.2. Compounding and Processing

The blends of NR and LLDPE were prepared in a Haake Rheodrive 5000 (drive unit) and a Haake Rheomix 600 with a roller blade (mixer) at a temperature of 150°C and a rotor speed of 55 rpm for about 13 min. Table I shows the composition of the blends for the first part of this study whereas Table II shows the different loading of WRHA

TABLE I Composition of the blends

<i>Blend component</i>	<i>A</i>	<i>B</i>	<i>C</i>	<i>D</i>	<i>E</i>
Natural rubber (SMR L)	70	60	50	40	30
LLDPE	30	40	50	60	70

TABLE II Different loadings of WRHA in NR/LLDPE blends

<i>Blend component</i>	<i>A</i> ( <i>control</i> )	<i>F</i>	<i>G</i>	<i>H</i>	<i>I</i>
Natural rubber (SMR L)	30	30	30	30	30
LLDPE	70	70	70	70	70
WRHA	–	15	30	45	60

in NR/LLDPE blends at 70/30 blend ratio (wt/wt). At the end of 13 minutes, the blend was taken out and sheeted through a laboratory mill at 2.0 mm nip setting. Samples of the blends were compression moulded in an electrically heated hydraulic press. Hot-press procedures involved preheating at 100°C, followed by heating at 150°C, and subsequent cooling under pressure. The total moulding time was 15 min.

### 2.3. Testing

Tensile tests were carried out according to ASTM D412 on a Monsanto tensometer T10. Dumbbell specimens of 2 mm thick were cut from the molded sheets with a Wallace die cutter. A crosshead speed of 50 cm/min was used and the test were performed at  $25 \pm 3^\circ\text{C}$ . Three specimens were used and the average was calculated in each case. ASTM Shore A was used to measure hardness of the blends according to ISO 48 (1979).

### 2.4. Oil Resistance

The test was conducted according to ASTM D471. The test specimens were immersed in ASTM No. 3 oil at room temperature for 70 h. The test specimens were then removed from the oil, wiped with tissue paper to remove excess oil from the surface, and weighed.

The percentage mass swell was then calculated as follows:

$$\text{Change in mass} = \frac{W_2 - W_1}{W_2} \times 100 \quad (1)$$

where  $W_1$  and  $W_2$  are the weights of the samples before and after immersion respectively.

## 2.5. Scanning Electron Microscopy

Examination of the fracture surface was carried out using a scanning electron microscope model Leica Cambridge S-360. The objective was to study the mode of fracture. All the surfaces were examined after first sputter coating with gold to avoid electrostatic charging and poor image resolution.

## 3. RESULTS AND DISCUSSION

### 3.1. The Effect of LLDPE Content on Mechanical Properties of NR/LLDPE Blends

Figures 1 and 2 show the effect of LLDPE content on tensile strength and tensile modulus,  $M_{100}$  (modulus at 100% elongation) of the NR/LLDPE blends. It can be seen that as the LLDPE content increases the tensile strength and tensile modulus increase as a result of increasing blends rigidity. The increasing of LLDPE content in NR/LLDPE blends also decrease the elongation at break,  $E_b$  (Fig. 3). This is a common observation since many researchers [11–12] also reported the decreasing of elongation at break with addition of polyolefin content into the rubber matrix. With increase LLDPE content, the stiffness and brittleness of the blends increased gradually with an associated decrease in the elongation at break. Figure 4 also shows that the hardness of the blend increased with increasing LLDPE content. This result is expected because as more LLDPE is incorporated into the rubber matrix, the plasticity of the rubber chain is reduced, resulting in more rigid blends.

### 3.2. Oil Resistance

One of the important features of polymeric materials is their ability to dissolve molecules of gas, vapour, liquid, and sometimes solute

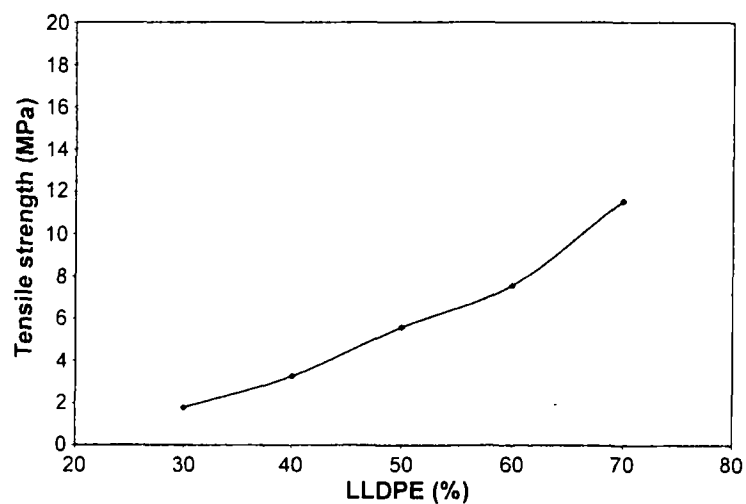


FIGURE 1 The effect of LLDPE content on the tensile strength of NR/LLDPE blends.

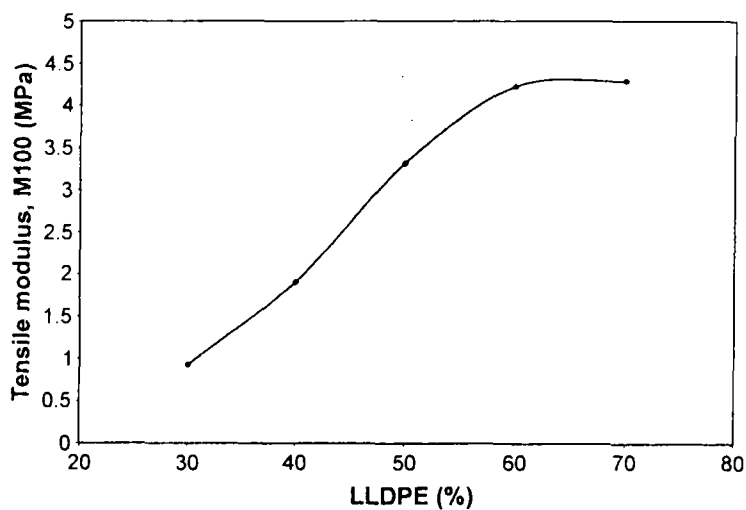


FIGURE 2 Variation of tensile modulus with LLDPE content for NR/LLDPE blends.

and to allow the transport of these molecules through the solid phase [13]. The property of transport has many practical applications, such as the use of polymers as barrier layers, cable coatings, food packings, and encapsulation of electronic circuit [14]. The phenomenon of transport through rubbery polymers is controlled by

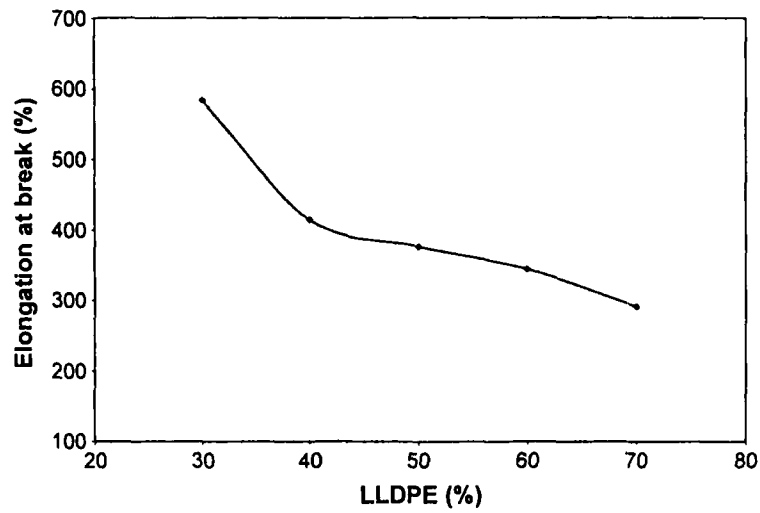
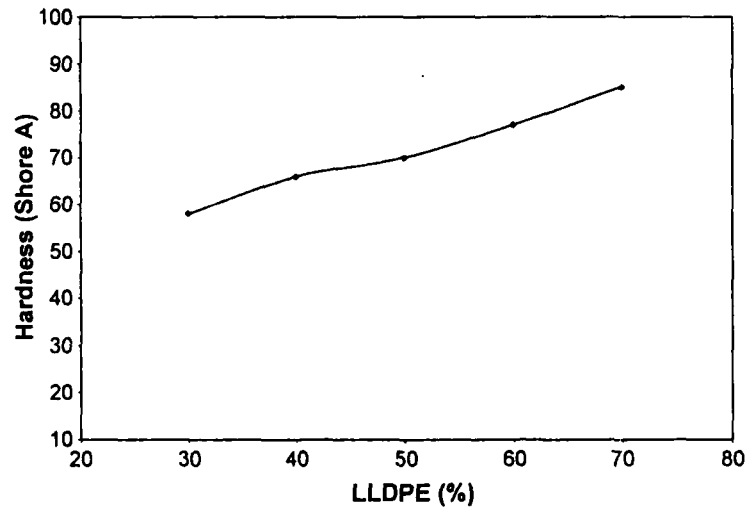
FIGURE 3 Elongation at break *versus* LLDPE content for NR/LLDPE blends.

FIGURE 4 The effect of LLDPE content on the hardness of NR/LLDPE blends.

penetrant size [15] polymer structure [16], crosslink density, mode of crosslinking presence of fillers [17], temperature, *etc.* Figure 5 shows the effect of LLDPE content on the mass swell of NR/LLDPE blends at room temperature. It can be seen that the property

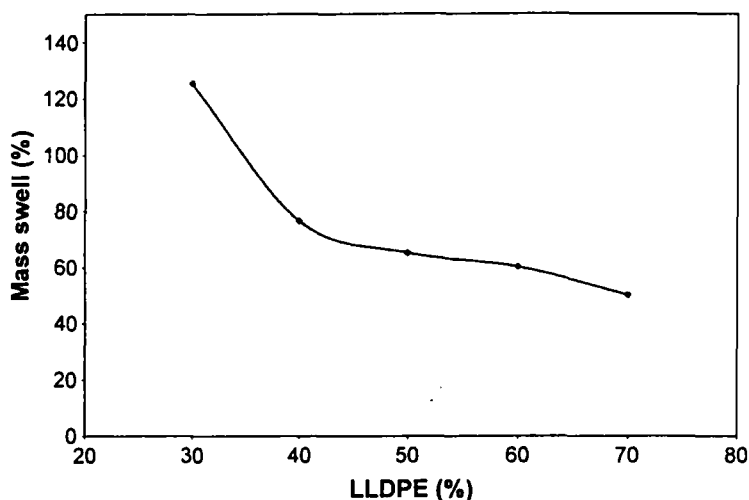


FIGURE 5 Variation of mass swell with LLDPE content for NR/LLDPE blends.

decreases with increasing LLDPE content in the blends. As the LLDPE content increases within the blends, the blends will become stiffer as shown in Figure 2 and less penetrable by the oil.

### 3.3. Scanning Electron Microscopy (SEM)

Figures 6(a), (b) and (c) show the comparison tensile fracture surfaces of NR/LLDPE blends at different LLDPE content. In Figure 6(a), the fracture surface of 70/30 NR/LLDPE blend exhibits that the LLDPE was dispersed as domains in a continuous rubber phase. However as the LLDPE content increases as shown in Figure 6(b) (50/50 NR/LLDPE blend), the two phases seem to fit together and appear to be interconnected in a 3-D network. As the LLDPE content is increased further (30/70 NR/LLDPE blend), the rubber is dispersed as domains in a continuous phase of LLDPE. It can be seen also in Figures 6(a), (b) and (c) that as the LLDPE content in NR/LLDPE increases, the nature of failure surface changes from smooth failure surface into rough failure surface. The rough failure surface of 30/70 NR/LLDPE blend indicates that a higher energy is needed to cause catastrophic failure compared to 70/30 NR/LLDPE and 50/50 NR/LLDPE blends.



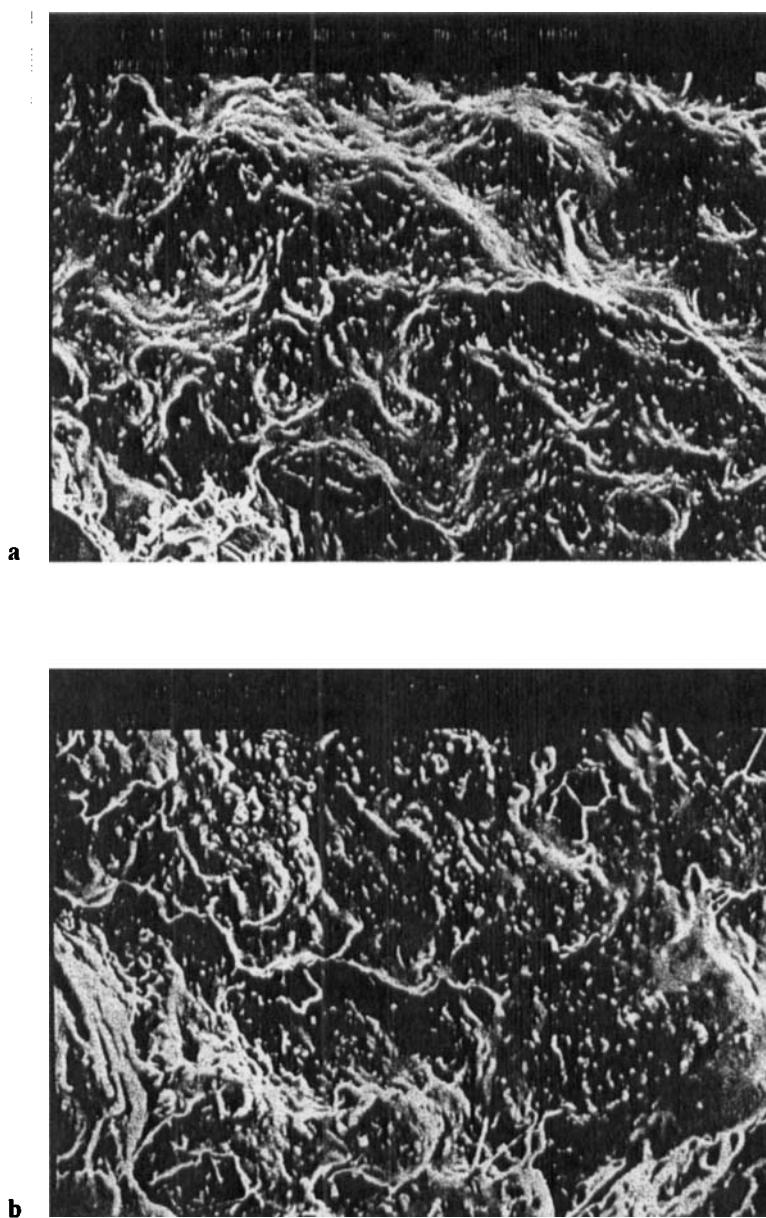


FIGURE 6 The tensile fracture surfaces of various NR/LLDPE blends (a) 70/30 NR/LLDPE blend (b) 50/50 NR/LLDPE blend and (c) 30/70 NR/LLDPE blend (Magnification  $\times 500$ ).



FIGURE 6 (Continued).

### 3.4. The Effect of WRHA Loading on Mechanical Properties of NR/LLDPE Blend

Figure 7 shows the effect of WRHA loading on the tensile strength of NR/LLDPE blends. It can be seen that the tensile strength decreased steadily as the loading of WRHA increased in the blends. This is not surprising since other researchers [18, 19] also reported that the incorporation of filler into thermoplastic matrix may not necessarily increase the tensile strength of a composite. According to Manson and Sperling [20] and Nielsen [21], in the particulate filled composites due to poor stress transfer at the filler-polymer interphase discontinuity is created which generates weak structure. Agglomeration of the filler particles and dewetting of the polymer at the interphase aggravate the situation by creating stress concentration points which account for the weakness in the composite.

Figure 8 illustrates the effect of WRHA loading on tensile modulus *i.e.*, modulus at 100% elongation ( $M_{100}$ ) of the NR/LLDPE blend. The increase in modulus with WRHA loading clearly indicates that addition of WRHA increases rigidity of NR/LLDPE blend through

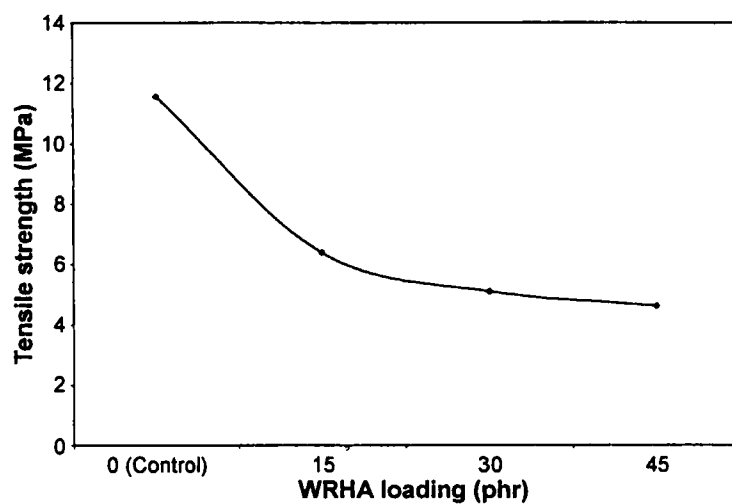


FIGURE 7 The effect of WRHA loading on the tensile strength of 30/70 NR/LLDPE blends.

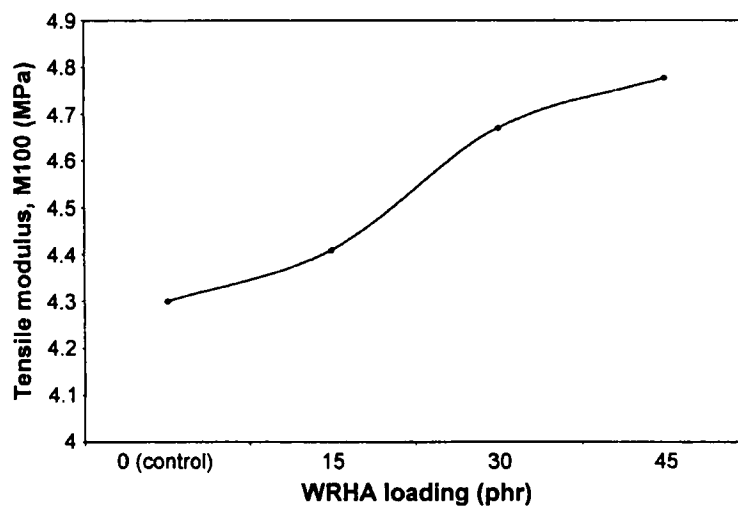


FIGURE 8 Variation of tensile modulus with WRHA loading for 30/70 NR/LLDPE blends.

introduction of restrictions in the mobility of the polymer molecules. The results for hardness test in Figure 9 also indicates that the incorporation of WRHA has increased the stiffness of the blends.

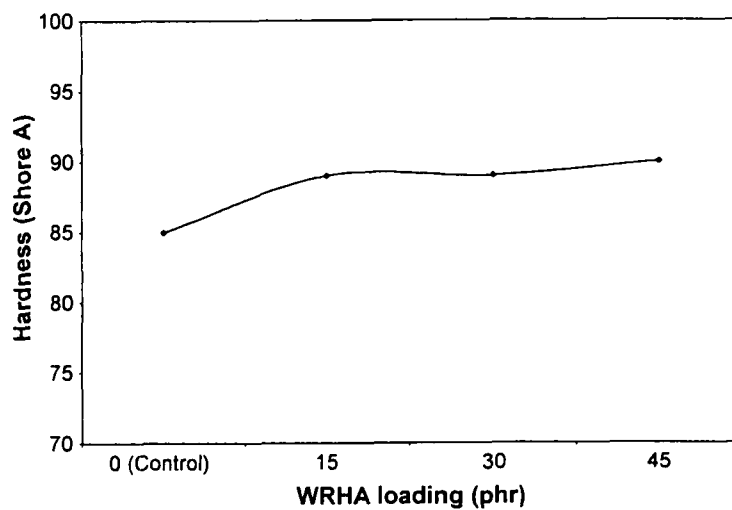


FIGURE 9 Hardness versus WRHA loading for 30/70 NR/LLDPE blends.

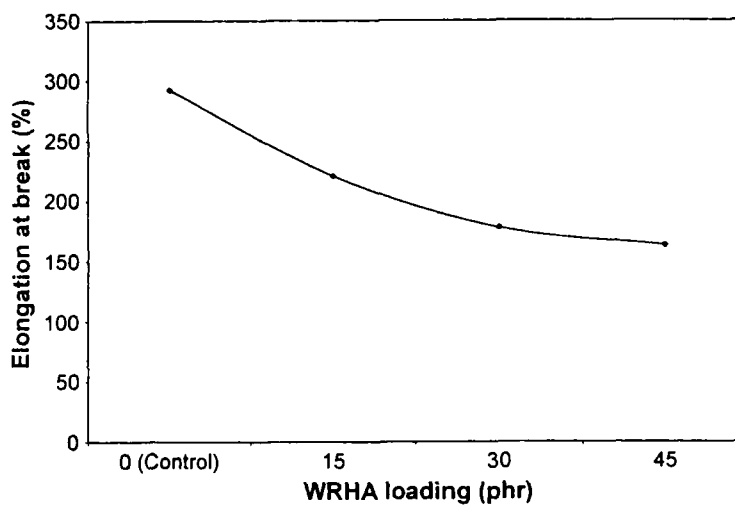


FIGURE 10 The effect of WRHA loading on the elongation at break of 30/70 NR/LLDPE blends.

The incorporation of WRHA into NR/LLDPE blends has resulted in a dramatic reduction in the elongation at break,  $E_b$  (Fig. 10). Incorporation of fillers that have poor adhesion to the polymer matrix

seems to cause interruption in the alignment process of the chains. When filler loading is increased, more weak interfacial regions between the filler surface and polymer matrix are formed. As crack travels easier through the weaker interfacial regions, the composite therefore fractures at lower value of elongation with increasing filler loading.

Figure 11 shows the effect of WRHA loading on mass swell of the NR/LLDPE blends. It can be seen that the property decreases with increasing filler loading. A relatively high loading of WRHA in the NR/LLDPE blends was thought to have a smaller distance (free volume) between the polymer networks. This resulted in a lower flexibility and mobility of the polymer chain. As a consequence of this, it was difficult for the oil to penetrate into the NR/LLDPE blends. Figures 12(a) and (b) show the tensile fracture surfaces of WRHA filled 30/70 NR/LLDPE blends at 15 and 60 phr loading of WRHA. The fracture surface for control blend (0 phr of WRHA) in Figure 6(c) exhibits the well blend of NR/LLDPE where the rubber particles dispersed in the continuous phase of LLDPE. It can be seen in Figure 12(b) that there are many clumps of de-wetted WRHA agglomerates on the surface (poor dispersion). According to Hess [22] the poor dispersion of filler give rise to certain detrimental effects such as

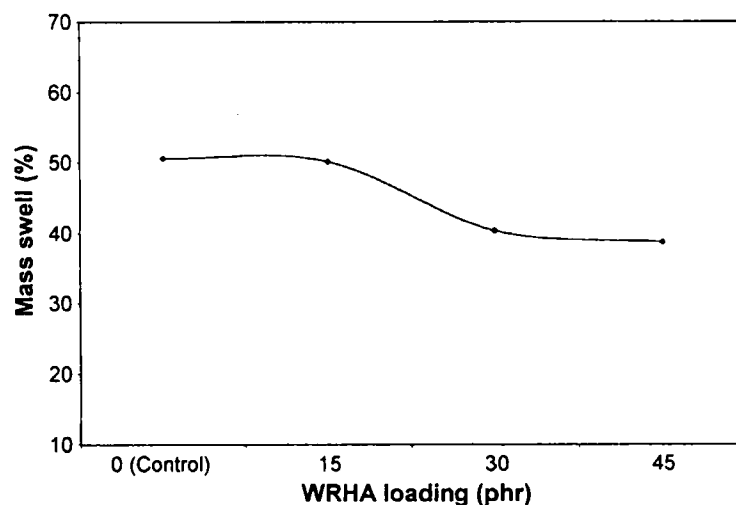


FIGURE 11 Variation of mass swell with WRHA loading for 30/70 NR/LLDPE blends.

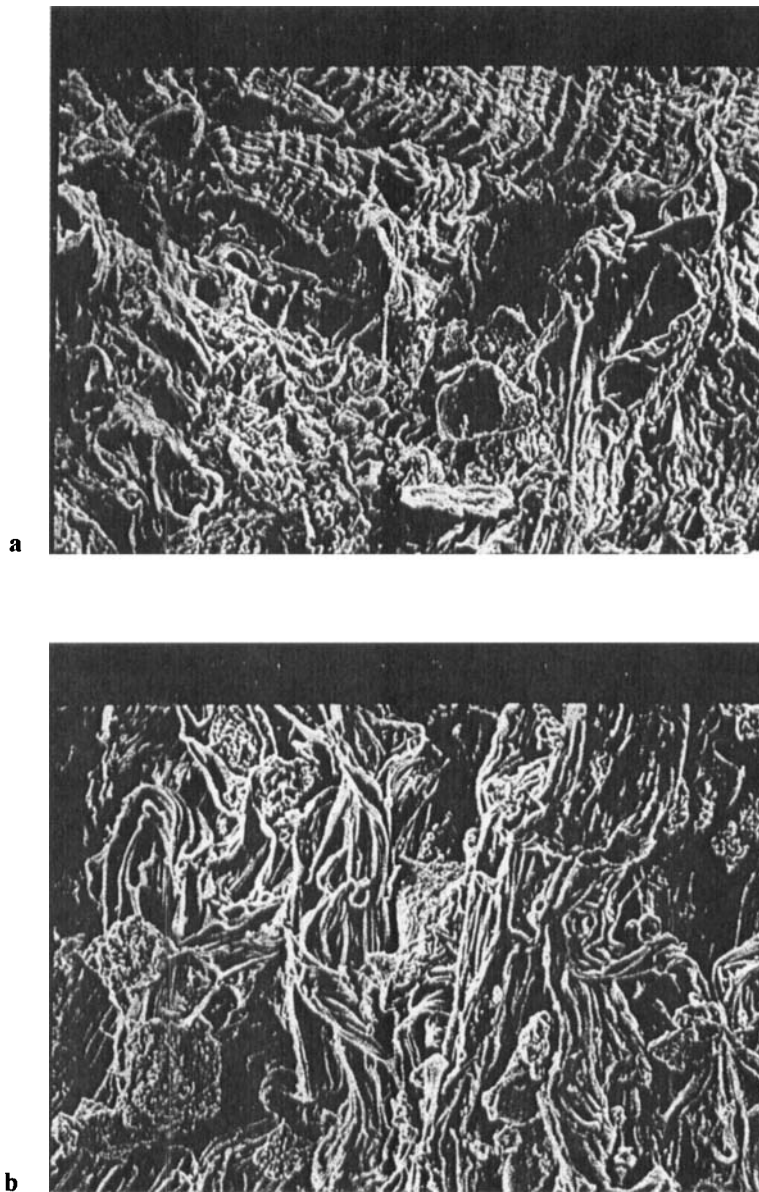


FIGURE 12 Tensile fracture surfaces of 30/70 NR/LLDPE blends with (a) 15 phr of WRHA and (b) 60 phr of WRHA. (Magnification  $\times 500$ ).

reduced product life, poor performance in service, poor product appearance, poor processing characteristics *etc.* However, the similar blend with 15 phr of WRHA in Figure 12(a) exhibits better dispersion with less de-wetted or undispersed WRHA agglomerates. These micrographs explain why the tensile strength decreases with increasing filler loading.

#### 4. CONCLUSION

1. The increasing content of LLDPE into NR/LLDPE blend has resulted in the improvement in tensile strength, tensile modulus and hardness of the blends. However the elongation at break and mass swell exhibits opposite trend. This observation is due to the increases in stiffness and brittleness of blends.
2. The rough failure surface of 30/70 NR/LLDPE blend indicates that a higher energy is needed to cause catastrophic failure compared to 70/30 NR/LLDPE and 50/50 NR/LLDPE blends.
3. The tensile strength of 30/70 (W/W) NR/LLDPE blends decreases with increasing WRHA loading. The poor performance in this property has been attributed to the poor filler-matrix interaction or compatibility and poor dispersion of filler in NR/LLDPE blends.

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