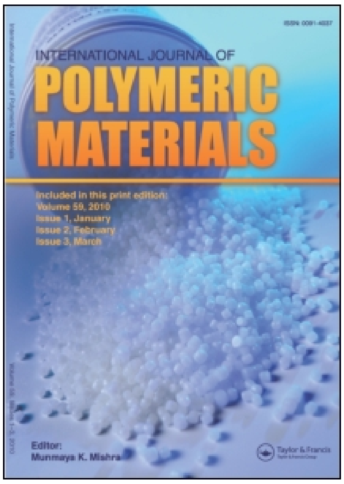

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

The Effect of Partial Replacement of Silica by White Rice Husk Ash in Natural Rubber Composites

H. Ismail[a]; F. L. Chung[a]
[a] School of Industrial Technology, Universiti Sains Malaysia, Minden, Penang, Malaysia

To cite this Article Ismail, H. and Chung, F. L.(1999) 'The Effect of Partial Replacement of Silica by White Rice Husk Ash in Natural Rubber Composites', International Journal of Polymeric Materials, 43: 3, 301 — 312
To link to this Article: DOI: 10.1080/00914039908009692
URL: http://dx.doi.org/10.1080/00914039908009692

Intern. J. Polymeric Mater., 1999, Vol. 43, pp. 301-312
Reprints available directly from the publisher
Photocopying permitted by license only

The Effect of Partial Replacement of Silica by White Rice Husk Ash in Natural Rubber Composites

HANAFI ISMAIL* and F. L. CHUNG

School of Industrial Technology, Universiti Sains Malaysia, 11800 Minden, Penang, Malaysia

(Received 10 March 1998)

The potential of using white rice husk ash (WRHA) to partially replace the silica content in silica filled natural rubber vulcanizates was studied. Results show that the optimum weight ratio of WRHA/silica to obtain maximum enhancement of tensile and tear strengths was 20/30 (phr/phr). The increase of WRHA in weight ratio of WRHA/silica decreases the hardness and elongation at break but increases the resilience of the vulcanizates. The cure time, t_{90} and scorch time, t_2 also decrease with increasing of WRHA in weight ratio of WRHA/silica.

Keywords: Natural rubber; fillers; white rice husk ash; mechanical properties

1. INTRODUCTION

In our previous works [1,2] the potential of white rice husk ash (WRHA) as a filler for natural rubber (SMR-L) compounds has been studied. The optimum loading of WRHA to obtain maximum physical properties was achieved at 10 phr after which there was deterioration in properties. However for epoxidized natural rubber (ENR 50), the maximum physical properties was observed to occur at 20 phr of filler loading [3,4]. It was reported that WRHA was about 96% silica content with mean particle size of 5.4 μm, surface area of 1.4 m^2/g and density of 2.2 g/m^3 [3, 4].

*Corresponding author.

In this study the effect of partial replacement of silica by WRHA in natural rubber compounds have been evaluated. The work has been based on SMR 10, WRHA and precipitated silica (grade vulcasil S) formulation used in previous studies [1, 2] and comprises exploration of: (a) the effect of partial replacement of silica by WRHA on scorch and cure times of the composites; (b) the effect of partial replacement of silica by WRHA on tensile strength, tear strength, elongation at break, resilience and hardness of the composites; and (c) examination of the fracture surface of the composites by using a scanning electron microscope (SEM).

2. EXPERIMENTAL

2.1. Materials

Natural rubber (SMR-L) was obtained from Rubber Research Institute of Malaysia (RRIM). White rice husk ash (WRHA) was supplied by Plastic Technology Centre, S.I.R.I.M., Malaysia. The physical properties of WRHA are given elsewhere [3, 4]. Precipitated silica (grade Vulcasil S) and other materials such as zinc oxide, sulphur, stearic acid, antioxidant and accelerators were purchased from Bayer (M) Ltd. All material were used as supplied and the semi-efficient sulphur vulcanization (semi-EV) system was employed.

2.2. Processing and Mechanical Properties Measurement

Formulations of the mixes are given in Table I. The mixing was carried out as per ASTM D 3182-80 on a two-roll laboratory size mixing mill (160 mm × 320 mm). Optimum cure time, t_{90} was determined by Monsanto Rheometer R-100. Vulcanization was carried out at 150°C on an electrically heated single daylight hydraulic press with a force of 10 MPa. The vulcanizates were tested for different mechanical properties according to respective ASTM standards. Tensile and tear properties were studied using an Instron Universal Testing Machine, model 1114 at a crosshead speed of 50 cm/min. For tensile properties, samples were punched out using die E from moulded sheets and were tested ASTM D412 method A. For tear strength measurements,

TABLE I Formulation to study the potential of WRHA to replace silica filler in natural rubber compounds

Ingredients	Mix No. A	B	C	D	E	F
Natural rubber (SMR-L)	100	100	100	100	100	100
Zinc oxide	5.0	5.0	5.0	5.0	5.0	5.0
Stearic acid	3.0	3.0	3.0	3.0	3.0	3.0
Flectol H[a]	1.0	1.0	1.0	1.0	1.0	1.0
CBS[b]	2.0	2.0	2.0	2.0	2.0	2.0
Sulphur	2.5	2.5	2.5	2.5	2.5	2.5
WRHA	0	10	20	30	40	50
Silica	50	40	30	20	10	0

[a] Poly-1,2-dihydro-2,2,4-trimethylquinoline.
[b] *n*-cyclohexylbenthiazyl sulphenamide.

standard test pieces (ASTM D624 – Type C) were used. Hardness measurements were made according to ASTM D2240 using Shore A durometer. Resilience test was done by using Wallace Tripsometer Dunlop according to BS 903: Part A8. The angle of rebound was measured and resilience calculated using the equation below:

$$R = \frac{1 - \cos(\text{angle of rebound}) \times 100}{1 - \cos(\text{angle of fall})}$$

SEM studies of the tear fracture surfaces were carried out on a scanning electron microscope, model Leica Cambridge S-360. The specimens were sputter coated with gold and then examined under SEM within 24 hours of testing. The angle of tilt of the specimens was 0° to the beam and the orientation of the photographs were kept constant.

3. RESULTS AND DISCUSSION

3.1. The Effect of Partial Replacement of Silica by WRHA on Cure Time, t_{90} and Scorch Time, t_2

Table II shows reduction in t_{90} and t_2 when the loading of WRHA in WRHA/silica weight ratio (phr/phr) increases. Precipitated silica is a filler which can retard curing rate [5–8]. Pure silica absorb the zinc oxide and accelerator during mixing. This reduces the zinc oxide and accelerator concentration which is needed in the vulcanization process,

TABLE II The effect of partial replacement of silica by WRHA on t_{90} and t_2 of the vulcanizates*

WRHA/silica (phr/phr)	0/50	10/40	20/30	30/20	40/10	50/0
t_{90} (min)	28.5	12.5	8.4	5.7	5.4	4.9
t_2 (min)	10.0	3.8	3.4	2.5	1.4	1.0

*Monsanto Rheometer R-100 at 150°C.

especially at higher concentrations of silica (≥ 30 phr) [5, 6]. So, the t_{90} of vulcanizates in Table II become shorter as the silica loading decreases. We have reported in our previous reports [1, 2] that the WRHA which contain various metal oxides can increase the curing rate. When the loading of WRHA in WRHA/silica weight ratio increases, the metal oxides content in rubber mixes also increases and contributes to the increasing of vulcanization rate.

3.2. Examination of Tear Failure Surface

Figures 1–3 show the comparison of tear failure surfaces of 20/30, 30/20 and 50/0, WRHA/silica (phr/phr) filled SMR-L vulcanizates respectively. From Figures 2 and 3, it can be seen that the failure surface structures of these two composites show more filler agglomeration

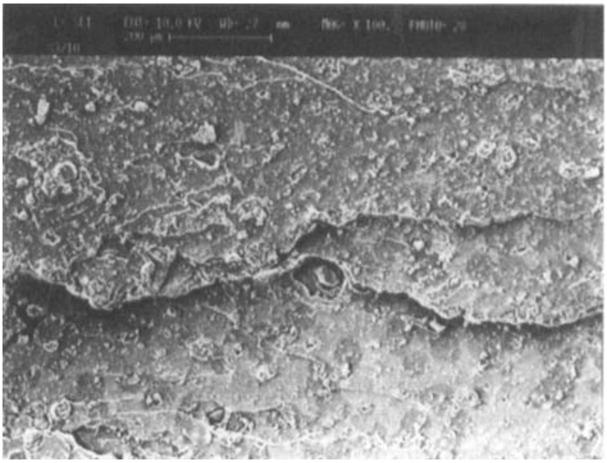

FIGURE 1 SEM micrograph of tear failure surface of 20/30, WRHA/silica (phr/phr) filled SMR-L vulcanizate at magnification of 100X.

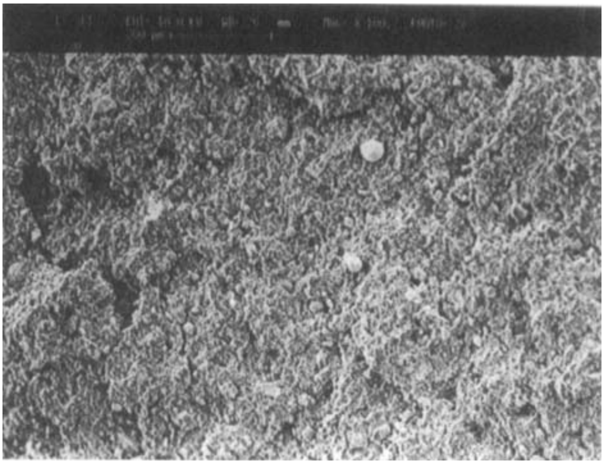

FIGURE 2 SEM micrograph of tear failure surface of 30/20, WRHA/silica (phr/phr) filled SMR-L vulcanizate at magnification of 100X.

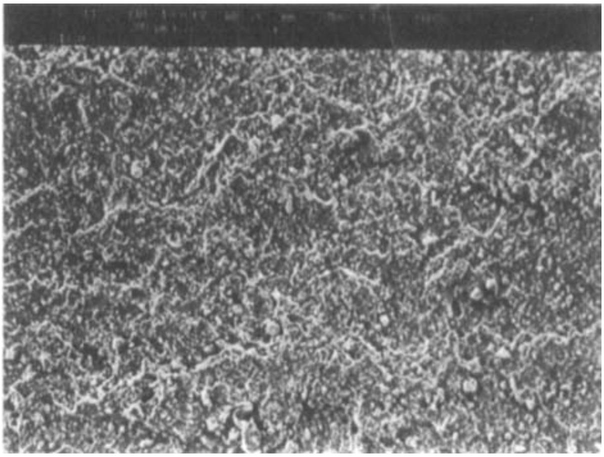

FIGURE 3 SEM micrograph of tear failure surface of 50/0, WRHA/silica (phr/phr) filled SMR-L vulcanizate at magnification of 100X.

and blank spots compared to Figure 1. This is due to increasing WRHA loading in the Figures 1–3. In our previous works [1, 2], the incorporation of WRHA more than 10 phr weakens the rubber-filler interaction. In vulcanizates containing both the WRHA and silica, when the loading of WRHA increases, the poor interaction of WRHA to rubber matrix is also increased, contributing to the formation of the weak points during tear test. It is expected that the tear strength of the 20/30, WRHA/silica (phr/phr) is higher than 30/20 and 50/0, WRHA/silica (phr/phr) filled vulcanizates.

3.3. The Effect of Partial Replacement of Silica by WRHA on Mechanical Properties

Precipitated silica is good reinforcing filler [8]. From Figures 4 and 5, it can be seen that the vulcanizate containing 30 phr of silica exhibits the highest tensile and tear strengths. Untreated silica has a high specific surface energy. Silica – silica interaction is stronger than silica – rubber interaction. The tendency of silica to form agglomerates is high especially at a higher concentration [9]. The agglomeration of silica reduces the interaction with rubber matrix and become the weak point in the vulcanizates. The tensile and tear strength of the vulcanizates was reduced when the silica loading exceeded 30 phr.

For WRHA, the maximum tensile and tear strengths of the vulcanizate (contain both WRHA and silica) occurred at WRHA loading of 20 phr (20/30 WRHA/silica phr/phr). Compared to vulcanizates filled with WRHA only, the optimum loading of WRHA was 10 phr [1, 2]. This means that both the silica and WRHA could be used in duo to reinforce natural rubber vulcanizates, and the fact that the latter, in this system could be used at higher concentration to give better tensile and tear strengths.

Figure 6 shows the effect of partial replacement of silica by WRHA on elongation at break, E_b. It can be seen that E_b decreases with increasing WRHA loading in the weight fraction of WRHA/silica filled vulcanizates. Our previous works [1, 2] showed that E_b decreases with increasing WRHA in the rubber vulcanizates. However Tultz *et al.* [5] and Bachmann *et al.* [6] reported that incremental silica loading increases the elongation at break. When the loading of WRHA in weight fraction of WRHA/silica increases, the effect of WRHA on E_b is more dominant than silica and this caused the E_b of the vulcanizates to decrease.

According to Jacques [10], the addition of particulate filler reduces the resilience of the rubber vulcanizate. The reduction of resilience

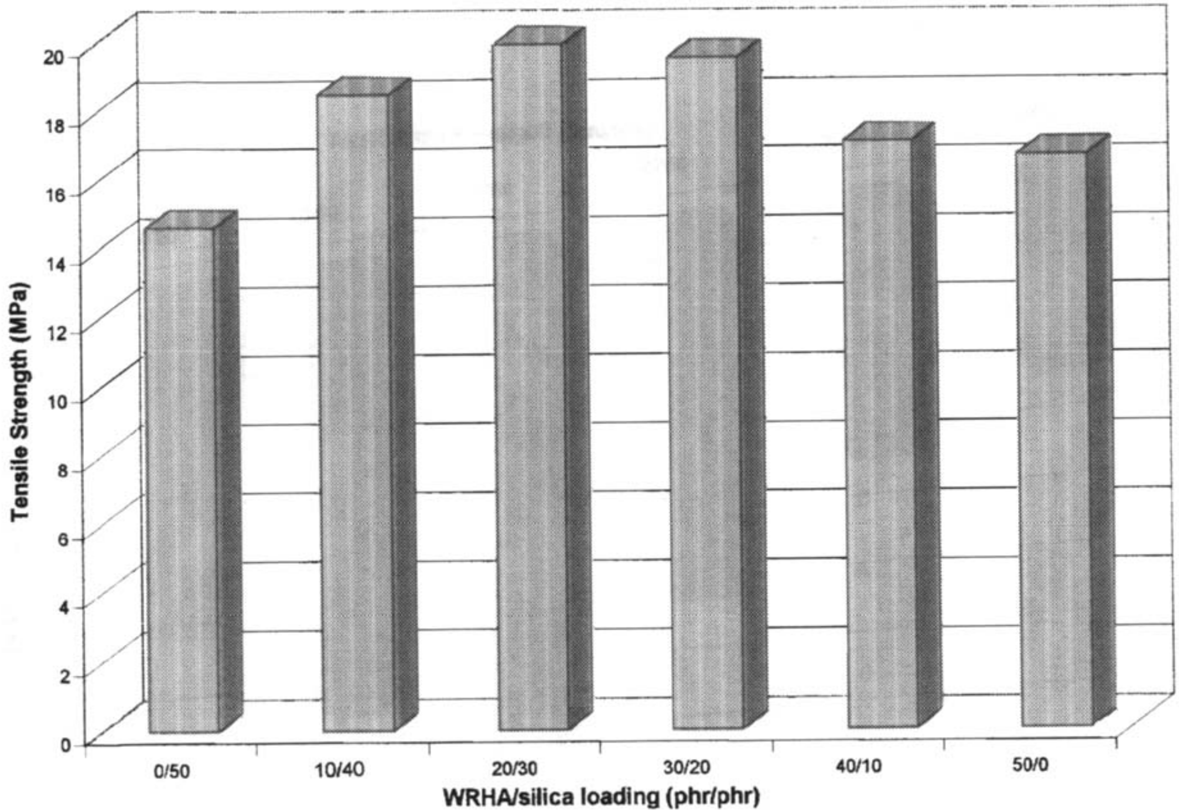

FIGURE 4 The effect of partial replacement of silica by WRHA on tensile strength of SMR-L vulcanizates.

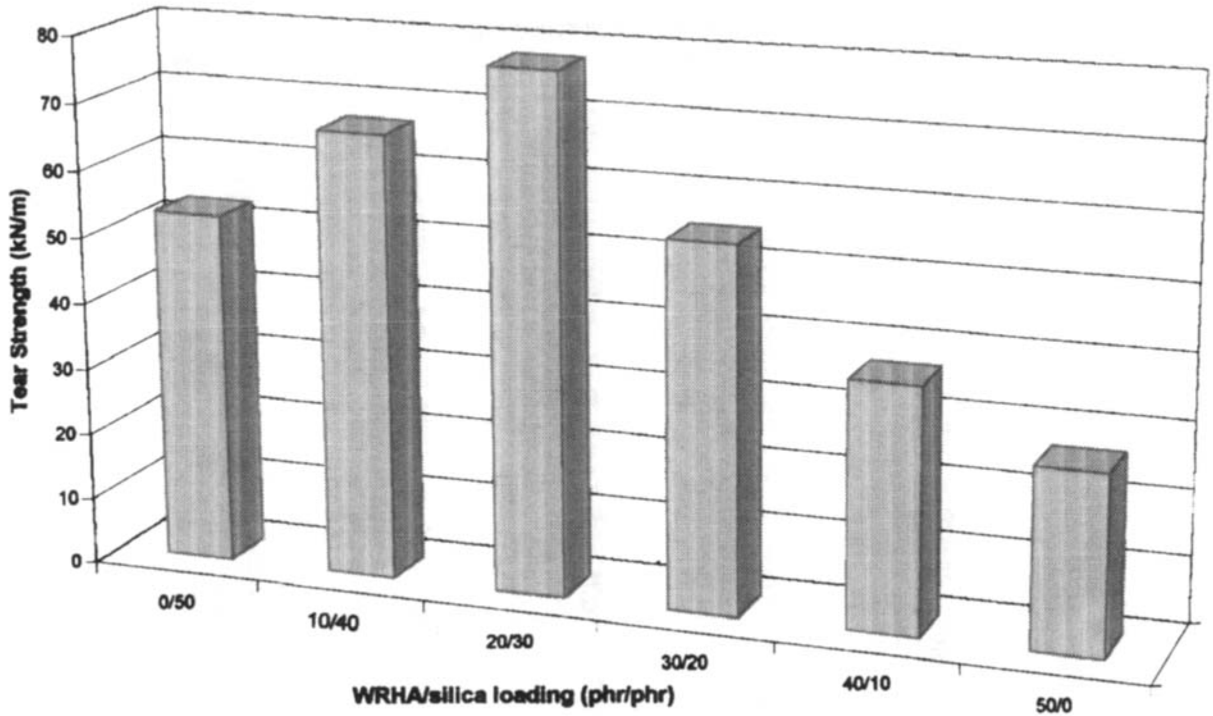

FIGURE 5 The effect of partial replacement of silica by WRHA on tear strength of SMR-L vulcanizates.

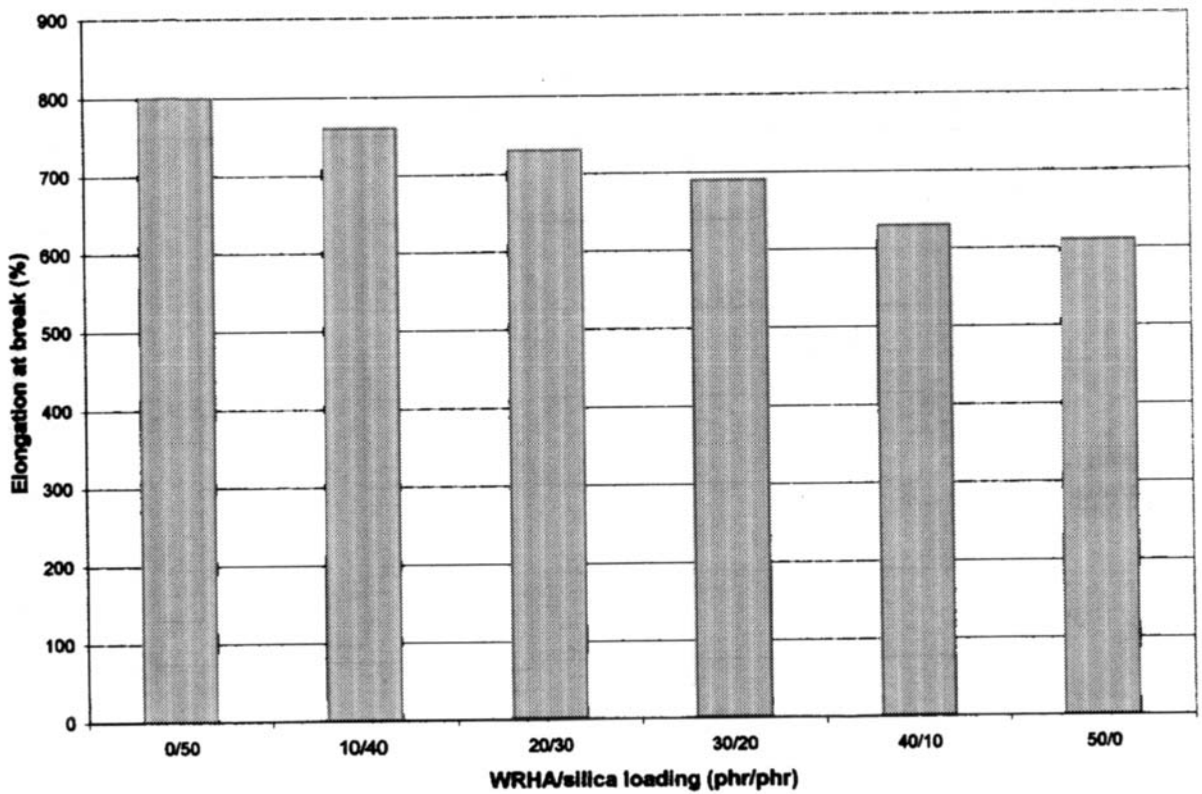

FIGURE 6 The effect of partial replacement of silica by WRHA on elongation at break of SMR-L vulcanizates.

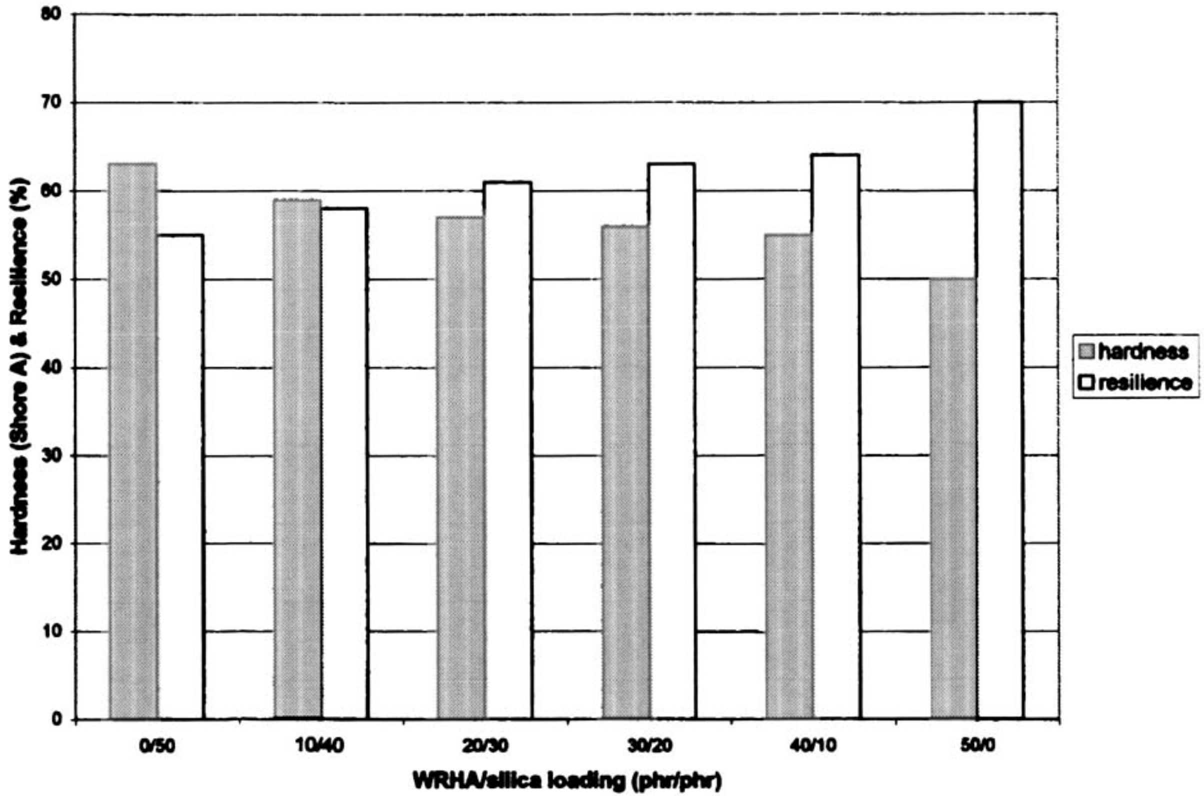

FIGURE 7 The effect of partial replacement of silica by WRHA on hardness and resilience of SMR-L vulcanizates.

become more significant if the filler used is reinforcing fillers such as carbon black and silica. The resilience is less affected by WRHA although at concentration of 50 phr [1, 2]. So for vulcanizates filled with both WRHA and silica, the resilience is more affected by silica. From Figure 7, it can be seen that the resilience decreases as the silica loading in the weight fraction of WRHA/silica increases.

Figure 7 also shows that the hardness of the vulcanizates increases with the increasing silica loading in the weight fraction of WRHA/silica. Jacques [10] reported that for particulate filler, the more reinforcing filler would result in harder vulcanizates. Silica is a more reinforcing filler then WRHA [8 – 11]. This explains why vulcanizates filled with higher loading of silica show higher hardness value.

4. CONCLUSION

1. White rice husk ash is a potential filler to partially replace silica in natural rubber compounds.
2. The increase of WRHA in weight ratio of WRHA/silica (phr/phr) decreases the cure time, t_{90} and scorch time, t_2 of the vulcanizates.
3. The increase of WRHA in weight ratio of WRHA/silica (phr/phr) weakens the rubber – filler interaction. However when WRHA used together with silica the optimum tensile and tear strengths occurred at 20 phr of WRHA compared to 10 phr when WRHA is used alone.
4. The hardness and elongation at break decreases with increasing WRHA in weight ratio of WRHA/silica (phr/phr) but the resilience shows opposite trend.

References

[1] Hanafi Ismail and Nasaruddin, M. N., *Polymer International*, in press.
[2] Hanafi Ismail and Chung, F. L., *Polym.-Plast. Technol. Eng.*, in press.
[3] Hanafi Ismail, Ishiaku, U. S., Arinab, A. R. and Mohd Ishak, Z. A. (1997). *Intern. J. Polymeric Mater.*, **36**, 39.
[4] Hanafi Ismail, Ishiaku, U. S., Azhar, A. A. and Mohd Ishak, Z. A. (1997). *J. of Elastomers and Plastics*, **29**, 270.
[5] Tultz, J. P., Jo-Ann, E. Bice, Timothy, A. Okel and Shailesh, D. Patkar (1997). *International Rubber Conference*, Kuala Lumpur, Malaysia, Oct. 6 – 9, pp. 417.

[6] Bachmann, J. H., Sellers, J. W., Wagner, M. P. and Wolff, R. F. (1959). *Rubb. Chem. Technol.*, **32**, 1286.
[7] Wolff, S., Burmester, K. and Tan, E. H. (1976). *Kautsch. Gummi Kunstst.*, **29**, 691.
[8] Fetterman, M. Q. (1986). *Rubber World*, **194**, 38.
[9] Wolff, S., Gorl, U., Wang, M. J. and Wolff, W. (1993). *European Rubber J.*, **171**, 16.
[10] Jacques, J. E. (1971). *Rubber Technology and Manufacture*, Ed. Blow, C. M., Butterworth and Co Ltd., London, pp. 321.
[11] Nasir, M. and Low, M. H. (1988). *Proc. International Rubber Technology Conference*, Penang, Malaysia, Oct. 17–19, pp. 370.