

PROPERTIES OF RICE HUSK POWDER/NATURAL RUBBER COMPOSITE

Razif Nordin¹, C. M. S. Said² and H. Ismail³

¹*Faculty of Applied Science, Universiti Teknologi MARA Sabah, L/B 71,
88997 Kota Kinabalu, Sabah, Malaysia*

²*Faculty of Applied Science, Universiti Teknologi MARA,
40450 Shah Alam Selangor, Malaysia*

³*School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia,
13400 Nibong Tebal, Penang, Malaysia*

ABSTRACT

The effects of epoxidized natural rubber (ENR) on cure characteristics, mechanical properties, and swelling behavior of rice husk powder (RHP) filled natural rubber (NR) composite were studied. Results indicated that the scorch time t_2 and cure time t_{90} of the composite decrease with increasing filler loading and with presence of a ENR. Minimum torque also increases with increasing filler loading but at similar filler loading shows lower value with the presence of ENR. The mechanical properties of composite viz. tensile strength, tensile modulus and hardness were also improved with additional of ENR.

INTRODUCTION

The interest in using agro-based fibre has received much attention from both the research and industry communities, and has been well reviewed [1], [2] for the last few years. On the environmental aspect, it would be very interesting if agro fibre used instead of inorganic materials as reinforcement in some structural applications. Agro fibres have many advantages compared to inorganic filler, such as they have very low density to volumetric cost, and they are recyclable and biodegradable. Additionally they are renewable raw materials and have relatively high strength and stiffness.

Agro fibres like crop species are cotton stalk, barley, rice, oat and wheat straws are agricultural products. Rice husk at present is a waste product of rice milling process and is produced about 2500 metric tones per year in the Malaysia [3]. Currently rice husk is used as bedding materials for animals and industrial applications of these materials are limited. Fortunately, efforts are already underway for producing value added products, such as composite materials. In recent research, rice husk is used as a reinforcing agent for thermoplastics [4] – [6]. It has been shown that the use of rice husk in thermoplastics composites results in an increase in tensile modulus and hardness. These properties were further enhanced with the use of a compatibilizer, which improved the interfacial adhesion between rice husk and thermoplastics matrix.

In the present study, a new type of composite material viz. rice husk powder (RHP) reinforced natural rubber composite was developed. The effect of filler loading and epoxidized natural rubber (ENR) as compatibilizer on the cure characteristics, tensile properties, and hardness of RHP filled natural rubber composites will be

reported. Scanning electron microscopy (SEM) studies were carried out to determine the failure mechanism of the composites.

EXPERIMENTAL

The formulation used in this study is shown in Table 1. Natural rubber (SMR L) and epoxidized natural rubber 25% (ENR) was obtained from the Rubber Research Institute of Malaysia (RRIM). The rice husk powder (RHP) used in this work was supplied by Bernas Perdana Sdn. Bhd. Penang, Malaysia. RHP was dried in an air oven at 100 °C for 18 hours to expel moisture prior to blending. In this study, the particle size of RHP was in the range of 60 – 90µm. Table 2 shows the chemical composition of the RHP fibre used in this study. Other chemicals such as zinc oxide, sulphur, stearic acid, N-cyclohexyl-2-benzothiazole sulfenamide, and 2,2-methylene-bis(4-methyl-6-tertbutylphenol) were all purchased from Bayer (M) Ltd.

Table 1 Formulation for rice husk powder reinforced natural rubber composite

Ingredients	phr								
	100	100	100	100	100	100	100	100	100
Natural Rubber	100	100	100	100	100	100	100	100	100
Rice husk powder, RHP	0	10	20	30	50	10	20	30	50
Epoxidized natural rubber, ENR	-	-	-	-	-	10	10	10	10

Zinc Oxide = 5.0 phr; Stearic Acid = 2.5 phr; Sulphur = 1.5phr; 2,2-methylene-bis-(4-methyl-6-tert-butylphenol)=2.0 phr; N-cyclohexyl-2-benzothiazole sulfenamide = 2.0 phr.

Table 2 Chemical composition of rice husk powder.

Chemical composition	%
Moisture	9.0
Cellulose	32.0
Hemicellulose	19.0
Lignin	16.0
Silica	13.0
Ash	11.0

The cure characteristics were studied using a Monsanto Moving Die Rheometer (MDR 2000) according to ASTM D 224. About 4 g samples of the respective composite were tested at a vulcanization temperature (150 °C). The composite were compression molded using hot press with a force of 10 MPa into 3 mm thick sheets at 150 °C, according to respective cure time t_{90} determined with MDR 2000.

Dumbbell shaped samples were cut from the moulded sheets according to ASTM D 3182. Tensile tests were performed at a crosshead speed of 500 mm/min using

Testometric Tensometer M500 according to ASTM D 412. Hardness measurements of samples were done according to ASTM D 1415 using a Wallace dead load, with hardness ranging from 30 to 85 IRHD (International Rubber Hardness Degree).

RESULTS AND DISCUSSION

The effect of RHP loading and ENR as compatibilizer on the scorch time, t_2 and cure time, t_{90} of NR composite can be seen in Table 3. It can be seen that both properties decreased with increasing RHP loading. The reduction in scorch time and cure time of rubber composite is due to longer mixing time of rubber composite during processing. As the filler loading increases in rubber composite, the time of incorporation also increases and consequently generated more heat due to friction. Similar trends were also observed by Ismail and Abdul Khalil [7], Geethamma et al [8] and Jacob et al. [9] using different types and size of natural fibre in the rubber composite. However at the same filler loading, NR composite with ENR had a shorter t_2 , and t_{90} than control composite (without ENR). This is because the activation effect of epoxide group and amine group on the ring opening pathway of sulphur which accelerated the curing process of NR composite [10].

Table 3 shows the effect of RHP loading on minimum torque M_L of RHP filled NR composite. It can be seen that the minimum torque increase with increasing filler loading. This observation is expected because as more filler particles are incorporated into rubber matrix, the flexibility of the rubber chain is reduced resulting in more processing difficulties. However at a similar filler loading, NR composite with ENR exhibit slightly higher minimum torque than NR composite without ENR. Similar trends were also observed by Ismail and Leong [11] at different ratio of NR and ENR grades.

The effect of RHP loading on torque difference (maximum torque – minimum torque), $M_H - M_L$ of NR composite with and without ENR are shown in Table 3. It can be seen that the torque difference increases with increasing RHP loading for both composite. The torque difference which indicates the extent of crosslink density of the rubber composite [12]. It clear that, at a similar filler loading, RHP/NR composite with ENR have higher torque difference than similar composite that do not contain ENR. This suggests that ENR has some effect on the crosslink density. In our previous work [13] we found that the incorporation of ENR in rubber composite contributed to extra degree of crosslinking.

The variation of tensile strength with RHP loading of NR composite with or without ENR is shown in Table 3. For both materials, the tensile strength increases up to 20 phr of RHP loading and then decreases with further increase of filler loading. Below 20 phr of RHP loading, smaller particle size and uniform dispersion of RHP in NR composite contribute to higher tensile strength. But, when more than 20 phr was used, the weak interaction and bonding between RHP particles and NR matrix was responsible for the deterioration in tensile strength. Furthermore, the particles of RHP are neither of

irregular shape nor size (Fig. 1). This can decrease the vulcanizates strength due to the inability of the filler to support stress transferred from polymer matrix [14]. However at a similar filler loading, the tensile strength of the RHP/NR composite increased significantly by incorporating ENR. This might be due to the ability of ENR to improve filler dispersion and enhance filler/matrix adhesion [15]. Another reason is epoxide group of ENR capable in increase the crystallization of NR composite thus yield higher tensile strength [16].

Table 3: Characteristics of Rice Husk Powder/Natural Rubber composite.

NR	100	100	100	100	100	100	100	100	100
RHP	0	10	20	30	50	10	20	30	50
ENR	-	-	-	-	-	10	10	10	10
Cure time, t_{90} (min)	11.15	8.42	7.14	6.42	6.20	6.31	5.69	5.27	4.89
Scorch time, t_2 (min)	6.20	4.72	4.04	3.50	3.12	3.79	3.29	3.01	2.34
M_L (dNm)	0.06	0.09	0.11	0.14	0.16	0.10	0.13	0.15	0.18
$M_H - M_L$ (dNm)	6.60	6.90	7.43	7.97	8.98	7.46	7.85	8.63	9.65
Tensile Str. (MPa)	19.60	21.81	23.00	18.06	12.18	23.69	24.29	19.30	13.34
M100 (MPa)	0.93	1.24	1.54	1.91	2.24	1.28	1.61	1.94	2.75
M300 (MPa)	3.27	3.33	5.02	6.14	6.79	3.85	4.76	6.24	7.24
E_b (%)	595	537	519	485	428	534	515	467	416
Hardness (IRHD)	40.50	42.50	45.00	47.50	50.00	44.00	47.13	49.10	53.00

Table 3 show the results for tensile modulus i.e. modulus at 100% elongation (M100) and modulus at 300% elongation (M300) and hardness of the NR composite. As expected, M100, M300 and hardness increase with increasing RHP loading whereas elongation at break decreases. However at a similar filler loading, RHP/NR composite with ENR show higher M100, M300, and hardness but lower elongation at break, E_b than RHP/NR composite without ENR. As discussed before, this is due to the improvement of crosslink density and better interaction between RHP and NR composite with the presence of ENR.

The comparison of SEM tensile fracture of RHP filled NR composite at 20 and 50 phr with or without ENR are shown in Fig. 2 – 5, respectively. The failure surface of RHP filled NR composite with ENR in Figs. 2 and 3 exhibit a more uniform dispersion of filler in rubber matrix. However for RHP/NR composite without ENR (Figs. 4 and 5), the failure surface exhibit many agglomerates and holes formation after RHP were pulled out from the rubber matrix. This type of failure indicates poor adhesion between filler and rubber matrix

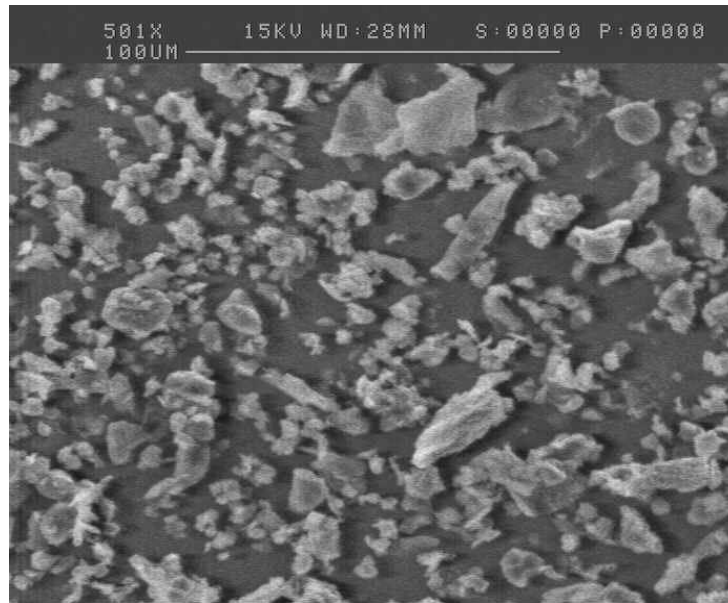


Figure 2: SEM of rice husk powder (magnification x 500)

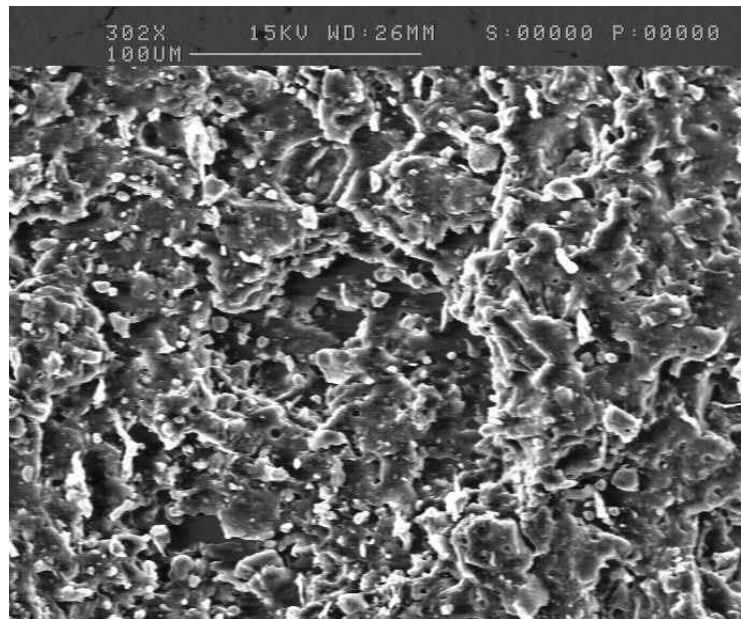


Figure 2: SEM tensile fracture of RHP filled NR composite at 20 phr of filler and with ENR at magnification x300

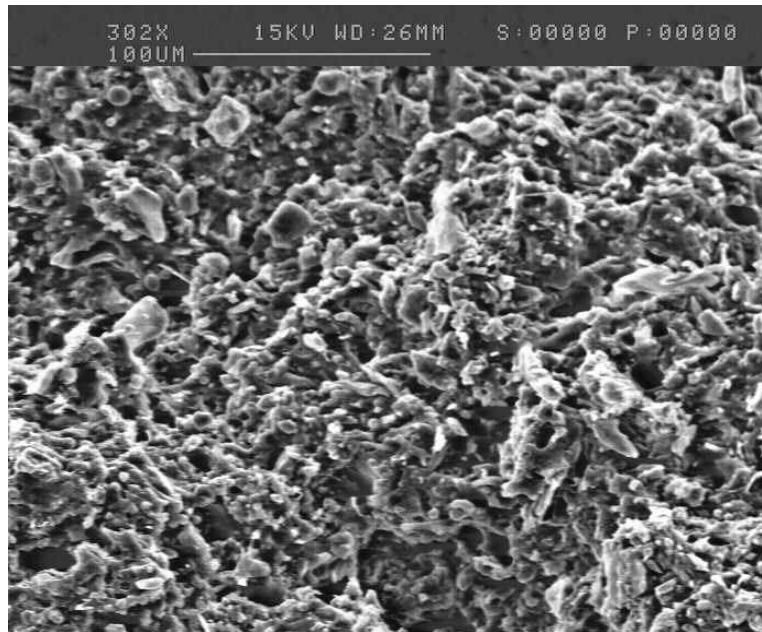


Figure 3: SEM tensile fracture of RHP filled NR composite at 50 phr of filler and with ENR at magnification x300.

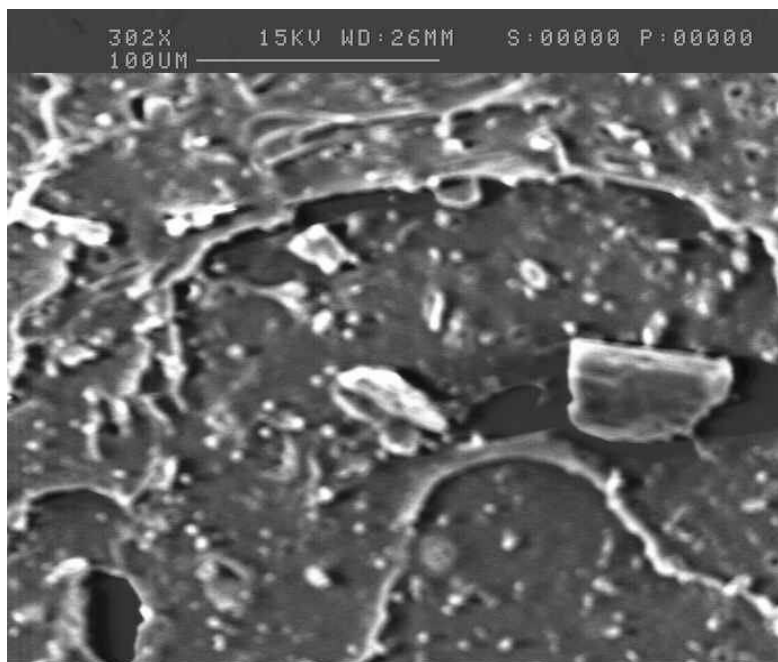


Figure 4: SEM tensile fracture of RHP filled NR composite at 20 phr of filler and without ENR at magnification x100.

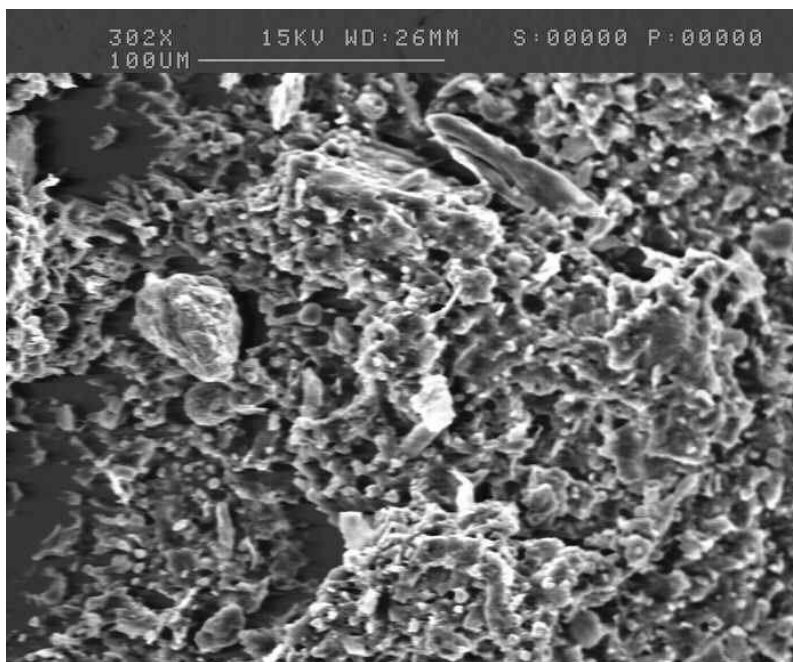


Figure 5: SEM tensile fracture of RHP filled NR composite at 50 phr of filler and without ENR at magnification x300.

CONCLUSIONS

Based on the above studies, the following conclusion can be drawn

1. The scorch time, and cure time of RHP/NR composite were shorter with the presence of ENR as compatibilizer.
2. Results from torque difference indicates that ENR has increased the crosslink density of RHP filled NR composite.
3. The mechanical properties such as tensile strength, tensile modulus, elongation at break, and hardness of RHP/NR composite were enhanced by the presence of ENR.
4. SEM tensile fracture of the blends shows that ENR improved the RHP dispersion and increase the interaction between RHP and NR matrix.

ACKNOWLEDGEMENTS

The authors would like to thank the Universiti Teknologi MARA for sponsoring this work under project IRPA (vote 100-CHS: IRDC 9/19/22). The authors would also to thank Mr. Megat Rosli and Mr Nurazminudin, from the Bernas Perdana Sdn. Bhd. Penang, Malaysia, for preparing rice husk powder samples.

REFERENCES

- [1]. English B., Chow P., Bajwa D.S., Processing into composites. In: Rowell R.W., Young R.A., Rowell J.K., Paper and composites from agro-based resources. New York, USA: Lewis Publishers 1997. pp 268 – 300.
- [2]. Sain M., Park S.H., Suhara F., and Law S., “Flame retardant and mechanical properties of natural fibre – PP composites containing magnesium hydroxide,” *Polym. Degradation Stability*, **83**, 363 – 367, 2004.
- [3]. IRRI 2004; www.irri.org
- [4]. Rozman H.D., Yeo Y.S., Tay G.S., and Abu Bakar A., “The mechanical and physical properties of polyurethane composites based on rice husk and polyethylene glycol,” *Polym. Testing*, **23**, 617 – 623, 2002.
- [5]. Hattotuwa G.B.P., Ismail H., and Baharin A., “Comparison of the mechanical properties of rice husk powder filled polypropylene composites with talc filled polypropylene composites,” *Polym. Testing*, **21**, 833 – 839, 2002.
- [6]. Yang H.S., Kim H.J., Son J., Park H.J., Lee B.J., and Hwang T.S., “Rice-husk flour filled polypropylene composites; mechanical and morphological study,” *Composites Structures*, **63**, 305 – 312, 2004.
- [7]. Ismail H and Abdul Khalil H. P. S., “The effects of partial replacement of oil palm wood flour by silica and silane coupling agent on properties of natural rubber composite,” *Polym. Testing*, **20** (1), pp 33-41, 2000
- [8]. Geethamma V.G., Joseph R., and Thomas S., “Short coir fiber-reinforced natural rubber composites: Effects of fiber length, orientation, and alkali treatment,” *J Appl. Polym. Sci.*, **55**, 583 – 594, 1995.
- [9]. Jacob M., Thomas S., and Varughese K.T., “Mechanical properties of sisal/oil palm hybrid fiber reinforced natural rubber composites,” *Composites Sci. Technol.*, **64**, pp 955-965, 2004
- [10]. Poh B.T. and Wong K.W., “Effect of blend ratio on Mooney scorch time of rubber blends,” *J. App. Polym. Sci.*, **69**, 1301-1310, 1998
- [11]. Ismail H. and Leong H.C., “Curing characteristics and mechanical properties of natural rubber/chloroprene rubber and epoxidized natural rubber/chloroprene rubber blends,” *Polym. Testing*, **20**, 509 – 516, 2001.
- [12]. Ramesan M.T., Mathew G., Kuriakose B., and Alex R., “Role of dichlorocarbene modified styrene butadiene rubber in compatibilisation of styrene butadiene rubber and chloroprene rubber blends,” *Eur. Polym. J.*, **37**, 719 – 728, 2001.
- [13]. Ismail H., and Nordin R., “Effect of Epoxidised Natural Rubber (ENR) and Ethylene co Acrylic Acid (EAA) Copolymer on Properties of Silica Filled Natural Rubber/Recycle Rubber Powder Blends,” *Polym. Plast. Technol. Eng.*, **43**, 285 – 300, 2004.
- [14]. Poh B.T. and Wong K.W., “Effect of blend ratio on Mooney scorch time of rubber blends,” *J. App. Polym. Sci.*, **69**, 1301-1310, 1998.
- [15]. Ismail H., and Nordin R., “Effect of Epoxidised Natural Rubber (ENR) and Ethylene co Acrylic Acid (EAA) Copolymer on Properties of Silica Filled

- Natural Rubber/Recycle Rubber Powder Blends,” *Polym. Plast. Technol. Eng.*, **43**, 285 – 300, 2004.
- [16]. Johnson T. and Thomas S., “Effect of epoxidation on the transport behavior and mechanical properties of natural rubber,” *Polym.* **41**, 7511 -7522, 2004.