



Chiang Mai J. Sci. 2008; 35(1) : 141-149

www.science.cmu.ac.th/journal-science/josci.html

Contributed Paper

Effect of Filler Loading on Curing Characteristics and Mechanical Properties of Thermoplastic Vulcanizate

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Received : 17 October 2007

Accepted : 30 October 2007

ABSTRACT

The curing characteristics and mechanical properties of natural rubber/polypropylene blends were investigated with the aim of improving tensile strength. Two different types of filler were tested: carbon black and reclaimed rubber. It was found that, the increasing carbon black compositions improve the tensile strength of the NR/PP blends. Reclaimed rubber shows beneficial effect by increasing the scorch time and decreasing the cure time of the NR/PP blends. And the use of reclaimed rubber results in compositions of improved elasticity and tensile mechanical performance.

Keywords: Curing characteristics, mechanical property, natural rubber/polypropylene blend.

1. INTRODUCTION

Thermoplastic elastomers (TPEs) are polymeric materials, which combine the excellent processability of the thermoplastic materials at high temperatures and a wide range of physical properties of elastomers at service temperature [1]. TPE grades are often characterized by their hardness. Olefinic thermoplastic vulcanizates (O-TPVs) are one of a class of TPE. These materials are composed of vulcanized rubber component in a thermoplastic olefinic matrix. O-TPVs have a continuous thermoplastic phase and a discontinuous vulcanized rubber phase. O-TPVs are dynamically vulcanized during a melt-mixing process in which vulcanization of rubber polymer takes place. O-TPVs's principal uses are automotive applications,

appliance uses, building/constructions, prominent electrical uses, business machines and uses in healthcare application [2-3]. Natural rubber (NR) has good resilience, high tensile strength, low compression set, resistance to wear and tear and good electrical properties. For these reasons, NR is usually used for wide range of application. Polypropylene (PP) due to its intrinsic properties such as translucent, good chemical resistance, tough, good fatigue resistance, integral hinge property, good heat resistance. It does not present stress-cracking problems and offers excellent electrical at higher temperatures. These include a lower density, higher softening point (PP doesn't melt below 160 °C) and higher rigidity and hardness. Easy incorporation of high loadings

of fillers and reinforcing agents, and ability to produce blends with other polymers including rubbers makes PP versatile [3-4]. Fillers are incorporated into polymer matrix mainly to achieve improvement of service properties or to reduce the material cost. Ismail and Suryadiansyah studied the effects of filler loading on tensile properties of PP-NR-RR composites. The results indicated that the tensile strength increase with increasing carbon black [5]. The effect of partial replacement of natural rubber (NR) by reclaimed rubber (RR) in natural rubber/polypropylene (NR/PP) thermoplastic elastomer blends was investigated. It is shown that replacement of up to half of the NR by RR is possible without altering the mechanical properties of the blend to any significant extent. Furthermore, the behavior of RR-NR/PP blends was similar to that of NR/PP on remolding; both systems retained their mechanical properties for up to four remolding cycles at 180 °C [6]. Recycled rubber (RR) was used to prepare polypropylene (PP)/RR blends with different RR content. A similar series of blends using natural rubber was also prepared. The results indicated that at similar rubber content, PP/RR blends have higher, tensile strength and Young's modulus but lower elongation at break and stabilization torque than PP/NR blends. Scanning electron microscopy (SEM) examination of the tensile fracture surface of PP/RR blends indicates that a higher energy is needed to cause catastrophic failure compared to PP/NR blends [7].

2. MATERIALS AND METHODS

2.1. Materials

The natural rubber (NR) sample used in this study was the product of Thailand. The polypropylene (PP) sample (grade 7500J) used in this study had the melt index of 12 and the bulk density of 0.905 g/cm³. Blending of NR and PP by NR/PP compositions of 80/20

by weight, was performed in a Brabender plasticorder (E-350, free volume =300 cm³, fill factor =0.85) with a fixed rotor speed of 50 rpm at 160 °C. A compatibilizer (polystyrene-modified natural rubber: SNR) was added. The formulation of the compound (that used a sulfur conventional vulcanization system), in parts per hundred rubber (phr), was binder 100, zinc oxide (ZnO) 5, stearic acid 2, MBTS 2.5, TMTD 2, DPPD 1, Wax 1 and sulphur 3. In this study, the effect of carbon black and reclaimed rubber content was of main interest. So, filler contents were steadily increased from 0 to 50 phr. The sample descriptions are summarized in [Table 1](#).

Table 1. Compound formulation used in NR/PP blends.

Componen.nts	phr	role
NR 20	80	Binder
PP	20	Binder
SNR	10	Compatibilizer
Reclaimed rubber	0-50	Filler
Carbon black	0-50	Filler.
Zinc oxide	5	Activator
Stearic acid	2	Activator
MBTS	2.5	Accelerator
TMTD	1.5	Accelerator
DPPD	2	Antioxidant
Wax	1.5	Antiozonant
Sulphur	3	Curing agent

2.2 Mixing and measurements of cure characteristics

Mixing was carried out using a laboratory two-roll mill in accordance with ASTM method designation D 3184-80. The cure characteristics of the mixes were determined

by using a Monsanto Rheometer model MDR 2000 at 140 °C. The respective cure time (t_{90}), and scorch times (t_2) values were obtained from the rheograph. The compounds were then compression molded at 140 °C based on respective t_{90} values.

2.3 Measurement of mechanical properties

Specimens for tensile and tear testing were punched out from the vulcanized sheets. Tensile testing was done as per ASTM D412, using dumb-bell specimens and at a rate of

grip separation of 500 ± 50 mm/min. For the hardness test, a Shore A type durometer was employed to find out the hardness of the vulcanizates. The method adopted is the same as that of ASTM D 2240 and the readings were taken after 5 s of indentation. Resilience was done according to ASTM D 2632 using a Vertical Rebound. Figure 1, that plot of stress versus strain. Measures the area underneath the stress-strain curve called toughness. Toughness is really a measure of the energy a sample can absorb before specimen breaks.

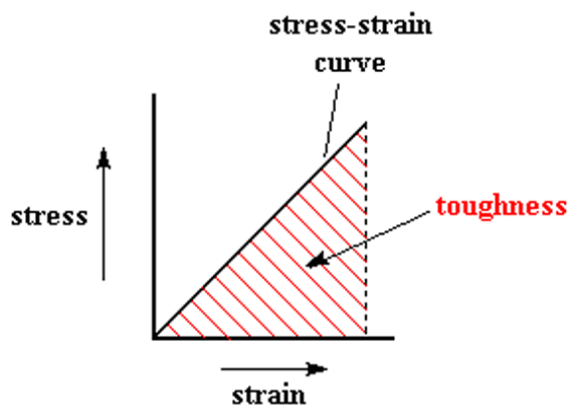


Figure 1. the plot of area underneath the stress-strain curve.

3. RESULTS AND DISCUSSION

3.1 Curing characteristics and mechanical properties of varied carbon black content.

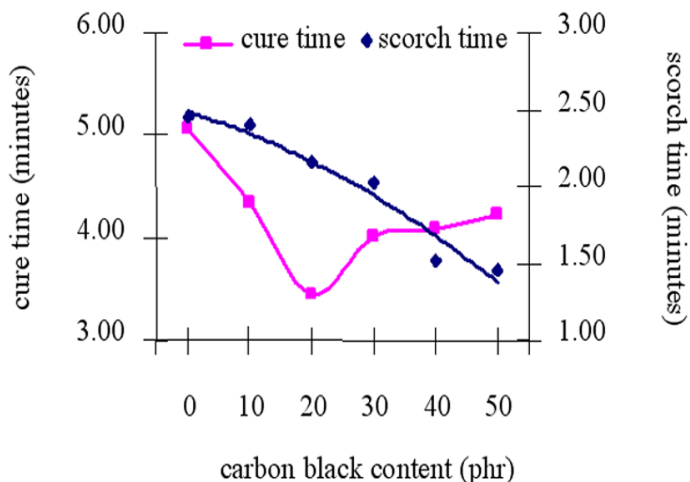


Figure 2. The effect of carbon black content on the cure time and scorch time of NR/PP blends.

Figure 2 demonstrates that the viscosity of the carbon black filled NR/PP blends increase with increasing carbon black loading, so the cure time tended to be dependent of carbon black content. And found a decrease

in scorch time with increasing carbon black content. This is due to the restriction of mobility and deformability of the matrix with the introduction of mechanical restraint [5].

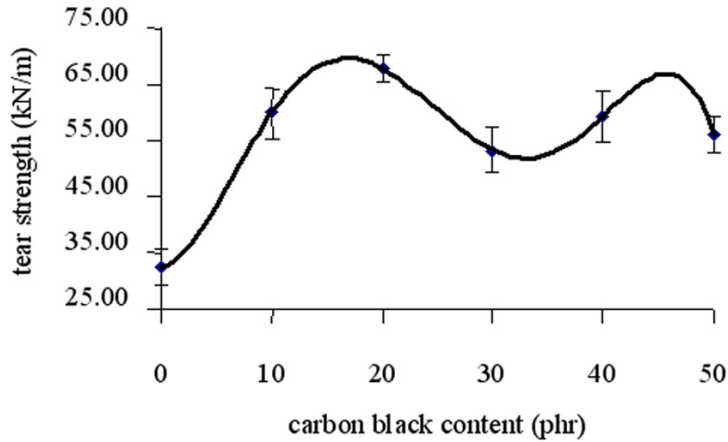


Figure 3. The relationship between tear strength and RR contents.

Engineering applications of thermo-plastic elastomer necessitates higher performance reliability which depends on various failure properties, including tear. Tear strength is a measure of the resistance to failure of a material when it is subjected to continue stretching. Higher tear resistance means better toughness. The related tear strength is given in

Figure 3, an optimum crosslinked NR-carbon black filled surface shows proper polymer-filler interaction. The matrix in the case of optimum cross-linking is properly restrained and the tear strength is enhanced. However, at an excess crosslink density level, the matrix become stiff and failure becomes brittle in nature [8].

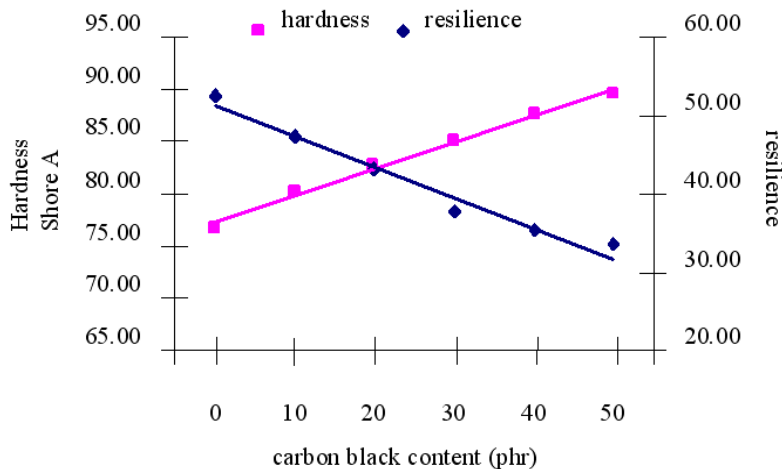


Figure 4. The effect of carbon black content on the mechanical properties of NR/PP blends.

Figure 4 shows the hardness and resilience of the NR/PP blends as a function of filler loading. It can be seen that hardness increase with increasing carbon black loading. In other word, the resilience of carbon black filled NR/PP blends decrease with increasing

carbon black loading. It inferred that carbon black could restrict the mobility and deformability of the matrix macromolecules at higher degree with increasing carbon black loading [5].

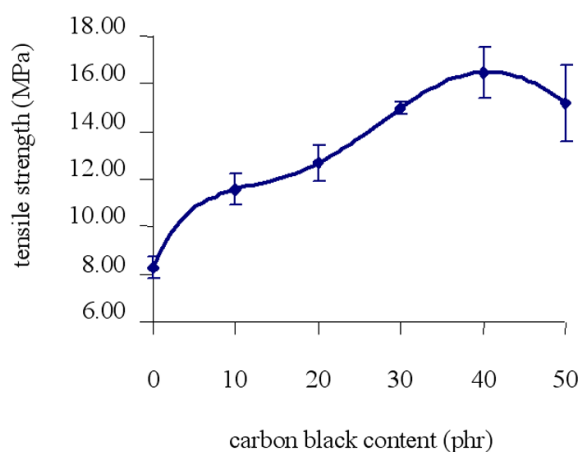


Figure 5. The effect of carbon black content on the tensile strength of NR/PP blends.

The effect of various filler loading on tensile strength of carbon black filled NR/PP blends is shown in Figure 5. It can be seen that the tensile strength increase with increasing carbon black content. This observation is due to the carbon black (N330) is reinforcing filler

which has good surface activity, chemical properties and non-uniform of porous surface which contribute to maximum interphase interaction between polymer chain and filler [5].

Table 2. Toughness values as a function of carbon black content

Carbon black content (phr)	toughness (N.mm/mm ³)
0	822.54
10	1333.36
20	1640.74
30	1295.56
40	1609.76
50	1100.47

The dependence of toughness of NR/PP blends on the filler loading is shown in Table 2. It is obvious that the toughness tended

to be dependent of the tear strength. Higher tear resistance means better toughness [8].

3.2 Curing characteristics and mechanical properties of varied reclaimed rubber content.

Figure 6 demonstrates that curing characteristics of the reclaimed rubber filled NR/PP blends indicated that an increase in the reclaimed rubber content decreased the cure time whereas the scorch time increased.

According to De et al., a decrease in cure time with increasing reclaimed rubber content was found owing to the presence of active crosslink sites in the reclaimed rubber [9]. And the blends with reclaimed rubber exhibit longer scorch time; this indicates that the incorporation of reclaimed rubber increases the processing safety of the blends.

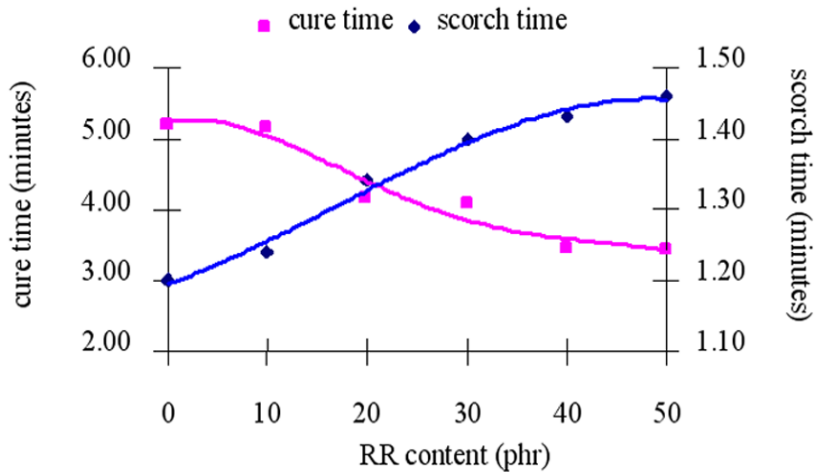


Figure 6. The effect of RR content on the cure time and scorch time of NR/PP blends.

Figure 7 shows the effect of reclaimed rubber content on the mechanical properties of NR/PP blends. Addition of a master batch of reclaimed rubber improved the adhesion between filler and matrix phase, so it could increase the resilience property of the blends.

Reclaimed rubber master batch is a softer material, results in decrease in flexural strength and hardness. Addition of reclaimed rubber master batch increase the total elastomeric content and, therefore, has a more pronounced effect on them [10].

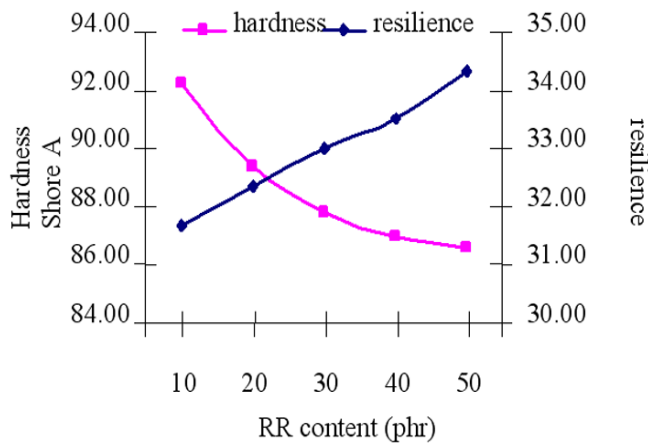


Figure 7. The effect of RR content on the mechanical properties of NR/PP blends.

The effect of various reclaimed rubber loading on mechanical properties of NR/PP blends are shown in Figure 8-9. The tensile and tear failure of these blends were improved with increasing in degree of cross-linking of rubber phase [11].

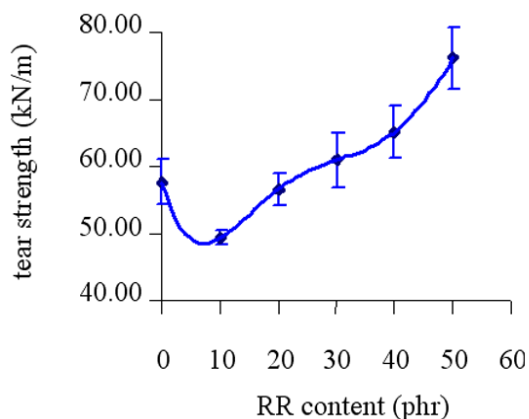


Figure 8. The relationship between tear strength and RR content.

The use of reclaimed rubber results in compositions of improved elasticity and tensile mechanical performance. This finding was attributed to interfacial entanglement and co-crosslinking phenomena [12].

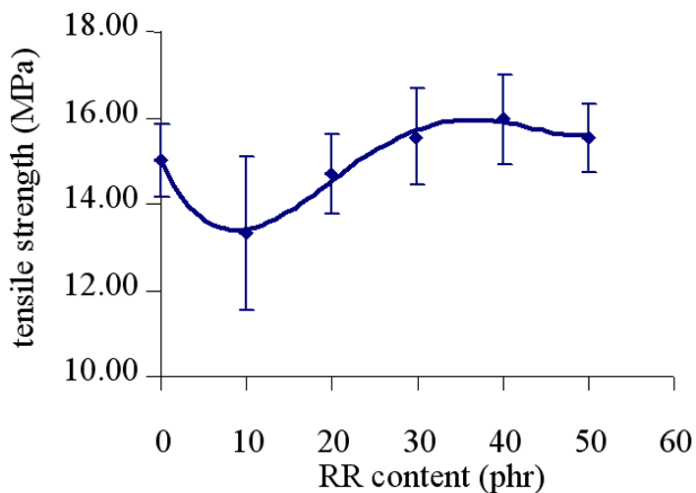


Figure 9. The effect of RR content on the tensile strength of NR/PP blends.

Table 3 shows the toughness values as a function of reclaimed rubber content, the area under the stress-strain curve increased after the reaction demonstrating that energy absorbed by the specimen before fracture also increased.

Table 3. Toughness values as a function of RR content

reclaimed rubber (phr)	tensile strength (MPa)	toughness (N.mm/mm ³)
0	15.02	647.63
10	13.33	621.32
20	14.71	976.48
30	15.56	1102.36
40	15.97	1224.93
50	15.55	1196.46

It was also observed that in cross-section of tensile specimen of NR/PP blends. This change of behavior may be explained by the increase of interfacial adhesion and increase of interaction between the stress concentrate zones in the matrix. An increase of interfacial adhesion suppresses production of voids or flaws in the matrix [13].

4. CONCLUSIONS

From this study, the following conclusions can be drawn:

(1) The increasing carbon black compositions improve the tensile strength of the NR/PP blends.

(2) Reclaimed rubber shows beneficial effect by increasing the scorch time and decreasing the cure time of the NR/PP blends.

(3) The use of reclaimed rubber results in compositions of improved elasticity and tensile mechanical performance.

ACKNOWLEDGEMENT

The authors are grateful to the scholarship of Graduate School, Prince of Songkla University and the Thailand Research Fund (TRF) under Contract No.RDG4850033.

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