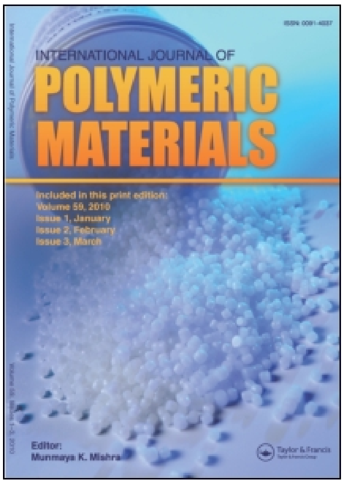

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The Effect of Filler Loading on Curing and Mechanical Properties of Natural Rubber/recycled Rubber Powder Blends

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THE EFFECT OF FILLER LOADING ON CURING AND MECHANICAL PROPERTIES OF NATURAL RUBBER/RECYCLED RUBBER POWDER BLENDS

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Curing characteristics and mechanical properties of carbon black– and silica-filled natural rubber (NR)/recycled rubber powder (RRP) blends were studied. Results indicate that the minimum torque and maximum torque increase with increasing filler loading in the compounds, whereas scorch time shows a decreasing trend. Cure time of carbon black–filled NR/RRP blends decreases with increasing filler loading whereas silca-filled NR/RRP blends show an opposite trend. Incorporation of filler loading has improved the tensile modulus, hardness, tear strength, and resistances toward swelling. However, elongation at break and resilience exhibit a different trend. For tensile strength, optimum values were obtained at 15 phr of both fillers. Overall results show that carbon black (N550) is more suitable to be used as a filler in natural rubber/recycle rubber powder blends compared to silica (Vulcasil S).

Keywords: cure characteristics, mechanical properties, recycle rubber powder, silica, carbon black

INTRODUCTION

Accumulations of waste rubber have created major environmental problems, as waste rubber is non-degradable material. This usual method of disposing of waste rubber, such as discarding in a landfill or burning,

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are not suitable since they causes severe environmental pollution and are uneconomical [1]. Thus an approach to reutilize waste rubber, for a better disposal and use, is needed. An approach to reutilize waste rubber can be done by mechanical [2–3] and chemical processes [4–5]. However, more researchers [6–10] tend to reutilize waste rubber by mechanical process, as it's more cost effective compared to chemical process.

Recently the authors reported the potential of recycle rubber powder as a filler in natural rubber compounds [11] and the effect of partial replacement of natural rubber with recycle rubber powder [12]. In both studies, the results indicate that the curing characteristics and mechanical properties were not much altered by the incorporation of recycled rubber powder in natural rubber compounds. Therefore, the use of recycled rubber powder in natural rubber compounds has a potential to partially replace natural rubber compounds. However, the effect of various fillers in natural rubber/recycled rubber powder blends should not be neglected as natural rubber/recycled rubber powder blends alone are not enough to fulfill the product specifications in the rubber industry. Therefore, the purpose of this study is to determine the curing characteristics and mechanical properties of carbon black– and silica-filled natural rubber/recycled rubber powder blends. Three different filler loadings were used, such as 15, 30, and 50 phr.

EXPERIMENTAL

Materials

Natural rubber (SMR L) was purchased from Kumpulan Guthrie Sdn. Bhd., Seremban, Malaysia. The recycled rubber waste (powder) product from the sanding process (polishing) of rubber ball and artifical eggs was obtained from Watas Holding (M) Sdn. Bhd., Penang, Malaysia. In this study, the particle size of recycled rubber powder was in the range of 250–500 μm, carbon black in the range of 39–55 nm (N550), and silica (Vulcasil S) in the range of 11–19 nm. Table 1 shows the formulation used in this study.

Cure Characteristics

Cure characteristics were studied using a Monsanto Moving Die Rheometer (MDR 2000) according to ASTM D 224. About 4 g samples of the respective compound were tested at a vulcanization temperature (150°C). The Mooney viscosity was determined by using a Monsanto automatic Mooney Viscometer (MV 2000) at 120°C. The testing procedure was conducted according to the method described in ASTM D

TABLE 1 Formulation of Compounds

Ingredient	phr	
Natural Rubber (SMR L)	50	50
Recycled Rubber Powder (RRP)	50	50
Zinc oxide	5.0	5.0
Stearic acid	2.0	2.0
Sulphur	2.5	2.5
N-cyclohexyl-2-benzothiazole Sulfanamide	0.6	0.6
2,2-methylene-bis-(4-methyl-6-tertbutylphenol)	2.0	2.0
Carbon black (N550)	0, 15, 30, 50	—
Silica (Vulcasil S)	—	15, 30, 50

1646.3 mm thick sheets were compression molded at 150°C with force of 10 MPa using a hot press according to the respective cure time, t_{90} determined with the MDR 2000.

Swelling Study

Swelling of natural rubber/recycled rubber powder blends was studied in toluene and ASTM Oil No. 1 according to ASTM D 471. Cured test pieces of compounds of dimension 30 × 5 × 2 mm were weighed using an electrical balance and this was considered to be original weight (M_1). Calculation of the swelling percentage was as follows:

$$\text{Swelling percentage} = \{(M_2 - M_1)/M_1\} \times 100 \tag{1}$$

where M_1 = initial mass of specimen, g, and M_2 = mass of specimen, g, after immersion.

Mechanical Properties

Dumbbell shaped samples were cut from the molded sheets according to ASTM D 3182. Tensile and tear tests were performed at a crosshead speed of 500 mm/min using a Monsanto Tensometer M500 according to ASTM D 412 and ASTM D 624 (die B), respectively. 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). Resilience was studied using a Wallace Dunlop Tripsometer according to ASTM D 1054-91 whereas rebound resilience was calculated according to the following equation:

$$\text{Percentage resilience} = \{(1 - \cos\theta_2)/(1 - \cos\theta_1)\} \times 100 \tag{2}$$

where θ_1 = initial angle (45°) and θ_2 = maximum rebound angle.

RESULTS AND DISCUSSION

Curing Characteristics

Table 2 shows the curing characteristics of carbon black– and silica-filled natural rubber/recycled rubber powder blends. It can be seen that carbon black–filled NR/RRP blends show decreasing trend of cure time with increasing filler loading whereas silica-filled NR/RRP RRP blends exhibit an opposite trend. It is believed that the ability of silica to react with curative ingredients [13–15] leads to depletion of the amount of curing agents in the blends. Therefore, it will take a longer time to vulcanize silica-filled natural rubber/recycled rubber powder compounds. On the other hand, increasing loading of carbon black in natural rubber/recycled rubber powder compounds decreases the cure time of the blends. This indicates that carbon black has the ability to activate the vulcanization process in natural rubber/recycled cycled rubber powder compounds. Hilyard et al. [16] reported that carbon black has the ability to activate the vulcanization process through the promotion of hydrogen sulfide formation and the rupture of S–N linkage when heated with sulfanamides in rubber compounds either in the presence or absence of curing ingredients.

It is also obvious that increasing filler loading has reduced the scorch time of natural rubber/recycled rubber powder blends (Table 2), especially with silica-filled blends. An insufficient amount of accelerator or curing agent and migration of sulphur from natural rubber to recycled rubber powder [17–19] might be the reasons for reduction of scorch time.

The effect of filler loading on minimum torque of natural rubber/recycled rubber powder blends is shown in Table 2. It is clear that incorporation of fillers in natural rubber/recycled rubber powder blends has increased the minimum torque. However, at a similar filler loading, silica-filled natural rubber/recycled rubber powder blends

TABLE 2 Curing Characteristics of Carbon Black and Silica-filled Natural Rubber/Recycled Rubber Powder Compounds

	Carbon black (phr)				Silica (phr)		
Curing characteristics	0	15	30	50	15	30	50
Cure time (min)	8.23	7.55	7.34	6.91	17.48	25.41	28.24
Scorch time (min)	1.82	1.79	1.71	1.49	1.04	0.15	0.08
Minimum torque (d Nm)	0.27	0.40	0.90	3.25	0.73	8.34	25.65
Maximum torque (d Nm)	13.04	16.95	23.34	32.22	19.4	33.04	55.00

show higher minimum torque than carbon black–filled natural rubber/recycled rubber powder blends. Strong filler-filler interaction of silica has contributed to the higher values of minimum torque. Meanwhile, for carbon black, an increase of minimum torque was mainly attributed by polymer-filler interaction rather then filler-filler interaction [20–22]. As minimum torque is related to viscosity and processability of compounds, the processing of compounds becomes more difficult with increasing amount of fillers. A measurement of Mooney viscosity from Mooney viscometer (Figure 1) shows that silica-filled natural rubber/recycled rubber powder compounds exhibit higher value of Mooney viscosity compared to carbon black. Therefore processing of silica is much more difficult compared to carbon black particularly at filler loading more than 30 phr.

A similar trend was observed for maximum torque as carbon black and silica loading increase in natural rubber/recycled rubber powder blends (Table 2). Because maximum torque is correlated to modulus, polymer-filler interaction and filler-filler interaction will contribute to the value of maximum torque. It is clear that, at a similar filler loading, silica filled natural rubber/recycled rubber powder blends show higher maximum torque value than carbon black–filled natural

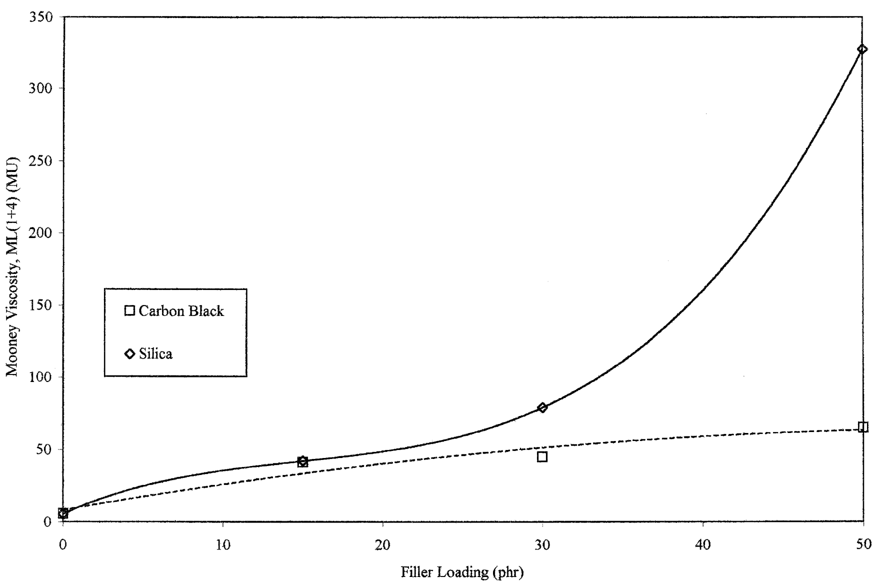

FIGURE 1 The effect of filler loading on Mooney viscosity, ML $(1+4)$ of natural rubber/recycled rubber powder blends.

rubber/recycled rubber powder blends. This might be due to the tendency of silica to have strong filler-filler interactions compared to carbon black [23].

Mechanical Properties

The effect of filler loading on tensile strength of natural rubber/ recycled rubber powder blends is shown in Figure 2. It can be seen that the optimum tensile strength occurred at 15 phr of both fillers. At a similar filler loading, carbon black–filled natural rubber/recycled rubber powder blends exhibit higher tensile strength than silica-filled natural rubber/recycled rubber powder blends. It's believed that uniform dispersion of filler and better filler-natural rubber/recycled rubber powder interaction enhance the tensile strength of natural rubber/recycled rubber powder compounds. However, when more than 15 phr was used, the deterioration of tensile strength was caused by decreasing polymer-filler interactions and increasing of filler-filler interactions (agglomeration) in the blends, particularly silica-filled natural rubber/recycled rubber powder blends. The other

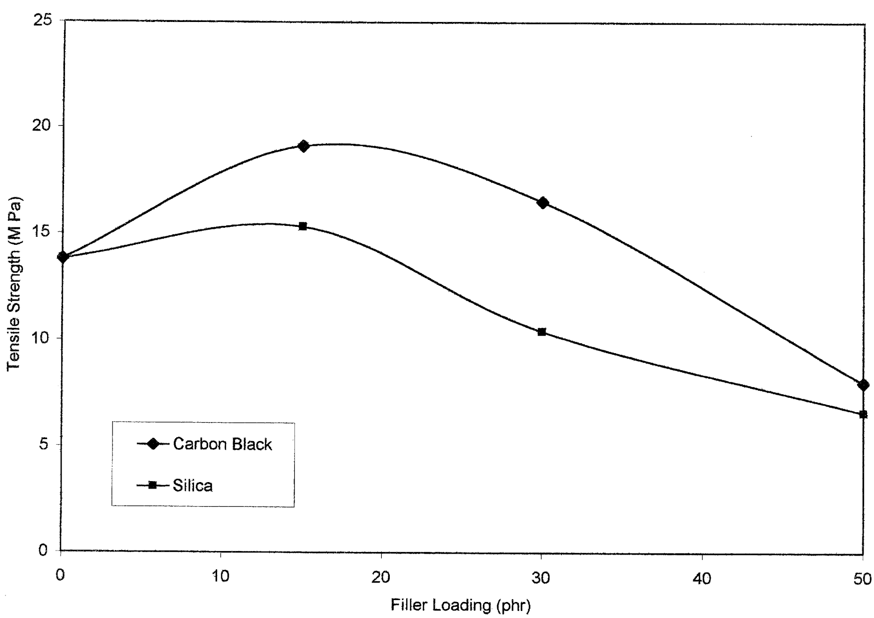

FIGURE 2 The effect of filler loading on tensile strength of natural rubber/ recycled rubber powder blends.

possibility of reduced tensile strengths after 15 phr of filler is due to the presence of recycled rubber powder (already crosslink) in the blends, which might also weaken the filler-natural rubber/recycled rubber powder interaction.

Figure 3 shows the effect of filler loading on tear strength of silica and carbon black–filled natural rubber/recycled rubber powder blends. Although the tear strength increases slightly with increasing filler loading, the tear strength of carbon black–filled natural rubber/recycled rubber powder blends still exhibits better strength than silica-filled natural rubber/recycled rubber powder blends. It is clear that carbon black N550 is more suitable to use as a filler in natural rubber/recycled rubber powder blends compared to silica (Vulcasil S). This might be due to the absence of a silane coupling agent, which is needed to couple the non-polar natural rubber/recycle rubber powder matrix and polar silica. The study of silane coupling agent in silica-filled natural rubber/recycled rubber powder blends will be carried out in the near future.

The variations of elongation at break and tensile modulus, M100 (stress at 100% elongation) with filler loading are shown in Figures 4 and 5. It can be seen that at a similar filler loading, silica-filled natural

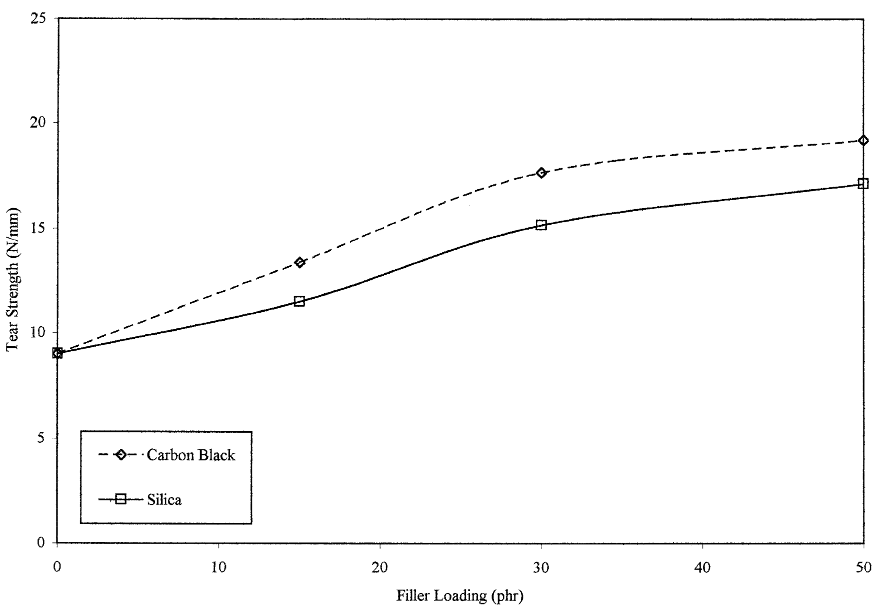

FIGURE 3 The effect of filler loading on tear strength of natural rubber/recycled rubber powder blends.

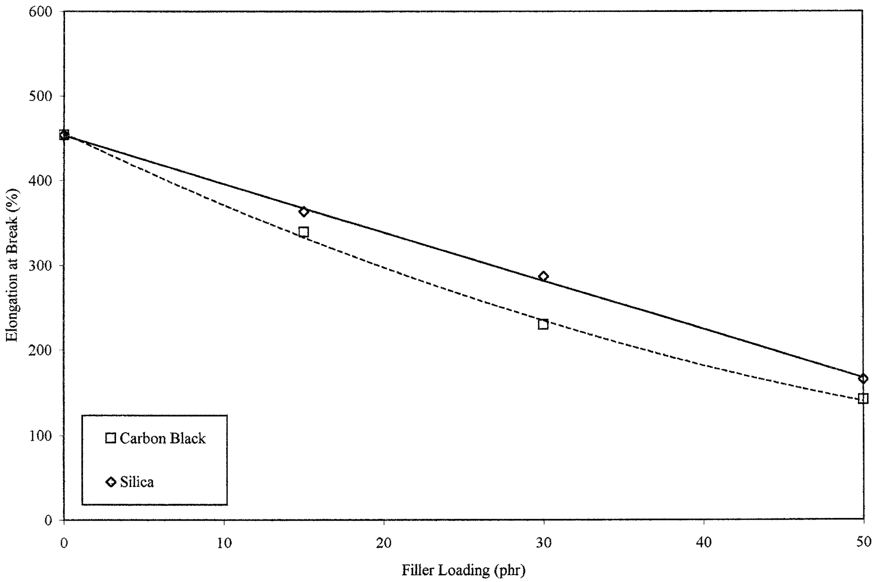

FIGURE 4 The variation of elongation at break and filler loading of natural rubber/recycled rubber powder blends.

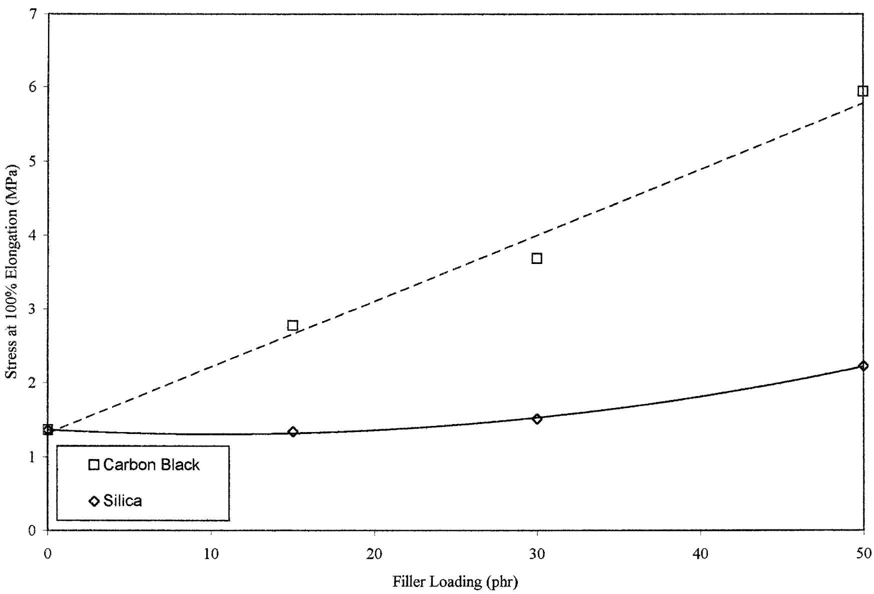

FIGURE 5 The effect of filler loading on stress at 100% elongation of natural rubber/recycled rubber powder blends.

rubber/recycled rubber powder compounds give higher values of elongation at break and lower values of M100 compared to carbon black. This is expected as carbon black has strong polymer-filler interaction, which restricts the mobility of natural rubber and recycled rubber chains. For silica, a weak polymer-silica interaction led to lower value of M100 and higher value of elongation at break than carbon black–filled NR/RRP blends.

Figures 6 and 7 show the effect of filler loading on the hardness and resilience of natural rubber/recycled rubber powder blends. It is clear that increasing of filler loading increased the hardness values. As hardness depends on the behavior of particulate fillers in the compounds, filler that has strong filler-filler interaction or agglomeration such as silica (high agglomeration especially in low polarity elastomer) will induce higher hardness value compared to carbon black. As hardness increases, reduction in resilience will occur (Figure 7) [24].

Figures 8 and 9 show the effect of filler loading on swelling percentage of natural rubber/recycled rubber powder compounds in toluene (after 24 h) and in ASTM Oil No. 1 (after 168 h), respectively. Both figures indicate that carbon black–filled natural rubber/recycled rubber

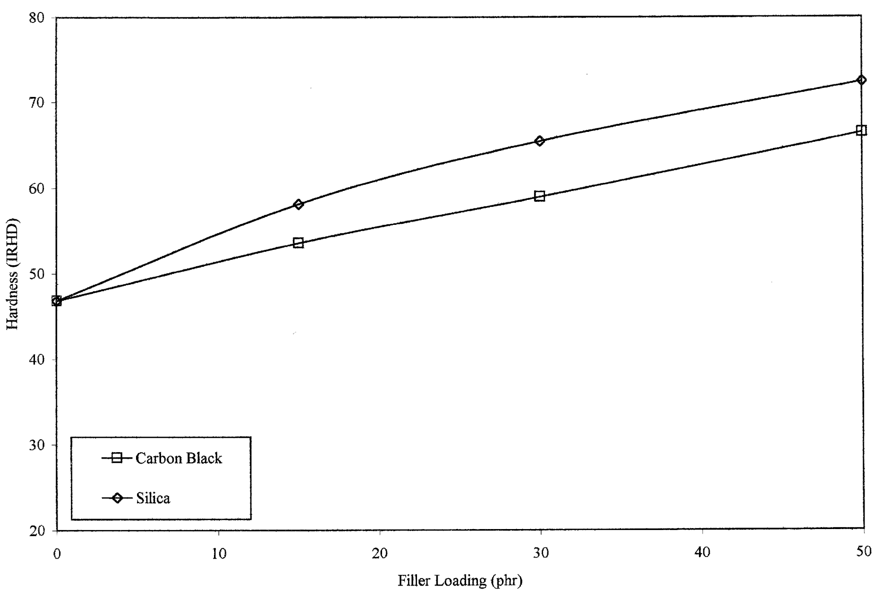

FIGURE 6 Relationship between filler loading and hardness of natural rubber/recycled rubber powder blends.

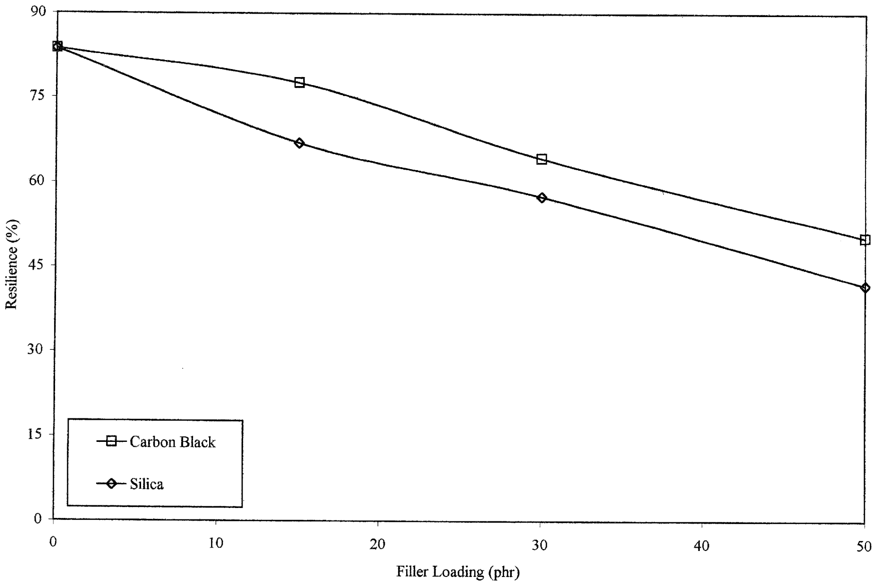

FIGURE 7 Relationship between resilience and filler loading of natural rubber/recycled rubber powder blends.

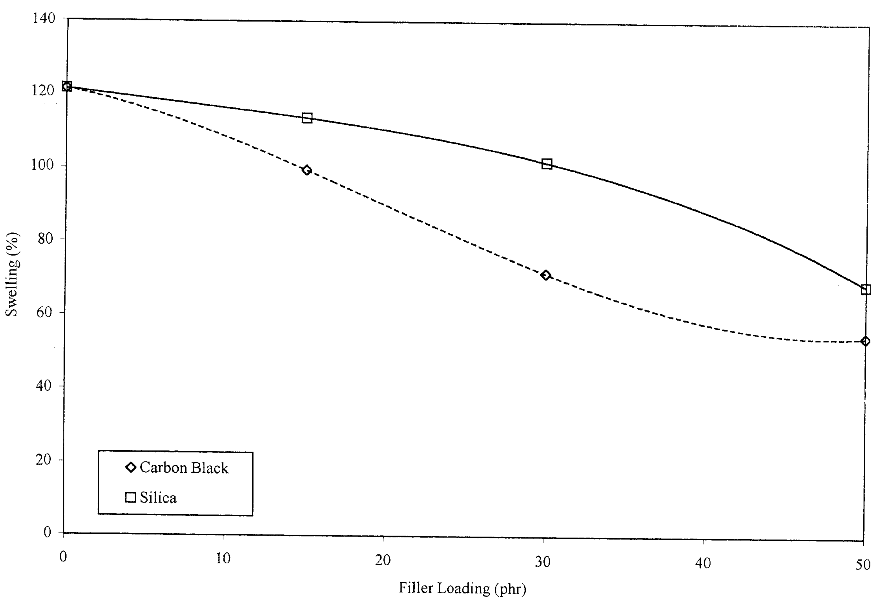

FIGURE 8 Relationship between filler loading and equilibrium of swelling (in toluene after 24 hours) of natural rubber/recycled powder blends.

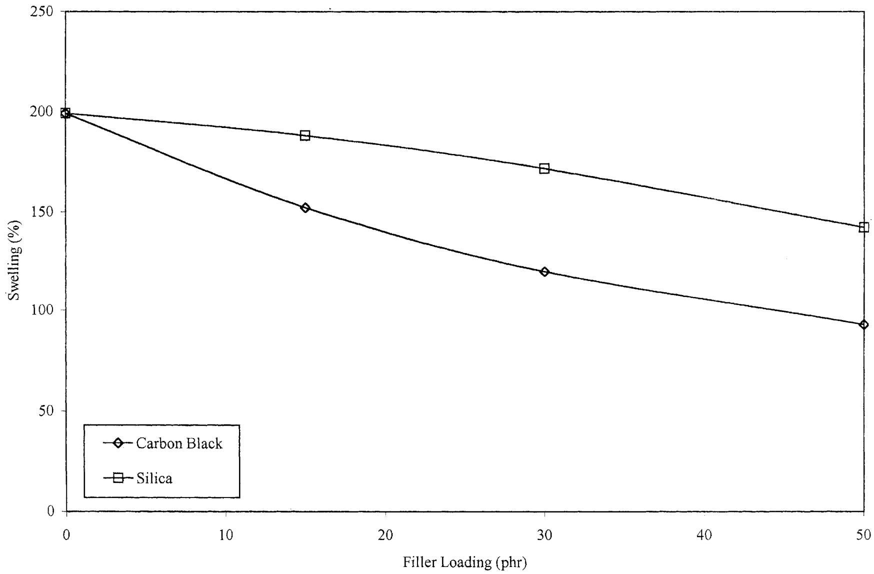

FIGURE 9 Relationship between filler loading and equilibrium of swelling (in ASTM Oil No. 1 after 168 hours) of natural rubber/recycled rubber powder blends.

powder blends give better resistance towards penetration of solvents and oil compared to silica. As discussed before this is due to better carbon black–natural rubber/recycled rubber powder interaction compared to silica–natural rubber/recycled rubber powder interaction.

CONCLUSIONS

From this study, the following conclusions can be drawn:

1. The cure time and scorch time were affected by the increase of carbon black and silica loading in natural rubber/recycled rubber powder blends. At a similar filler loading, carbon black shows shorter cure time but longer scorch time compared to silica.
2. Incorporation of silica shows significant increment of minimum torque and Mooney viscosity compared to carbon black–filled natural rubber/recycled rubber powder blends.
3. Carbon black–filled natural rubber/recycled rubber powder blends show better tensile strength, tear strength, tensile modulus, and swelling resistance than silica-filled natural rubber/recycled rubber powder blends.

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