

Studies on Fly-Ash-Filled Natural Rubber Modified with Cardanol Derivatives: Processability, Mechanical Properties, Fracture Morphology, and Thermal Decomposition Characteristics

A. R. R. Menon, T. A. Sonia, J. D. Sudha

Regional Research Laboratory (CSIR), Thiruvananthapuram 695 019, Kerala, India

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ABSTRACT: Fly ash, a by-product of thermal power stations, was used as a filler in natural rubber (NR) in presence of 5–10 phr of phosphorylated cardanol prepolymer (PCP) and hexamethylene tetramine cured PCP (PCPHM). The compositions modified with the cardanol-based resins showed lower power consumption for mixing, lower cure time, improved tensile properties and tear strength, and higher thermal stability. Scanning electron microscopy of the fracture surfaces of the tensile-failed specimens showed finer and more uniformly

distributed filler particles in the rubber matrix in the presence of PCP/PCPHM. The cardanol-based resins are expected to function as a coupling agent between the filler and rubber leading to the improvement in mechanical properties. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 4801–4808, 2006

Key words: fly ash; natural rubber; phosphorylated cardanol prepolymer; mechanical properties; morphology; thermal stability; coupling agent

INTRODUCTION

Fly ash, a waste by-product generated abundantly in thermal power stations has been posing severe environmental problems the world over, particularly during the last decade. Consequently, it is capturing the interest of researchers and environmentalists to devise effective strategies for its disposal/reuse.^{1–6} Fly ash has been used to synthesize zeolites,^{1,2} in blends with cement for civil works,³ for treatment of waste water,^{4–5} and as a supplementary additive with organic wastes and fertilizers for improving the yield of agricultural crops.⁶

Apart from the various applications of fly ash mentioned above, it has recently been used as a filler in various polymer matrices.^{7–14} Thus, fly ash has been used as a filler in polyester,^{9–11,14} nylon,¹² and epoxy resins.¹³ Fly ash has also been used as a filler in various rubbers.^{15–20} Mishra et al. have used nano calcium carbonate (CaCO₃) as a reinforcing filler in polybutadiene rubber, in comparison with fly ash.¹⁵ They observed improvements in tensile strength, toughness, hardness, flammability, and tear strength for the nano CaCO₃ filled rubber, as compared to fly-ash-filled rubber. Sombatsompop et al. have recommended the use

of fly ash as a filler for NR compounds with silica contents of 0–30 phr.¹⁶ They have reported that the vulcanizate properties of fly-ash-silica-filled vulcanizates were similar to those of precipitated silica filled vulcanizates at silica contents of 0–30 phr. Also, NR compounds filled with fly-ash-silica were reported to have relatively low cure time and low viscosity. Garde et al. indicated that the mechanical properties of polyisoprene rubber loaded with fly-ash particles were inferior to those of polyisoprene filled with silica.¹⁷ Alkadasi et al. used fly ash treated with a silane coupling agent (Si-69) as a filler in polybutadiene rubber (BR).¹⁸ The rubber composites with treated fly ash showed better reinforcing properties. It has also been reported by the same authors that fly ash treated with a 2% solution of a titanate coupling agent could improve the mechanical properties of BR vulcanizates.¹⁹ Hundiwale et al. studied elastomer–filler interaction by incorporating calcium carbonate, barium sulphate, and fly ash as inert fillers in styrene butadiene rubber (SBR).²⁰ It was found that amongst the three fillers, barium sulphate contributes the least towards reinforcement, while the reinforcement caused by fly ash was almost equal to that caused by calcium carbonate. They have also studied the effect of fly ash as a filler in butadiene rubber, in comparison with calcium carbonate and talc.²¹

Phosphorylated cashew nut shell liquid (PCNSL)²² and phosphorylated cardanol prepolymer (PCP)²³ synthesized from cashew nut shell liquid (CNSL) and cardanol, respectively, which are renewable natural

Correspondence to: A. R. R. Menon (ravindranathamenon@yahoo.co.in).

resources obtained from the cashew industry, have been found to be suitable for various applications such as adhesives for bonding plywood, precursors for friction-modifying additives in automobile brake linings, foundry core oil binders, and flame-retardant plasticizers for plastics. Previous studies in unfilled mixes have shown that PCNSL and PCP²⁴ could function as multifunctional additives in natural rubber (NR) and synthetic rubbers in various roles as a crosslinkable plasticizer, a softener, a tackifier, an antidegradant, an improver of mechanical properties of vulcanizates (tensile strength, tear strength, and fatigue resistance), and a coupling agent between silica filler and NR, leading to improvement in resistance to crack propagation.²⁴⁻³⁰ It has been observed recently that at a dosage of 5 phr, PCP could function as a coupling agent between NR and silica particles, thereby improving the filler dispersion and consequently the mechanical properties of the vulcanizates.³⁰ This is expected to be due to the amphiphilic nature of PCP, with a nonpolar hydrocarbon side-chain and a polar phosphate group in the same molecule. The unsaturated hydrocarbon side chain fraction of PCP may become co-crosslinked with that of NR and the phosphate group of PCP can have polar-polar interaction, such as hydrogen bonding with the silanol groups on the surface of the silica particles, thereby acting as a coupling agent between the rubber and the filler. Silica is known to be the major constituent (46.25%) of the chemical composition of fly ash particles.¹⁶ Also, it is reported that the hydroxyl groups on the surface of silica particles can adsorb polar materials via hydrogen bonds³¹⁻³³ and can react with polar coupling agents.³⁴⁻³⁶ Thus, it is possible that PCP may act as a coupling agent between the polar silica particles of fly ash and the nonpolar NR, thereby improving the efficiency of fly ash as a reinforcing filler.

Hence, the present study is such an attempt to explore the effect of the cardanol derivatives, namely, PCP and hexamethylene tetramine cured PCP (PCPHM) on the processability and mechanical properties of fly-ash-filled NR systems.

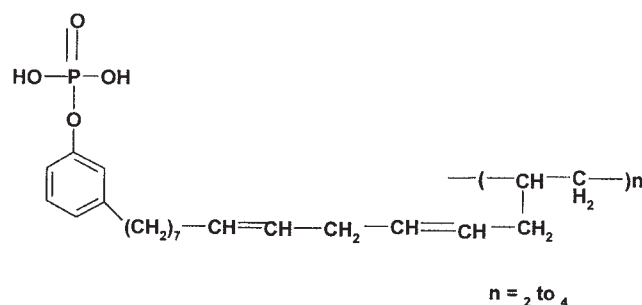
EXPERIMENTAL

Materials

PCP (I)—an oligomer of monocardanyl phosphoric acid—was synthesized at Regional Research Laboratory (CSIR), Thiruvananthapuram, according to a patented process.²²

Prepolymer of PCP

Hexamethylene tetramine cured PCP (PCPHM) was prepared by mixing PCP with HMTA (8%) in a Brabender Plasticorder at 120°C and at a rotor speed of



Scheme 1

10 rpm for 15 min. Fly ash having particle size ranging from 0.5 to 7 μm was obtained from National Thermal Power Corporation (Tuticorin, Tamil Nadu, India). Natural rubber (grade, RSS-V) and other rubber chemicals (laboratory reagent grades) were obtained locally.

Methods

Semi-efficient vulcanization systems of NR containing 0, 5, 10, 15, and 20 phr of fly ash and 5 and 10 phr each of PCP/PCPHM (compositions as given in Table I) were prepared by mixing in a Brabender Plasticorder at room temperature and at a rotor speed of 30 rpm for 20 min. The power consumption for mixing (P) was obtained (in $\text{m}\cdot\text{kg}$) from the equilibrium torque at the 15th minute (M) and the angular velocity of the rotor (ω), using the relation $P = M\omega$, where $\omega = 2\pi S/60$ with S the rotor speed (in rpm).

TABLE I
Composition of Mixes

Mix code ^a	Fly ash (phr)	PCP (phr)	PCPHM (phr)
FA0	0	—	—
FA0-PCP5	0	5	—
FA0-PCPHM5	0	—	5
FA5	5	—	—
FA5-PCP5	5	5	—
FA5-PCP10	5	10	—
FA5-PCPHM5	5	—	5
FA5-PCPHM10	5	—	10
FA10	10	—	—
FA10-PCP5	10	5	—
FA10-PCP10	10	10	—
FA10-PCPHM5	10	—	5
FA10-PCPHM10	10	—	10
FA15	15	—	—
FA15-PCP5	15	5	—
FA15-PCPHM5	15	—	5
FA20	20	—	—
FA20-PCP5	20	5	—
FA20-PCP10	20	10	—
FA20-PCPHM5	20	—	5
FA20-PCPHM10	20	—	10

^a Base mix: NR 100, ZnO₅, stearic acid 2, sulphur 2, and MBT 2.

TABLE II
Processability Characteristics of Fly-Ash-Modified (10 phr)
NR Containing Different Dosages of PCP/PCPHM

Mix code ^a	Power consumption for mixing, <i>P</i> (kW)	Cure time at 150°C (min)
FA10	7.444	11
FA10-PCP5	6.484	6
FA10-PCP10	6.084	7
FA10-PCPHM5	7.204	5
FA10-PCPHM10	7.204	5

^a Base mix: NR 100, ZnO₅, stearic acid 2, sulphur 2, and MBT 2.

The cure time of the mixes was measured at 150°C using the Brabender Plasticorder at a rotor speed of 30 rpm. Test samples were prepared by compression molding the above mixes at 150°C at the respective cure times.

Tensile properties of the vulcanizates were measured using a Universal Testing Machine (Hounsfield, Model H5 KT) at a crosshead speed of 500 mm/min as per ASTM D-412-1978. Tear strength of the samples were also measured on the same equipment as per ASTM D-624-86.

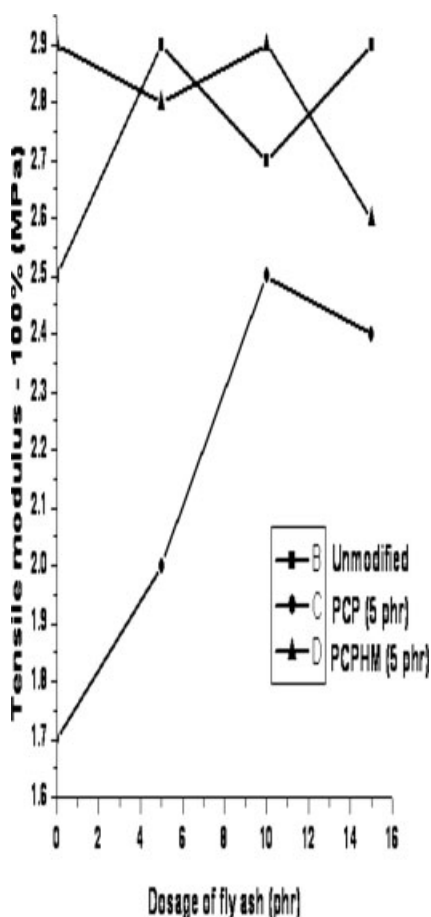


Figure 1 Effect of dosage of fly ash on tensile modulus of NR vulcanizates containing PCP/PCPHM.

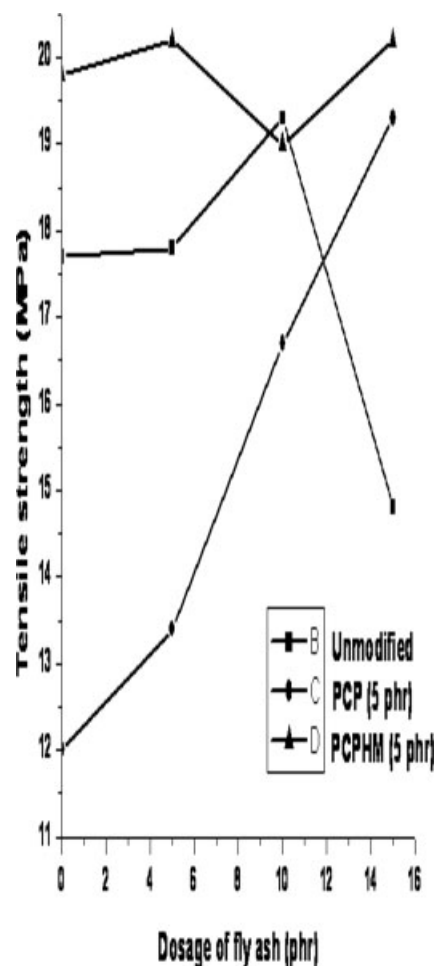


Figure 2 Effect of dosage of fly ash on tensile strength of NR vulcanizates containing PCP/PCPHM.

The morphology of the fracture surfaces of the specimens was studied using a scanning electron microscope (Model JEOL-JSM-5600LV SEM) after sputter coating the surfaces with a fine coater (Model JEOL-JFC 1200).

Thermogravimetric analysis (TGA) of the vulcanizates was carried out in nitrogen atmosphere at a heating rate of 20°C/min on a thermogravimetric analyzer (Model Shimadzu DTG 60).

RESULTS AND DISCUSSION

Processability characteristics

Mixing and curing/vulcanization constitute two of the critical unit operations in the manufacture of rubber products. It is essential that the power consumption/energy requirement in these stages is kept to a minimum from economic considerations.

Mixing

Table II shows the results on the processability characteristics of the mixes containing 10 phr of fly ash, as

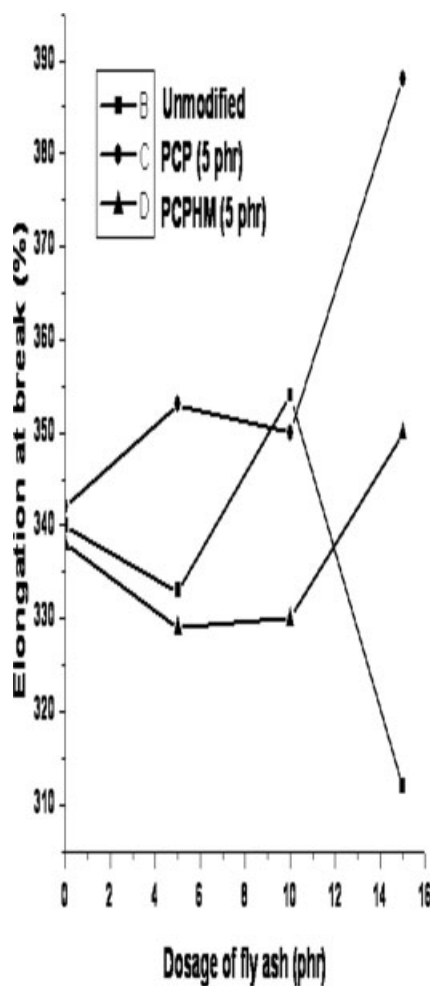


Figure 3 Effect of dosage of fly ash on elongation at break of NR vulcanizates containing PC /PCPHM.

obtained from the Brabender Plasticorder. It shows a steady decrease in the power consumption for mixing with the increase in dosage of PCP from 0 to 10 phr. The corresponding values for the mixes containing 5 and 10 phr of PCPHM are also lower than that of the unmodified mix. These results show the plasticizing effect of the cardanol-based resins in the fly-ash-filled NR mixes. This is expected to be due to the presence of the unsaturated aliphatic side-chain fraction of these resins which could enhance the segmental mobility of NR. It may be noted that the plasticizing efficiency of PCP is higher than that of PCPHM and that it increases with the increase in dosage of the resin from 5 to 10 phr. This is quite expected due to the higher degree of mobility of the molecules of the former, as compared to that of the latter which is crosslinked with hexamine. Plasticizers are known to increase the segmental mobility of the main chain of the polymer to which these are incorporated. The melt viscosity of a polymer at any specified temperature is often considered to be a measure of the mobility of the polymeric chains relative to each other and is controlled by the

flexibility of the chains.³⁷ 3-(tetrabromopentadecyl) 2,4,6 tribromo phenol (TBPTP)—a low-molecular-weight flame retardant derivative of cardanol—was reported to exhibit a plasticization effect in ethylene vinyl acetate copolymers, as indicated by considerable reduction in the power consumption for mixing.³⁸ The higher plasticization effect of PCP in NR is comparable to this. However, it can be noted that the power consumption for mixing does not decrease with the increase in dosage of PCPHM from 5 to 10 phr. This could be due to the reduced mobility of the side chain fraction of PCPHM resulting from intramolecular crosslinking at the benzene ring of the molecule.

Curing

It can be noted from the results in Table II that there is a significant reduction in the cure time of the mixes in presence of the cardanol-based resins. The higher cure rate of the PCP modified mixes may be due to enhanced mobility of isoprene chains in presence of PCP and/or

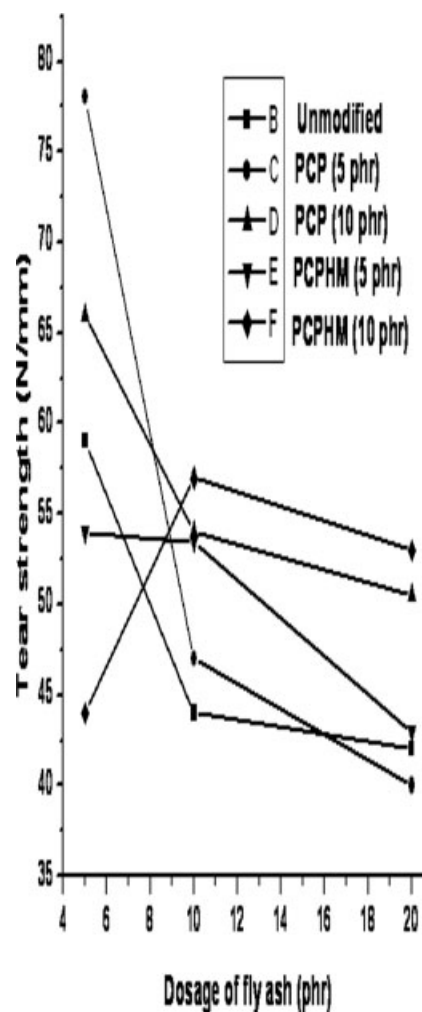


Figure 4 Effect of dosage of fly ash on tear strength of NR vulcanizates containing PCP/PCPHM.

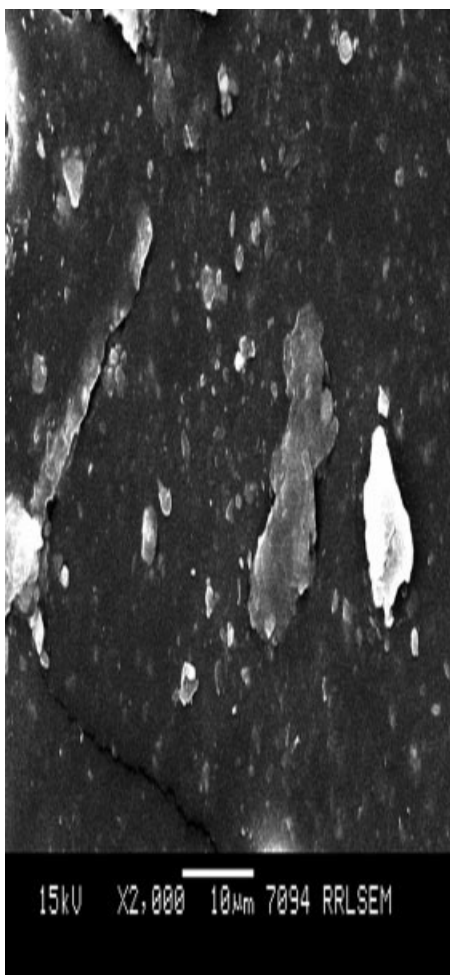


Figure 5 SEM of NR vulcanizate containing 10 phr of fly ash ($\times 2000$).

to the immediate access to sufficient unsaturation sites of the side chain of PCP for crosslink formation with the isoprene main chains.^{26,27} The mixes containing PCPHM show the lowest cure time. This may be due to the higher alkalinity of this resin, as basic materials in general are known to improve the rate of cure in elastomers.

Mechanical properties

For most practical applications, it is essential that the vulcanizates of the filled compositions should possess the required physico-mechanical properties.

Tensile properties

The variations in the tensile properties of the fly-ash-filled NR vulcanizates with the dosage of filler are shown in Figures 1–3. Figure 1 shows that at the filler dosage of 10 and 15 phr, the tensile moduli at 100% extension of the vulcanizates containing 5 phr of PCP are lower than that of the unmodified vulcanizates. Also, in the presence of 15 phr of fly ash, the vulcani-

zate containing 5 phr of PCPHM shows a lower modulus than that of the unmodified vulcanizate containing the same dosage of fly ash. The lower values of the tensile modulus are a reflection of the softening effect of the resins resulting from the plasticization effect. Also, Figures 2 and 3 show that in presence of 15 phr of fly ash, the vulcanizates containing PCP/PCPHM show considerably higher tensile strength and elongation at break than that of the unmodified vulcanizate.

Tear strength

The results given in Figure 4 show that tear strengths of the vulcanizates containing 10 phr of fly ash and 5–10 phr of PCP/PCPHM are higher than that of the unmodified sample containing the same dosage of filler.

The improvement in mechanical properties as mentioned above for the PCP/PCPHM modified vulcanizates could probably be partly due to the compatibilizing effect of these amphiphilic resins between the

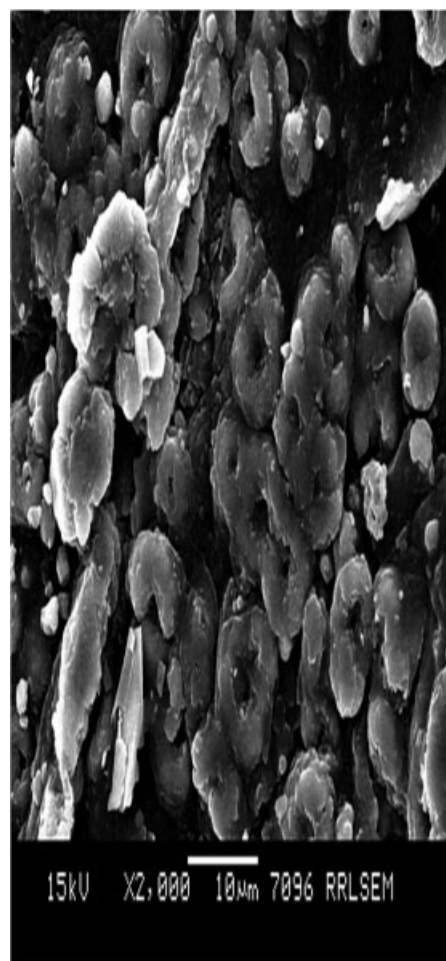


Figure 6 SEM of NR vulcanizate containing 10 phr of fly ash and 5 phr of PCP ($\times 2000$).

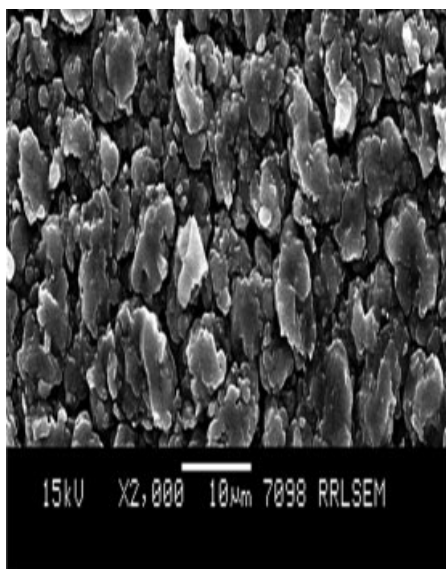


Figure 7 SEM of NR vulcanizate containing 10 phr of fly ash and 10 phr of PCP ($\times 2000$).

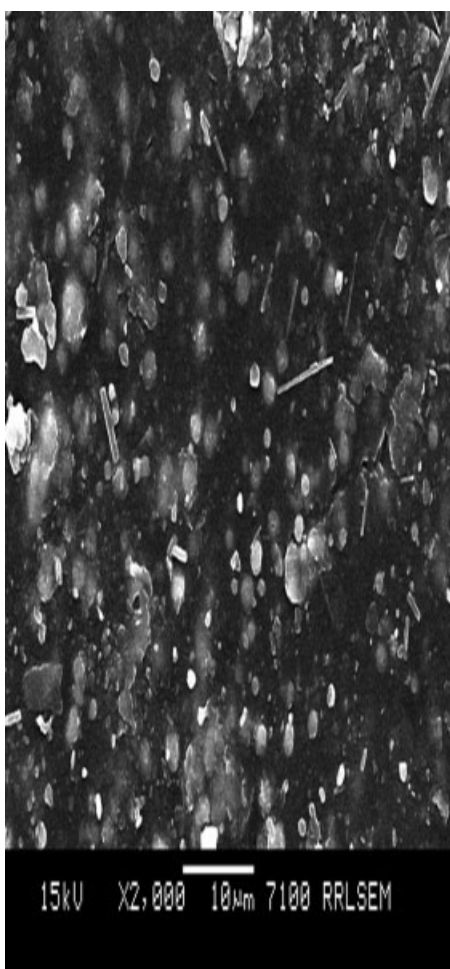


Figure 8 SEM of NR vulcanizate containing 10 phr of fly ash and 5 phr of PCPHM ($\times 2000$).

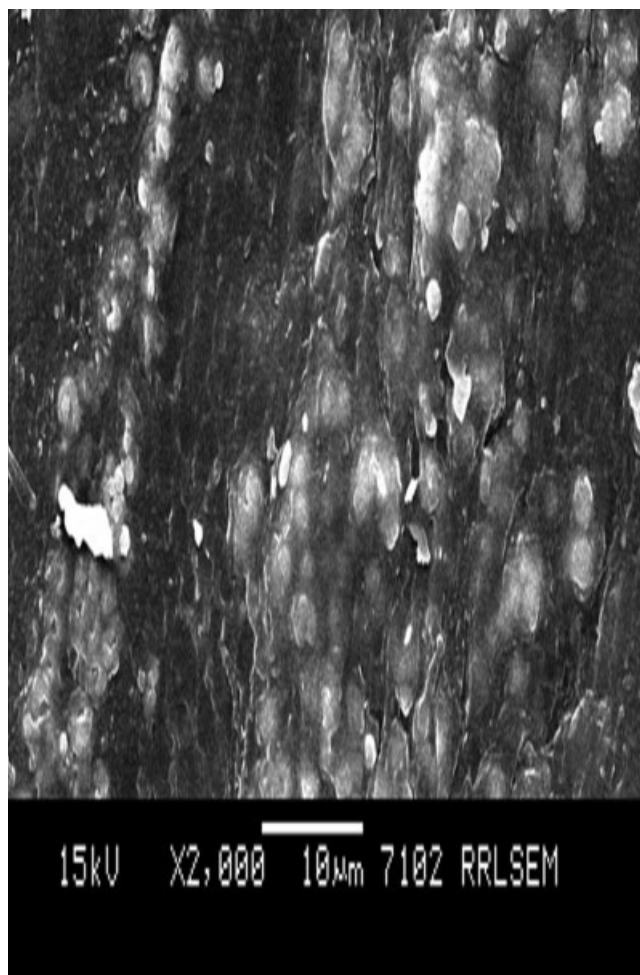


Figure 9 SEM of NR vulcanizate containing 10 phr of fly ash and 10 phr of PCPHM ($\times 2000$).

polar silica particles of fly ash and the nonpolar NR in a manner, as reported earlier for silica filled NR.³⁰ In PCP-modified unfilled NR vulcanizates, simultaneous improvements in tensile strength, elongation at break, and tear strength have been reported previously.^{25,26} This was ascribed to the formation of an entangled network structure of PCP with NR.²⁶ It is expected that the enhancement in the hydrophobicity of the particles of fly ash resulting from its interaction with PCP/PCPHM will improve the reinforcement potential of fly ash.

3. Morphology of the fracture surfaces

The scanning electron micrographs of the tensile fracture surfaces of the fly-ash-filled NR vulcanizates are given in Figures 5–9. The SEM micrograph of the unmodified NR vulcanizate containing 10 phr of fly ash (Fig. 5) shows particles of nonuniform size with bigger agglomerates of the filler. In presence of 5 phr of PCP, the filler agglomerates are smaller and uniform in size, as shown by Figure 6. The microstructure

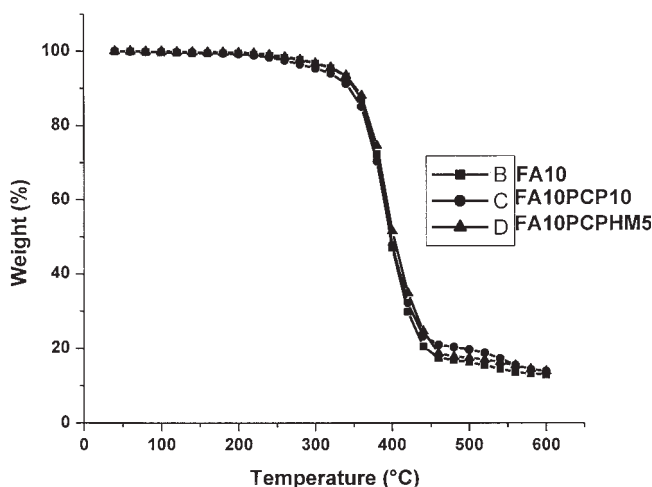


Figure 10 TGA traces of fly-ash-filled (10 phr) NR vulcanizates containing PCP/PCPHM.

of the fly-ash-filled NR vulcanizate containing 10 phr of PCP (Fig. 7) shows filler particles with lower size. In contrast to the above, the fractographs of the vulcanizates containing 5 phr of PCPHM (Fig. 8) and 10 phr of PCPHM (Fig. 9) show that the fly ash particles are much smaller and uniformly distributed in the rubber matrix. It can be noted that in the vulcanizate containing 5 phr of PCPHM (Fig. 8), the particles of fly ash are much smaller, with uniform dispersion as against that containing 10 phr of PCPHM (Fig. 9). Probably, a lower degree of softening in the case of the former results in the generation of higher shear stresses in the internal mixer, resulting in better size reduction and dispersion of the fly ash particles. It is expected that the finer and more uniform distribution of the fly ash particles in the NR matrix is partly responsible for the higher mechanical properties of the cardanol resin modified vulcanizates.

The degree of reinforcement provided by a filler in an elastomer depends on a number of variables, the most important of which is the development of a large polymer-filler interface.³⁷ Thus, in silica-filled elastomers containing coupling agents the observed improvement in mechanical properties is ascribed not only to improved pigment-elastomer adherence, but also to better dispersion of the filler in the elastomer matrix.³⁸ The plasticizing and softening effect of PCP and PCPHM on NR improves the distribution and dis-

persion of fly ash in the NR matrix, particularly at the lower dosage of the filler (10 phr). Besides, it is possible that these additives may act as coupling agents between NR and the particles of fly ash as mentioned above. Such an interaction in turn will lead to an improvement in mechanical properties, as observed in the present study.

Thermal decomposition characteristics

The thermal decomposition characteristics of the vulcanizates were studied by dynamic thermogravimetry. The temperature at which 50% of the weight of a given polymer is vaporized under standard pyrolytic conditions (T_{50}) has been considered to be a quantitative measure of thermal stability.³⁹ The TGA traces of the NR vulcanizates containing 10 phr of fly ash in presence and absence of PCP/PCPHM are given in Figure 10. The thermal stability index (T_{50}) of the different vulcanizates are shown in Table III. Figure 10 and the results in Table III show some improvement (though to a lower extent) in the thermal stability of the fly-ash-filled NR vulcanizates in presence of 10 phr of PCP and 5–10 phr of PCPHM. This may be due to the formation of a higher degree of crosslinking in these systems or thermally stable structures subsequently decomposing at higher temperatures, similar to that reported earlier for unfilled⁴⁰ and filled⁴¹ NR vucanizates.

CONCLUSION

The processability characteristics, mechanical properties, morphology of fracture surfaces, and thermal decomposition characteristics of fly-ash-filled NR containing different dosages (5–10 phr) of PCP/PCPHM have been studied. At optimal dosages of 10–15 phr of fly ash in NR, the PCP/PCPHM-modified mixes showed lower power consumption for mixing, lower cure time, higher tensile modulus, tensile strength, elongation at break, tear strength, better dispersion of finer particles of filler in the rubber matrix, and improved thermal stability. It is expected that the cardanol-based resins act as coupling agents between particles of fly ash and NR, thereby improving the mechanical properties.

TABLE III
Thermal Stability Index (T_{50}) of Fly-Ash-Modified (10 phr) NR Containing Different Dosages of PCP/PCPHM

Mix code ^a	FA10	FA10 PCP5	FA10 PCP10	FA10 PCPHM 5	FA10 PCPHM 10
Property					
Thermal stability index, T_{50} (°C)	397.5	396	401.3	401.6	401.4

^a Base mix: NR 100, ZnO₅, stearic acid 2, sulphur 2, and MBT 2.

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