

# Regeneration of Carbon Black from Waste Automobile Tires

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**ABSTRACT:** Carbon black, an important ingredient in tires, was regenerated from waste automobile tires. Waste tires were cut to separate the tread cap, tread base, side wall, and inner ply and were crushed and pyrolyzed in a muffle furnace at 430°C for 3 h. This black was again heated at 430°C for 1.5 h to increase the surface area to get regenerated black (RB). To find out the effect of heating during pyrolysis, the original black was also heated at 430°C for 1.5 h to get heat-treated black (HTB). The regenerated black (RB) was characterized by different methods. It was mixed in different formulations, with few required properties of the final mixed compounds measured, and then cured maintaining technical cure time and temperature; physicochemical properties of the cured specimen were measured and compared with that of the virgin black (VB) and the heat-treated

black (HTB). From all these results it was concluded that regenerated black has a higher surface area, higher elongation at break, and higher heat buildup but a lower cure rate, a lower modulus, and a comparable state of cure, dispersion, hardness, tensile strength, and tear strength compared with those of the virgin black. It was also found that for the regenerated black, the structure and agglomerate size were slightly changed. Some polar acidic groups were incorporated into the carbon black during pyrolysis or during heat treatment. Regenerated black had better aging properties than did the virgin or heat-treated black. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 89: 465–473, 2003

**Key words:** aging; ESCA; processing; pyrolysis

## INTRODUCTION

The enormous number of used tires has been threatening the environment, especially in the United States and Canada, where one used tire is dumped per person per year. Most of these used tires are not recycled but are simply dumped in open or landfill sites, where they breed mosquitoes and rats and can be the cause of uncontrolled fires.<sup>1</sup> To solve this environmental problem, waste tires are being reused in a manner that is both environmentally friendly and economically viable.

There are several disposal methods<sup>1</sup> for waste tires, including (1) landfills, dumping, stockpiling; (2) incineration; (3) retreading; (4) reclamation; and others. Among the above disposal methods landfills are more expensive, with increasing restrictions being applied, particularly since the recent spate of fires in landfills. Deliberate burning in the open air is wasteful and against air pollution regulations and is thus no longer allowed. Of the above described disposal methods, reclamation is the most environmentally friendly. However, 1989–90 statistics show that only 5% of the total amount of waste tires is reclaimed.<sup>1</sup>

There is a market for scrap rubber in the form of crumb, but the market is limited and the economics debatable.<sup>1</sup> Pyrolysis of scrap rubber from tires to remove and recycle raw material meets the ideal objective and can be done economically. The current work was mainly focused on regeneration of carbon black from waste automobile tires.

Tires contain 20%–30% carbon black and 3%–7% oil, which are valuable and can be regenerated easily by simple pyrolysis under controlled conditions. There are different methods for regeneration of carbon black from waste automobile tires, including: (1) vacuum pyrolysis,<sup>2</sup> (2) inert atmosphere pyrolysis,<sup>1</sup> (3) pyrolysis in a muffle furnace, (4) microwave conversion of scrap tires,<sup>1</sup> (5) nitric acid digestion, (6) metathesis degradation,<sup>3</sup> and (7) supercritical and subcritical water oxidation.<sup>4</sup> In the current study the third process, pyrolysis in a muffle furnace, was followed for the regeneration of carbon black from waste automobile tires because of its simplicity in operation. Because the raw materials required for the regeneration of carbon black are mainly waste tires (only transportation and labor costs are required for its collection, i.e., cheap material) and electrical energy, the process should also be economically viable.

However, regenerated carbon black is different from virgin commercial carbon black. Different blacks may be mixed up along with the inorganic components, and additives of the tire may change the morphology of the regenerated black. Regenerated black

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may contain adsorbed oxygenated or acidic groups and hydrocarbon or carbonaceous deposits on the surface<sup>2</sup>.

So, the aim of this work was to regenerate carbon black from waste automobile tires and to characterize it to find out its properties and morphological change from that of virgin commercial black. For this study some required formulations with virgin, heat-treated, and regenerated blacks were made to study the processibility, curing characteristics, and physical properties.

## EXPERIMENTAL

### Materials

Natural rubber (RMA-4, ISNR-20, EBC-3X, J.K. Supplying Agency, Cochin, India), polybutadiene rubber (Cisamer 1230, Indian Petrochemical Corporation, Ltd., Vadodara, India), acetone, hydrochloric acid [E. Merck (India) Ltd., Mumbai, India], chloroform (Ranbaxy Laboratories Ltd., India), Peptizer (pentachlorothio phenol; I.C.I. Ltd., Rishra, India), zinc oxide [Meta Zinc (India) Ltd.], stearic acid (Godrej Soaps, Ltd., Malanpur, India), N330-, N550-, and N660-type carbon black (Cabot India, Ltd.), aromatic oil (Hindustan Petroleum Corporation, Ltd., India), antioxidant [polymerized 2,2,4-trimethyl-1,2-dihydro quinoline (TMQ); Bayer India Ltd., India], antiozonate [diphenyl para phenylene diamine (DPPD)], *N*-1,3-dimethylbutyl-*N'*-phenyl para phenylenediamine (6PPD) from Bayer India Ltd., India], microcrystalline wax (Indian Petrochemical Corporation, Ltd., Vadodara, India), sulfur (Jain Chemicals, Ltd., Thane, India), accelerator [*N*-oxydiethylene benzothiazole-2-sulfenamide (NOBS)], tetramethyl thiouram monosulfide [TMTM; M/S Bayer India Ltd., Thane, India], prevulcanization inhibitor (PVI: *N*-cyclohexyl thiophthalimide).

### Regeneration of carbon black from waste tires

A known truck tire was taken that contained N330 in tread cap and in side wall, N550 in tread base, and N660 in inner ply. The known tire was taken to compare the characteristic properties of the known regenerated black with that of virgin black.

The whole tire was cut into small pieces by knife and then different portions—tread cap, side wall (for N330), tread base (for N550), and inner ply (for N660)—were separated by a slicing machine. Those portions were taken and crushed in a two-roll mill into pieces about 10 mm<sup>3</sup> in size. The crushed product was then taken in a crucible, which was heated at 430°C in a muffle furnace (from M/S Tempo I & E, India) for 3 h. Initially, the temperature of the furnace was kept at 200°C. Then the temperature was increased slowly at a rate of 10°C/min. The logic behind this was that initially when the rubber compound was heated at a

lower temperature, volatile matters would evolve that could replace oxygen from the system, making it a partially oxygen-free system.

After pyrolysis was completed, a black residue was left that contained carbon black, carbonaceous particles, and other inorganic materials. To ensure the complete elimination of any organic materials, the residue was washed first with acetone and then acetone + chloroform (1:1). To remove ZnO, the residue was treated with concentrated HCl. Finally, to remove HCl it was washed with water, checked with silver nitrate to make it free from chlorine, and then dried at 100°C.

This black was heated for a second time at 430°C for 1.5 h to improve its surface roughness and surface area by removing carbonaceous particles as much as possible.

### Heating of the virgin black

During pyrolysis regenerated black experiences heat, which has an effect on the properties and morphology of the regenerated black. To find out the effect of heat on properties and morphology, virgin black was also heated under the same conditions (at 430°C for 1.5 h).

To find out the change in properties and morphology because of pyrolysis or heat treatment, all the properties for the virgin blacks (VB), heat-treated blacks (HTB), and regenerated blacks (RB) were measured and compared. The probable reasons for the change in properties were explained accordingly.

### Characterization of carbon blacks

All the blacks were characterized by the following tests.

#### Iodine adsorption number

In the iodine adsorption number test (ASTM D 1510-96), about 0.5 g of dried (at 125°C for 1 h) carbon black was shaken with 25 mL of iodine solution in a stoppered conical flask for 1 min and centrifuged in a centrifuge (M/S Remi, India) for 2 min. Then the decanted iodine solution was titrated with a standard sodium thiosulfate solution, and then the adsorbed iodine was calculated in milligrams of iodine per gram of black.

#### Nitrogen surface area measurement

In the nitrogen surface area measurement test<sup>2</sup> (ASTM D 4820) a small amount of carbon black sample was placed in a known volume cell of a nitrogen surface area analyzer (Gemini 2370, Micromeritics, USA) and evacuated to less than 5 millitorr of vacuum. Using the ideal gas equation, the volume of nitrogen required to give a predetermined relative pressure was calculated and dosed into the sample cell. Any additional nitro-

TABLE I  
Formulations

Ingredients	Tread cap	Side wall	Tread base	Inner ply
Natural rubber	100.00	70.00	100.00	100.00
Butadiene rubber	—	30.00	—	—
Pentachloro Thiophenol	0.20	0.12	0.12	0.07
Carbon black	HAF-50.00	HAF-45.00	FEF-45.00	GPF-40.00
ZnO	5.00	6.00	5.00	4.00
Stearic acid	3.00	2.00	3.00	2.50
Antioxidant/antiozonate	3.25	3.70	4.30	0.60
Microcrystalline wax	—	2.00	2.00	—
Process oil	6.00	5.00	10.00	7.00
Sulfur	2.25	1.44	1.50	1.50
Accelerator	0.50	0.65	1.00	1.30
Prevulcanization Inhibitor	0.20	0.35	0.10	0.10

gen required to attain this relative pressure was a result of adsorption by the carbon black. Based on the volume of nitrogen adsorbed at various relative pressures, the surface area was calculated applying Brunauer–Emmet–Teller (BET) theory and the BET equation:

$$\frac{1}{V[P_0/P^{-1}]} = \frac{1}{V_m C} + \frac{C-1}{V_m C} \times \frac{P}{P_0}$$

where  $V$  = volume of nitrogen adsorbed (in  $\text{cm}^{-3}$ );  $P$  = pressure (in KPa);  $P_0$  = saturation vapor pressure of nitrogen (in KPa);  $V_m$  = volume of nitrogen that covers one monomolecular layer (in  $\text{cm}^{-3}$ ); and  $C$  = BET constant.

#### Determination of structure of carbon black by DBP absorption

In determination of the structure of carbon black by DBP absorption (ASTM D 2414-96), about 20 g of the sample was weighed and transferred to the absorptometer (model E, C. W. Brabender Co., USA) mixing chamber, and then the instrument was switched on. When the instrument stopped, the amount of DBP absorbed was recorded on a digital counter. From this result the DBP required for 100 g of carbon black was calculated.

#### Determination of pH

To determine the pH (ASTM D 1512-95), about 5 g of sample was crushed in a mortar and then put in a 100-mL beaker to which 50 mL of distilled water was added. The mixture was then boiled for 10 min after which the beaker was covered with watch glass until cooled. The supernatant liquid was poured off, and the pH was determined from the remaining slurry by a pH meter (model EA-940, ORION, USA).

#### Determination of surface structure with SEM

Silver paint was put on a metal stuff, and one pellet of carbon black was placed on it. Then it was sputtered to coat with gold and a photograph was taken by scanning electron microscopy (SEM; JEOL, Japan).

#### Electron spectroscopy for chemical analysis

The electron spectroscopy for chemical analysis<sup>5</sup> (ESCA) technique involves the measurement of binding energies of electrons in molecules or lattices. Hence, it is used to find out the binding energy of the surface groups present in VB, HTB, and RB. The sample was irradiated with X-rays, and ejected electrons were analyzed by a double-focusing electrostatic energy analyzer (VG Scientific Ltd., UK), and the system was maintained under a vacuum of  $< 10^{-8}$  torr.

#### Mixing

Mixing was done according to ASTM D 3182 in two stages: (1) master batch mixing and (2) final batch mixing.

The master batch mixing was carried out in a Brabender Plasticorder (model PL 2000-3, Brabender OHG, Duisburg). Rubbers and other ingredients, except vulcanizing agents, were weighed according to the appropriate formulations (Table I), considering 65 g to be the total batch weight. Rubber and peptizer were put in the Brabender chamber and masticated for 30–45 s. Then the other ingredients were added and mixed. The whole mixture was scraped after 2 and 4 min, and the mixture was dumped after 6 min. The mixing was done at 100°C with 60 rpm rotor speed.

Final batch mixing was done in a two-roll mill (model MML; Santosh Rubber Machinery Pvt. Ltd., Mumbai, India) by doing about 20 passes.

#### Characterization of the final mixed compounds

After the mixing was completed, the final mixed compounds were characterized by the following methods.

**TABLE II**  
**Curing Condition**

Specimen	Curing temperature (°C)	Curing time (min)
Tensile sheet (for TS tear strength)	141	45
Heat buildup specimen	141	60

According to ASTM Standard D 5289, the cure characteristics of the mixed products were determined with a rheometer (model MDR 2000 E; Monsanto, USA). A processibility tester (MPT; Monsanto, USA) was used to measure process properties of the compounds containing VB, HTB, and RB. A dispersometer (model 1000; Optigrade, AB Sweden) was used to determine dispersibility and the agglomerate size of the carbon black, according to ASTM Standard D 2663.

### Bound rubber<sup>6</sup>

Using wire-net small boxes that had been prepared, about 2–3 g of the final mixed compound was put in the wire-net box, which was then immersed in toluene. The solvent was changed every day until it became black (4–6 days). Then the wire box was taken out, the sample was dried, and TGA (model TGA-7; M/S Perkin–Elmer, USA) was performed to determine the percentage of polymer and black, from which the percentage of bound rubber was calculated.

### Molding

All the samples were molded according to ASTM Standard D 3182 in a molding press (Hind Hydraulics & Engineers) under a pressure of 5 MPa. Times and temperatures for different specimen are given in Table II.

### Testing of cured specimens

#### Hardness

Hardness was measured according to ASTM Standard D 1415, for IRHD using a dead load tester (H. W. Wallace & Co.) and according to ASTM Standard D 2240 for Shore A, using a Durometer (Rahul & Co., Mumbai, India).

#### Tensile and tear strength

Tensile strength; 100%, 200%, and 300% modulus; elongation at break, and tear strength were measured using a Zwick UTM-1445 (Zwick, Germany) according to the standard procedure ASTM D 412-80.

### Heat buildup

Heat buildup was measured with a Goodrich flexometer (Ferry Machine Co., USA) according to ASTM Standard D 623-78. A specimen 25–26.5 mm in height and 17.8–19.9 mm in diameter was preconditioned at about 100°C for 30 min, and then the test was performed for another 30 min until a reading was finally taken. Heat buildup = final reading – initial reading.

### Specific gravity

The weights of the sample, empty specific bottle, bottle + water, and bottle + water + sample were taken separately. Then the specific gravity of the compound was determined from eq. (1):

$$\text{Specific Gravity} = \frac{(F - A)}{(B - A) - (E - F)} \quad (1)$$

where  $A$  is the weight of the specific gravity bottle,  $B$  is the weight of the specific gravity bottle + water,  $E$  is the weight of the specific gravity bottle + water + sample, and  $F$  is the weight of the specific gravity bottle + sample.

### Volume fraction and swelling index<sup>7</sup>

To determine the volume fraction and swelling index, 0.2–0.3 g of the vulcanized sample was kept in toluene for 48 h in a closed vessel. The swelled sample was taken out, and the solvent was soaked by blotting paper and weighed immediately. Then the sample was kept at room temperature for complete drying, and a constant weight of dried sample was taken. The volume fraction and swelling index of the rubber were calculated on the basis of eqs. (2) and (3), respectively.

$$V_r = \frac{F - \frac{AT}{\rho_r}}{F - \frac{AT}{\rho_r} + \frac{B - A}{\rho_s}} \quad (2)$$

where  $A$  is the initial weight of the sample,  $B$  is the swollen weight of the sample,  $F$  is the dried or deswelled weight of the sample,  $T$  is the insoluble fraction (carbon black + ZnO) in the mix,  $\rho_s$  is the specific gravity of the solvent, and  $\rho_r$  is the specific gravity of the rubber.

$$\text{Swelling Index} = \frac{\text{Swollen weight}}{\text{Original weight}} \quad (3)$$

**TABLE III**  
**Characteristic Properties of FEF (N550) Blacks**

Properties	VB (N550)	HTB (N550)	RB (N550)
IODINE NUMBER (mg of I <sub>2</sub> /gm of black)	45.7	91.5	101.9
NITROGEN SURFACE AREA (m <sup>2</sup> /gm)	41.7	103.9	98.5
DBP Absorption (ml of DBP/100gm of black)	118.2	126.1	115.2
pH	7.4	3.6	5.9

VB: virgin black; HTB: heat-treated black; RB: regenerated black.

**Accelerated aging and aging properties**

Tensile specimen was kept in multicell aging oven [M/S Tempo (India) Ltd.] for 2 weeks at a temperature of 70°C. Then the specimen was taken out and kept 1 day at room temperature. The tensile properties were then measured using a Zwick UTM 1445.

**RESULTS AND DISCUSSION**

Though all the tests were performed for all the N330, N550, and N660 blacks, here results only for N550 are reports.

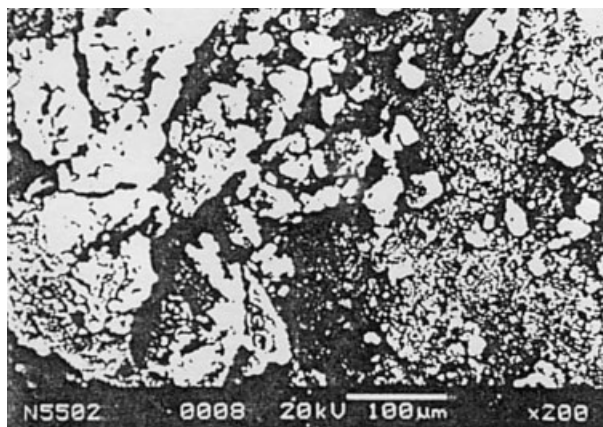
**Study of surface area, structure, and pH**

Iodine number, nitrogen surface area and DBP values are given in Table III. The results show that RB and HTB had a higher surface area than did the VB (confirmed from iodine number and nitrogen surface area). This is because of the increase in surface roughness and partial breakdown of agglomerate during different processing steps of manufacturing.<sup>2</sup>

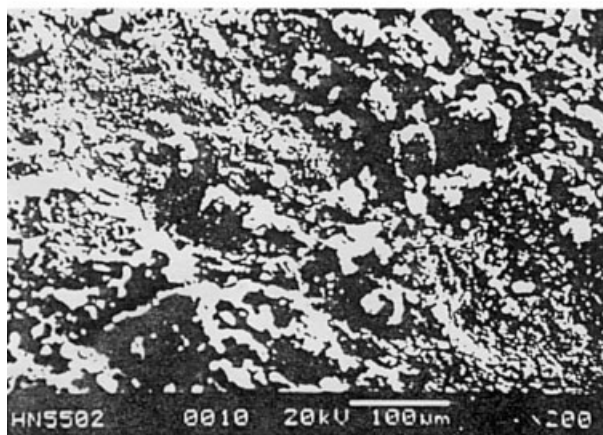
The DBP absorption value was slightly higher for HTB and slightly lower for RB than for VB, which indicates that the structure did not change too much during regeneration or heat treatment. The increase in surface area was greater for HTB than for RB. This is because of the partial deposition of carbonaceous particles, which come from the polymer and the different organic compounds. These carbonaceous particles smooth the surface of the carbon black, and as a result the resultant surface area of the RB was lower compared with that of the HTB.<sup>8</sup>

The pH for RB/HTB was lower compared with that of VB. This indicates that during pyrolysis or heat treatment partial oxidation of the blacks took place. So some acidic groups like carbonyl, alcoholic, and carboxylic groups and some thiocarbonyl, thiophenol, or thiocarboxylic groups may have been incorporated onto the surface of the carbon black. But the incorpo-

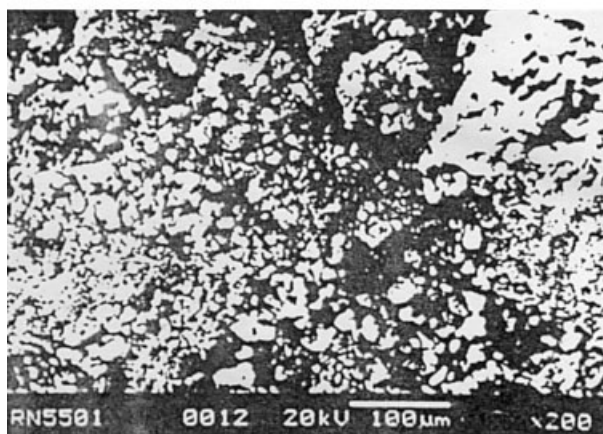
ration of acidic groups was greater for HTB than for RB. This may be because of the presence of some basic chemicals in the rubber compound, which partially neutralized the acidity of the RB.



(a)



(b)



(c)

**Figure 1** SEM photographs of: (a) VB (N550); (b) HTB (N550); (c) RB (N550).

### SEM study for different N550 blacks

Figure 1(a) shows the agglomerate structure of VB (N550); Figure 1(b) shows HTB (N550), where a few small particles having an average size of about  $25 \mu\text{m}$  are present. The surface roughness and porosity of HTB (N550) are higher than those of VB (N550). So the surface area and structure of HTB (N550) are higher compared with those of VB (N550). Figure 1(c) shows the RB (N550), for which it can be observed that the greatest number of small particles have an average size of about  $15 \mu\text{m}$ . Here, a partial breakage of agglomerate has taken place. So, an increase in the surface area but a decrease in structure can be observed.

### ESCA study of different N550 blacks

Figure 2(a) shows the ESCA peaks for  $C_{1s}$  of VB, HTB, and RB. Here the peak of HTB is slightly shifted to the right side from the peak of VB, and the peak of RB is again slightly shifted to the right side from the peak of HTB. We know if any polar atom or group is attached to another atom, the binding energy of the electrons of the second atom are increased, that is, ESCA peak is shifted to the right side. So, shifting of peaks indicates that maximum number of polar atoms or groups is attached with RB, a medium number of polar atoms or groups is attached with HTB, and the minimum number of polar atoms or groups is attached with VB. Figure 2(b) shows the ESCA peaks for  $O_{1s}$ . The broad peaks indicate the presence of different groups such as carbonyl, hydroxyl, carboxylic, and ether groups.<sup>5</sup> An extra peak [Fig. 2(c)] appears for RB, which is for chlorine, indicating that some chlorine has been incorporated, maybe because during washing with HCl, the chlorine was not completely removed.

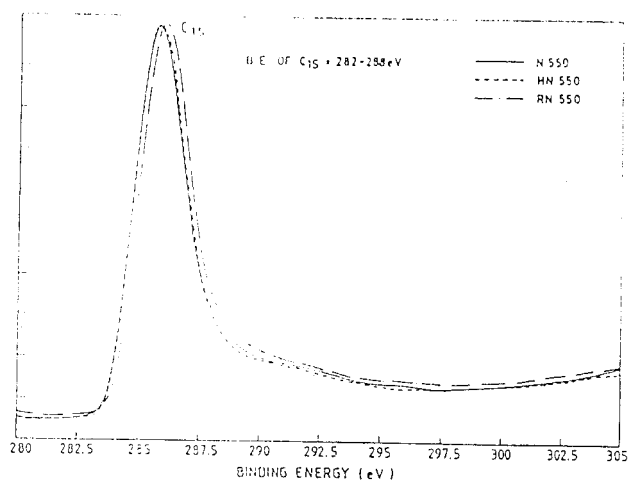
### Characterization of the carbon blacks in final mixed compounds

#### Rheometric study

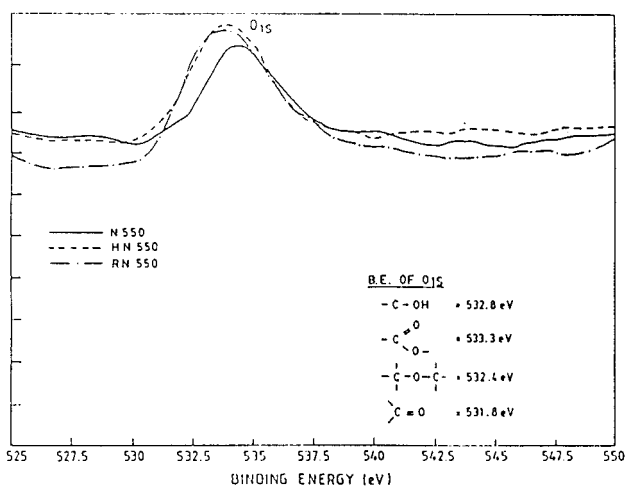
The rheometric study (MDR) results (Table IV and Fig. 3) show that both HTB and RB have a higher scorch safety, higher optimum cure time, and a lower rate of curing. Previously, it was seen that HTB and RB had a lower pH, that is, higher acidity. So these results are in good correlation.

Accelerators are generally basic. So, in the presence of any acidic groups, the activity of the accelerator will decrease, and as a result the compound shows more scorch safety, a higher optimum cure time, and a lower rate of curing.

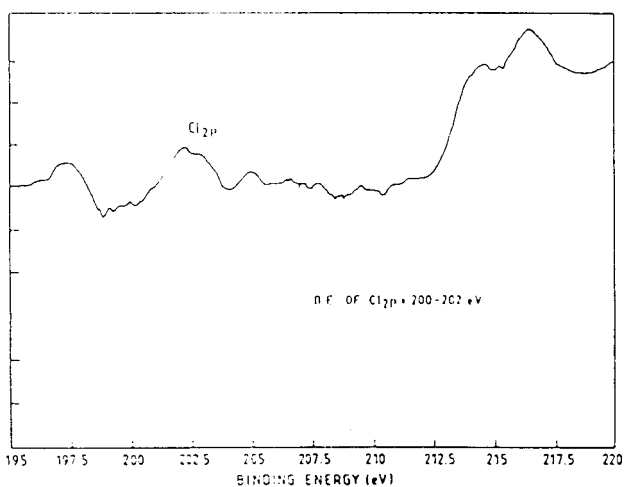
Because the acidity of HTB is maximum (minimum pH), its scorch time and optimum cure time should be maximum, and its rate of curing should be minimum. But in reality the scorch time and optimum cure time are maximum for regenerated black and the rate of curing is minimum. This may be because of the pres-



(a)



(b)



(c)

**Figure 2** (a) ESCA/XPS curve ( $C_{1s}$  peaks) for FEF (N550) black; (b) ESCA/XPS curve ( $O_{1s}$  peaks) for FEF (N550) black; (c) ESCA/XPS curve ( $Cl_{2p}$  peak) for RB (N550) black.

**TABLE IV**  
**Curing Characteristics of Tread Base Final Mixed Compound Containing FEF (N550) Black**

Properties	VB (N550)	HTB (N550)	RB (N550)
Maximum Torque (lb-in)	9.12	7.87	8.51
Minimum torque (lb-in)	0.70	0.82	0.70
TS02 (scorch time) in min	8.34	10.10	9.92
TC90 (optimum cure time) in min	13.02	17.23	16.12
Average rate of curing (lb-in/min.)	0.80	0.67	0.64

VB: virgin black; HTB: heated-treated black; RB: regenerated black.

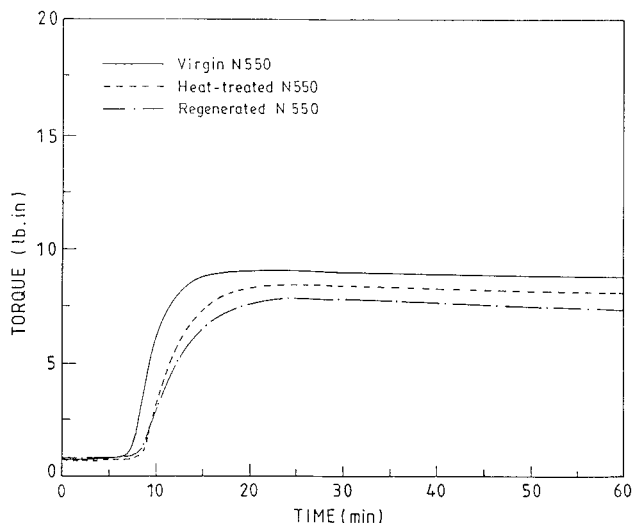
ence of chlorine in RB, which was confirmed from the ESCA result. As chlorine is acidic, it has a retarding effect on accelerator activity. Thus, the results of pH, ESCA, and MDR are correlated to one another.

**SEM study of different final mixed compounds containing different N550 blacks**

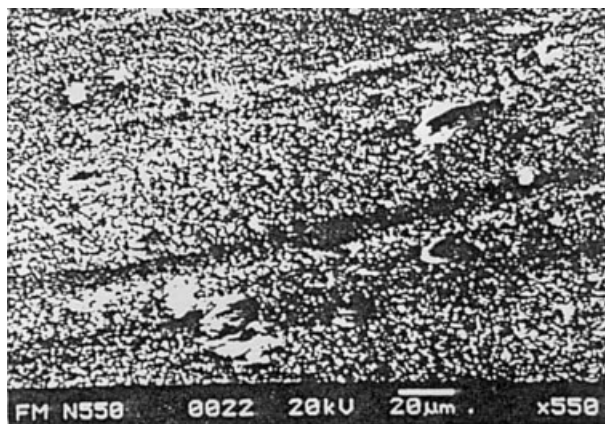
Figure 4(a) is the SEM photograph of the tread base compound containing VB, Figure 4(b) of the same compound containing HTB, in which a poorer distribution of carbon black can be observed, yielding a poorer failure property (tensile strength) than that of VB. Figure 4(c) shows the same compound containing RB, in which a better distribution of carbon black can be observed, giving a better failure property (tensile strength) compared with that of VB.

**Processing properties (MPT results) of different final mixed compounds containing different N550 blacks**

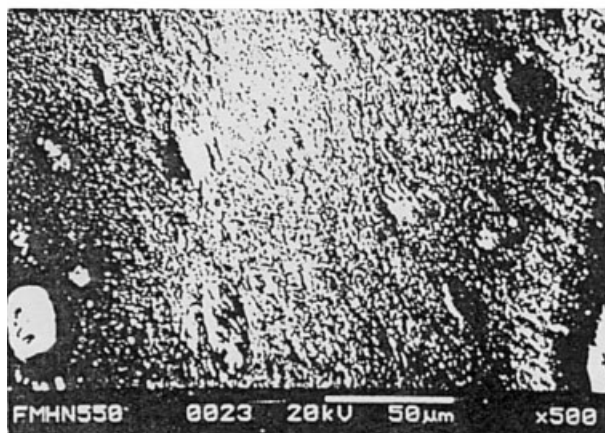
At a particular shear rate, shear stress is higher [Fig. 5(a)] for compounds containing RB, indicating a better



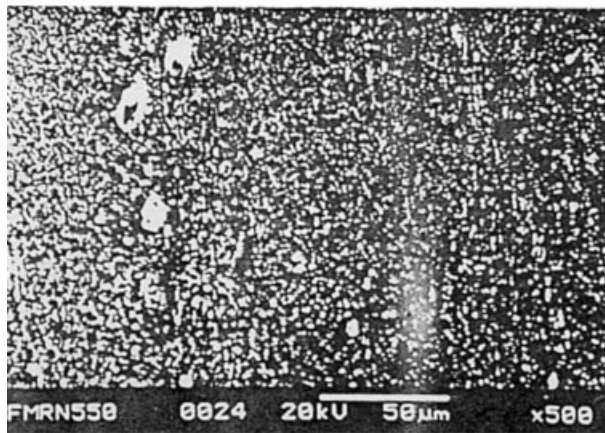
**Figure 3** MDR curve for final mixed compounds containing VB, HTB, and RB.



(a)



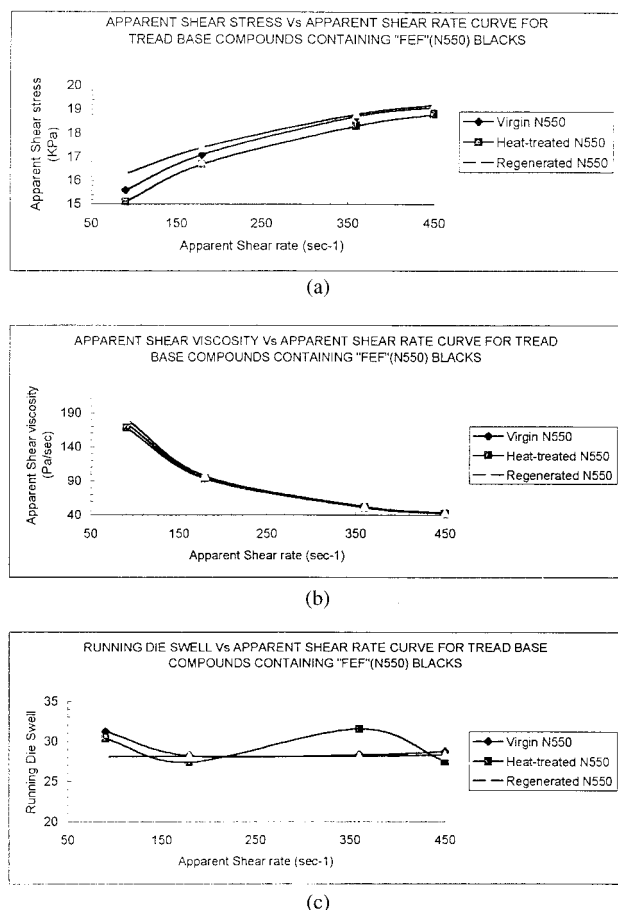
(b)



(c)

**Figure 4** SEM photographs of tread base compound containing (a) VB (N550); (b) HTB (N550); (c) RB (N550).

rubber-filler interaction, which is a result of the greater surface area of the RB. Viscosity at a particular shear rate is comparable [Fig. 5(b)] for all three compounds containing VB, HTB, and RB. The running die swell [Fig. 5(c)] is lower and more consistent for the compounds containing RB than for that of VB and HTB, which indicates a better rubber-filler interaction and better processibility.



**Figure 5** (a) Apparent shear stress versus apparent shear rate curve for tread base compounds containing different FEF (N550) blacks; (b) apparent shear viscosity versus apparent shear rate curve for tread base compounds containing different FEF (N550) blacks; (c) running die swell versus apparent shear rate curve for tread base compounds containing different FEF (N550) blacks.

### Dispersibility, agglomerate size, and bound rubber

The dispersibility (Table V) of HTB is slightly higher, whereas it is slightly lower for RB than for VB. The agglomerate sizes (Table V) of HTB and VB are comparable, with a slight decrease in agglomerate observed for RB. Bound rubber (Table V) for RB and

**TABLE V**  
Properties of FEF (N550) Blacks in Tread Base Compounds

Properties	VB (N550)	HTB (N550)	RB (N550)
Average dispersibility of carbon black in final mixed compound (1–10)	5.52	5.74	5.00
Average agglomerate size of carbon black (milimicron)	9.14	9.18	8.56
Bound rubber (%)	29.0	39.1	36.1

VB: virgin black; HTB: heated-treated black; RB: regenerated black.

**TABLE VI**  
Physical and Physicomechanical Properties of Tread Base Compounds Containing FEF (N550) Blacks

Properties	VB (N550)	HTB (N550)	RB (N550)
Specific gravity	1.097	1.121	1.100
Swelling index	2.46	2.46	2.68
Volume fraction	0.25	0.25	0.23
Hardness (Shore A)	55	54	53
Hardness (IRHD)	59	57	56
Tensile strength (MPa)	22.2	20.1	22.4
Modulus at 100% elongation (MPa)	1.9	1.6	1.4
Modulus at 200% elongation (MPa)	5.0	4.3	3.7
Modulus at 300% elongation (MPa)	8.9	8.1	7.4
Elongation at break (%)	566	562	588
Tear strength (N/mm)	51	55	55
Heat buildup (°C)	10.2	13.2	13.8

VB: virgin black; HTB: heated-treated black; RB: regenerated black.

HTB is higher compared with that of VB, indicating a better rubber–filler interaction in the green state.<sup>6</sup>

### Properties of carbon black–filled compounds

The swelling index (Table VI) of the cured samples containing VB and HTB is the same, but it is higher for RB, indicating that the state of cure is the same for compounds containing VB and HTB, but the cure is incomplete for the compounds containing RB. The lower volume fraction for the compounds containing RB also indicates undercuring or incomplete curing.<sup>9</sup>

Hardness (Table VI) has decreased slightly for the compounds containing HTB and decreased even more for the compounds containing RB. This is may be a result of the undercuring, because as already seen from the MDR result, the rate of curing is the minimum for the compounds containing RB, medium for the compounds containing HTB, and maximum for the compounds containing VB.

Tensile strengths (Table VI) are almost identical for the tread base compounds containing VB and RB, but a slight decrease in tensile strength was observed for the compounds containing HTB, most probably because of the poor distribution of carbon black [Fig. 4(c)] or possibly because of a lower elongation of break.

The modulus (Table VI) for the compounds containing HTB and RB is lower compared with that of the compounds containing VB. The main reason for this is the partial breakage and loosening of the agglomerate as a result of the heat treatment (confirmed by SEM photographs). Another possible reason is that, during extension, for the compounds containing RB or HTB no extra surface was exposed as surface area had already increased, but for compounds containing VB more surface



TABLE VII  
Tread Base Compound Containing FEF (N550) Black

Properties	Unaged compound containing VB (N550)	Aged compound containing VB (N550)	Unaged compound containing HTB (N550)	Aged compound containing HTB (N550)	Unaged compound containing RB (N550)	Aged compound containing RB (N550)
Hardness (Shore A)	55	60	54	60	53	58
Tensile strength (MPa)	22.2	13.6	20.1	15.4	22.4	15.9
100% modulus (MPa)	1.9	2.5	1.6	2.3	1.4	2.0
200% modulus (MPa)	5.0	6.2	4.3	5.6	3.7	5.0
300% modulus (MPa)	8.9	10.0	8.1	9.4	7.4	9.2
Elongation at break (%)	566	376	562	434	588	462

VB: virgin black; HTB: heated-treated black; RB: regenerated black.

area was available during extension, yielding more modulus. But modulus for compounds containing RB is lower compared with that of HTB, which may be because of compounds containing RB having the slowest cure rate and hence incomplete curing.<sup>9</sup>

Tear strength (Table VI) increased more for compounds containing HTB and RBN than for those containing VB.

The higher heat buildup (Table VI) of the compounds containing HTB and RB was a result of more filler–filler interaction, as in these carbon blacks the surface area is higher compared with that of VB. Some polar, acidic groups are also present, giving a better filler–filler interaction. These polar, acidic groups increase the surface energy of the blacks, and, as a result, during mixing carbon particles coalesce, giving a bigger agglomerate or cluster. As a result, filler–filler interaction increases, giving a higher heat buildup.<sup>9</sup>

### Aging study

After accelerated aging for 2 weeks at 70°C, hardness (Table VII) for all the compounds increased and the extent of the increase in hardness was almost same in all compounds (irrespective of the type of black).

After aging tensile strength decreased in all cases, but the relative decrease in tensile strength was maximum for the compound containing VB, minimum for the compound containing HTB, and medium for the compound containing RB.

After aging the modulus increased in all compounds, but the relative increase in modulus was always higher in compounds containing HTB and RB compared with that of compounds containing VB. The extent of the increase in modulus was comparable for compounds containing HTB and RB.

Elongation at break after aging decreased, but the relative decrease was maximum in the compound containing VB, medium for the compound containing HTB, and minimum for the compound containing RB. Because the rate of curing for the compounds containing RB and HTB were lower compared with that of those containing VB, these compounds have residual curing. So, after aging further, curing takes place, giving increased hardness and modulus.<sup>9</sup>

### CONCLUSIONS

1. Regenerated carbon black has a higher surface area because of the increase in surface roughness, compared with the respective virgin black.
2. The structure of regenerated black remains almost the same with that of the virgin black.
3. The agglomerate size of regenerated black is lower compared with that of virgin black.
4. Some polar acidic groups such as carbonyl, hydroxyl, carboxylic, thiocarbonyl, thiophenol, and chlorine groups are incorporated into regenerated carbon black.
5. Compounds containing regenerated black have a lower state of cure (undercure), lower cure rate, lower dispersion, lower hardness, and lower modulus than do compounds containing virgin black.
6. Compounds containing regenerated black have comparable tensile strength with respective compounds containing virgin black.
7. Compounds containing regenerated black had better scorch safety, a higher optimum cure time, higher tear strength, higher heat buildup, and higher elongation at break than do respective compounds containing virgin black.
8. Compounds containing regenerated black have better aging properties than do compounds containing virgin black.

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