OPTIMIZATION OF EPDM RECLAIM IN EPDM COMPOUNDS

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Ethylene-propylene rubbers (EPDM) continue to be one of the most widely used and fastest growing synthetic rubbers being used in specialty and general-purpose applications. In this study, EPDM-reclaim has been used as partial replacement for virgin EPDM, which eventually reduce the production cost, provided the material properties do not deteriorate significantly. Characterisation of vulcanisates illustrate that addition of up to ten parts of EPDM reclaim to virgin rubber do not significantly deteriorate vulcanisate properties. Tensile, tear, and hardness properties were maintained almost as that of the virgin EPDM vulcanisate. Also, there is no drastic increase in heat build-up after reclaim addition. There is only a marginal reduction in thermal stability with the incorporation of EPDM reclaim. The above results were well supported by swelling analysis and dispersion study which show that as the reclaimed rubber content increases, the level of dispersion gradually deteriorates. From the findings it may be concluded that incorporation of up to 10 parts reclaimed EPDM is justifiable in EPDM vulcanisates.

Keywords: EPDM, Mechanical properties, Reclaimed EPDM rubber

INTRODUCTION

Extensive utilization of rubber in various applications causes a problem in the disposal of the resulting rubber products after service. Among various possibilities of handling used and worn-out products, one of the most commonly applied methods is to dump them in a landfill, creating potential breeding places for disease-carrying insects. Furthermore, these piles can catch fire, which are practically impossible to extinguish and cause air, soil and surface-water pollution. Therefore, recycling of used rubber products is considered a way to save the environment and to reduce the material cost by reshaping the used rubber into a new product or otherwise replacing some fraction of the virgin material. Reclaimed rubber is an interesting raw material as it reduces the production costs of new rubber articles, and saves energy through shorter mixing times and lower power consumption.

Unlike thermoplastics, elastomers are thermoset which makes them more difficult to recycle. In order to make them re-processable, the three-dimensional network has to be broken down, the so-called reclaiming process. In this process, either sulphur crosslinks connecting the polymer chains or carbon-carbon bonds within the polymer backbone are to be broken. The first

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process is the preferred one, as the backbone of the polymer remains intact. Advantages of reclaimed rubber include shorter mixing time, lower power consumption and heat build-up, faster processing, lower thermoplasticity, reduced die-swell and shrinkage, higher cure rate, minimum reversion, and good ageing properties (Adhikari, 2000).

Ethylene propylene diene rubber (EPDM), with its highly saturated polymer back bone, is widely used to make rubber products with improved ageing and ozone resistance. EPDM reclaim as a replacement for virgin raw material can reduce the production cost. However, it is necessary to ensure that the material properties do not deteriorate significantly. Formulations with reclaimed rubber have lower processing temperature, and the material has a higher dimensional stability during calendaring and extrusion due to the residual three-dimensional network, which remains largely unaffected by the reclamation process. Improvement of ageing resistance is the most important advantage for cured articles containing reclaimed rubber.

Investigations were carried out on the utilization of waste EPDM as well as reclaimed EPDM rubber. The processability, curing characteristics and vulcanisate properties of EPDM compounds containing waste EPDM powder has been explored (Jacob, 2003). Mechanical properties of the epoxy/waste EPDM rubber composite also have been studied (Valasek, 2014). Two types of ethylene–propylene–diene monomer (EPDM) rubbers, namely an efficiently vulcanized (EV) and a semi-efficiently vulcanized (SEV), have been used to produce devulcanisates and most of the reclaims thus produced show slightly inferior mechanical properties compared to the virgin rubber. Also surface imperfections were observed on the vulcanisate containing high levels of devulcanising agent (Sutanto, 2006). EPDM-based ground waste (W-EPDM) is reutilized as rubber matrix in plug-material compounds for automotive applications. Effect of particle size and the amount of W-EPDM were studied. A new trial product of maleated ethylene - propylene rubber (EPM) and bitumen were used to improve interphase adhesion of the waste and the virgin EPDM (Karaagac, 2013). A ternary blend of polypropylene (PP), high density polyethylene (HDPE) and mechano-chemically devulcanised EPDM has been prepared using a laboratory twin-screw extruder (Jalilvand, 2007). Due to the high price of synthetic rubbers and growing concern about environmental issues, reclaimed rubber has been used as a component of a blend with virgin rubber or as a replacement for the virgin material in many applications (Premachandra, 2011).

In the present investigation, part of the virgin EPDM rubber has been replaced by EPDM reclaimed rubber and its effect on various vulcanisate properties has been studied.

MATERIALS AND METHODS

Characterization of reclaimed EPDM

The EPDM reclaim was procured from M/s. Gujarat Reclaim, Mumbai. The material has been characterized and its properties are given in Table 1.

Formulations used for the preparation of EPDM vulcanisates are given in Table 2. EPDM (Keltan 8570) was procured from Lanxess India Ltd., Thane. All the other chemicals used in the formulation are of rubber grade.

Table 1. **Properties of EPDM reclaim**

Mooney viscosity, ML(1+4) 100 °C, MU	30.1
Volatile matter, %	0.73
Acetone extract, %	30
Ash content, %	14
Carbon black, %	33
Rubber hydrocarbon content, %	25

Vulcanisates with varying reclaim loadings and their respective designations and descriptions are shown in Table 3.

Characterization of EPDM vulcanisates

Cure characteristics of the EPDM compounds were measured on a Monsanto rheometer (MDR 2000) as per ASTM D5289 and Mooney viscosity was measured using a Mooney viscometer (MV 2000, Alpha Technologies, USA). The EPDM vulcanisates were subjected to various physical characterisation techniques. Tensile properties and tear strength were determined using a universal testing machine, UTM (Zwick/Roell, Z005) with a cross-head speed of 500 mm/min using dumb-bell shaped and angle tear specimens respectively. Hardness was measured using a Shore A Durometer, Heat build-up was determined using BF Goodrich Flexomer, and dispersion was examined with a Dispergrader (Dynisco, USA). Thermal degradation study of the various vulcanisates has been carried out using a thermogravimetric analyser (Q 500, TA Instruments, USA).

The swelling study of the rubber specimens was carried out in toluene at ambient condition (27 °C) for 48h. Volume fraction of rubber (V_r) was calculated using the following equation (Zheng, 2004)

$$V_r = \frac{[(D - FW_i)\,\rho_r^{-1}]}{[(D - FW_i)\,\rho_r^{-1}] + A_0\rho_s^{-1}}$$

where, V_r is volume fraction of rubber in the swollen gel; *D*, the de-swollen weight of the composites; *F*, the fraction insoluble; W_i, the initial weight of the sample and A_0, the amount of solvent imbibed. ρ_r is the

Table 2**. Formulations of EPDM vulcanisates**

	(phr)			
EPDM (Keltan 8570)	100	90	80	70
EPDM reclaim (RHC 25%)	0	40	80	120
ZnO	5	5	5	5
Stearic acid	2	2	2	2
TDQ	2	2	2	2
Carbon black (GPF)	130	117	104	91
Naphthanic oil	100	88	76	64
MBT	1.5	1.5	1.5	1.5
Sulfasan R (DTDM)	1.2	1.2	1.2	1.2
ZDBC	2.5	2.5	2.5	2.5
TMTD	0.5	0.5	0.5	0.5
Sulphur	0.5	0.5	0.5	0.5

Table 3. **EPDM vulcanisate designations and descriptions**

Sample designation	Description
EP-EPRR-0	EPDM with 0 phr EPDM reclaim
EP-EPRR-10	EPDM with 10 phr EPDM reclaim
EP-EPRR-20	EPDM with 20 phr EPDM reclaim
EP-EPRR-30	EPDM with 30 phr EPDM reclaim

density of the rubber, while ρ_s is the density of the swelling solvent.

RESULTS AND DISCUSSION

Cure characteristics and Mooney viscosity

The t_5, cure index (t_{35}-t_5), and Mooney viscosity values of EPDM vulcanisates are reported in Table 4. It can be concluded that the presence of reclaimed EPDM has no significant influence on the scorch value and cure index. Whereas the Mooney viscosity is considerably affected by the amount of reclaimed EPDM in the compounds. The more the amount of reclaimed rubber incorporated the lower the Mooney viscosity becomes.

Physical properties

In Figures 1a and b, tensile strength and 300 per cent modulus are plotted against reclaimed rubber loading. There is a gradual reduction in tensile strength with reclaimed rubber loading. The change is steeper up to about 20 phr and then it appears to be tapering off. Also, it is observed that the tensile strength is reduced only marginally after ageing at 100°C for 168 hours. Similar effects are noted in the case of modulus also. The fall in modulus appears to be

Table 4. **Mooney viscosity and scorch values of EPDM compounds**

	t_5 (min.)	Cure index (t_{35}-t_5), min.	Mooney viscosity [ML (1+4) 100°C], MU
EP-EPRR-0	6.03	3.43	44.2
EP-EPRR-10	5.87	3.47	41.3
EP-EPRR-20	6.03	3.46	40.8
EP-EPRR-30	5.88	3.59	37.7

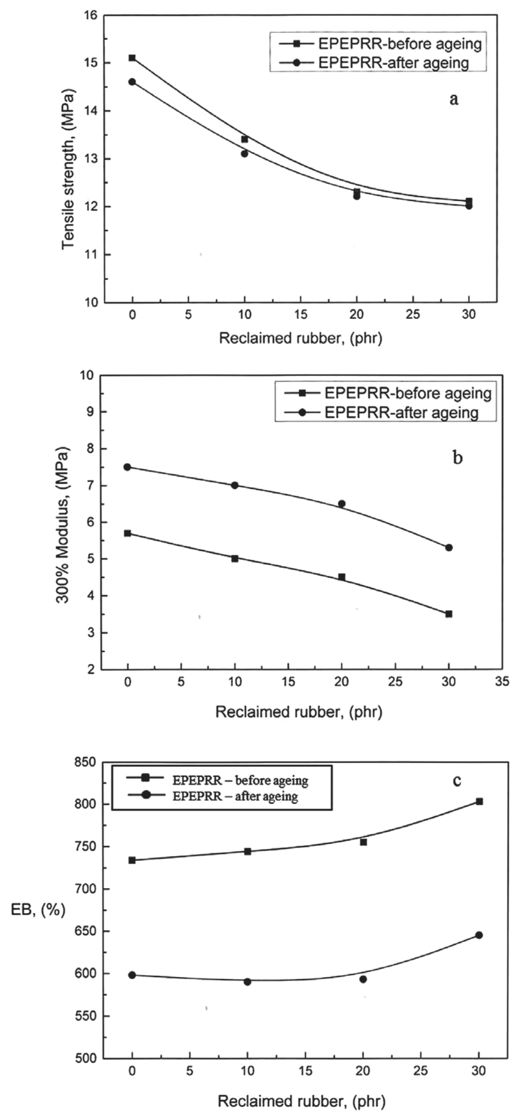

Fig. 1a-c: Tensile properties of EPDM vulcanisates progressive as the reclaim content increases. However, ageing causes a significant increase in modulus. Figure 1c depicts the variation in elongation at break with reclaimed rubber loading. There is a slight increase in elongation at break with reclaimed rubber loading which becomes more significant at higher loadings. Ageing causes a consistent reduction in EB. The increase in elongation at break and decrease in modulus is attributed to the increasing proportion of low molecular weight fraction of polymer due to reclaimed rubber.

Hardness

Figure 2 depicts the variation of Shore A hardness of EPDM vulcanisates. It can be seen that hardness of the vulcanisate gradually decreased on addition of reclaimed EPDM, which can be attributed to the presence of low molecular weight polymer fractions in the reclaimed rubber. The increased hardness after ageing is expected as vulcanisates undergo additional cross-linking upon heat ageing. The hardness values are in correlation with the modulus values of vulcanisates with the incorporation of reclaimed rubber.

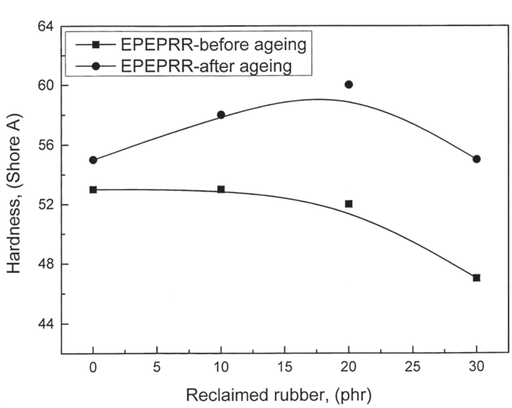

Fig. 2. Hardness of EPDM vulcanisates

Tear strength

EPDM vulcanisate exhibited higher tear strength, while on addition of reclaimed rubber the tear strength decreased dramatically (Figure 3). The decrease in tear strength with more reclaimed rubber content is quite natural as the tear strength is directly related to the molecular chain length. This shows that addition of reclaimed rubber deteriorate tear strength significantly which is detrimental in many applications.

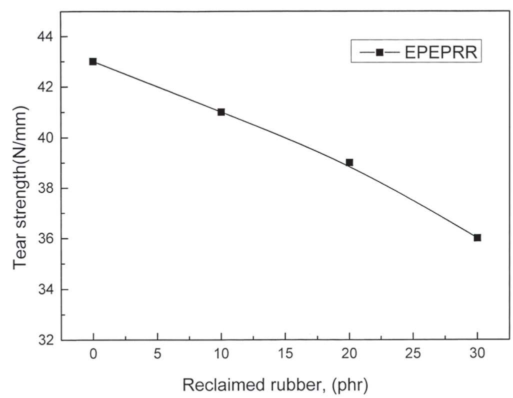

Fig. 3. Tear strength of EPDM vulcanisates

Heat build-up

Heat generation is an important parameter in certain rubber products such as tyre, conveyor belt etc. As shown in Figure 4, the virgin EPDM vulcanisate showed lower heat build-up, while the vulcanisate with reclaimed rubber showed higher heat build-up values. This trend was observed up to 20 phr reclaimed rubber and then tapered off showing no further effects upon higher loadings.

Dispersion study

Many properties of rubber such as tensile strength, tear strength, fatigue resistance,

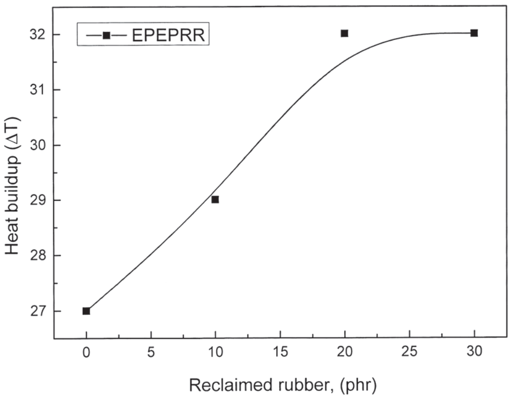

Fig. 4. Heat build-up of EPDM vulcanisates

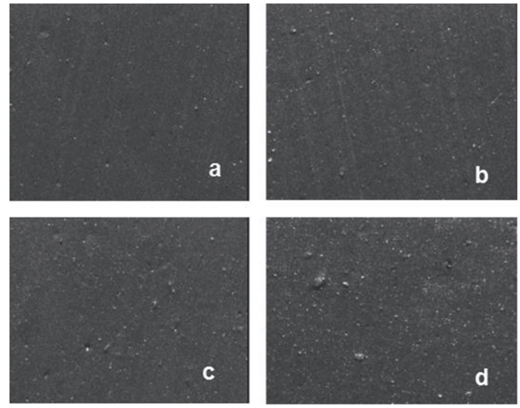

Fig. 5a-d. Dispergrader images of vulcanisates (a) EP-EPRR-0, (b) EP-EPRR-10, (c) EP-EPRR-20, and (d) EP-EPRR-30

and abrasion resistance are affected by dispersion of various ingredients, particularly reinforcing fillers such as carbon black. Table 5 displays the dispersion ratings of all four EPDM vulcanisates. Here, x, y, and z values indicate the filler distribution, agglomerate size and percentage dispersion of the vulcanisate, respectively. It is evident that the small additions of reclaimed EPDM result in some reduction in x, y and z values while with 30 phr addition there is large drop in these readings. This implies that mixing large quantity of reclaimed rubber in to virgin EPDM results in poor dispersion which again results in inferior vulanisate properties. The readings are well supported by the respective images of the vulcanisates which is shown in Figures 5a-d. The presence of improperly dispersed fillers is shown by irregularities that form "bumps" on the surface, indicating a poorer dispersion of fillers. From the images it may be confirmed that the larger the amount of reclaimed rubber in compound, the poorer the overall dispersion of the vulcanisate. The poor dispersion with the presence of reclaimed rubber may be attributed to the poor compound viscosity resulting from the lower molecular weight of reclaimed rubber fraction.

Swelling analysis

The volume fractions of rubber in various vulcanisates are reported in Table 6. With the presence of up to 20 phr of reclaimed rubber, the volume fraction of rubber slightly got increased. This may be due to the additional amount of curatives with the reclaimed rubber. It may be noted that, with higher amount of reclaimed rubber (30 phr), the volume fraction of rubber gets reduced. This may be because of the poor dispersion resulting from the addition of higher

Table 5. **Dispersion rating of EPDM vulcanisates**

	EP-EPRR-0	EP-EPRR-10	EP-EPRR-20	EP-EPRR-30
Dispersion ratings	x=8.2, y= 10, z=98.6	x=7.8, y= 9.9, z=96.4	x=7.8, y= 9.9, z=95.7	x=6.9, y= 9.8, z=92.1

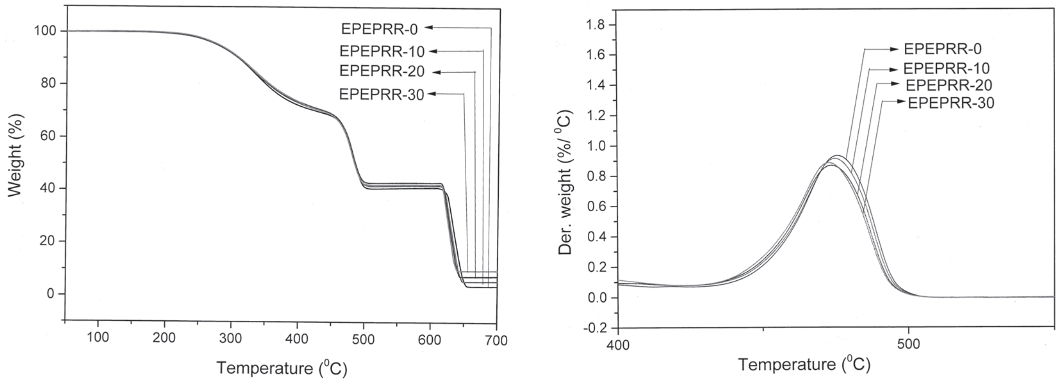

Fig. 6a-b. Thermal degradation curves and the corresponding derivative curves of EPDM vulcanizates

Table 6. **Volume fraction of rubber in EPDM vulcanisates**

Sample designation	Volume fraction of rubber, V_r
EP-EPRR-0	0.256
EP-EPRR-10	0.265
EP-EPRR-20	0.262
EP-EPRR-30	0.241

Table 7. Thermal degradation temperature of EPDM vulcanizates

Sample designation	Degradation temperature, °C
EP-EPRR-0	475.8
EP-EPRR-10	474.8
EP-EPRR-20	473.7
EP-EPRR-30	472.8

amount of reclaimed rubber which dominates the curative effects.

Thermo-gravimetric analysis

Thermal degradation curves and their corresponding derivative curves of EPDM vulcanisates are shown in Figures 6a-b. It may be observed that all the samples exhibit a two stage thermal degradation. The hydrocarbon content in the vulcanisates decomposes in the 400-430 °C range. Again when oxygen is passed at 600 °C, oxidative degradation occurs to the vulcanisates. Incorporation of EPDM reclaim imparts a gradual reduction in thermal stability of EPDM vulcanisate. This is due to the presence of low molecular weight polymer fraction in the reclaimed rubber.

CONCLUSIONS

Ethylene-propylene rubbers (EPDM) continue to be one of the most widely used and fastest growing synthetic rubbers having both specialty and general-purpose applications. Incorporation of reclaimed EPDM to virgin EPDM compound has been analysed based on vulcanisate properties. Mechanical properties such as tensile strength and modulus show gradual deterioration while hardness increases to some extent then decreases in correlation with the modulus values. Tear strength shows a drastic decrease while the heat build-up increases to some extent and then taper off. Overall results reveal that an addition of 10 phr of reclaimed EPDM in to EPDM compounds is acceptable when all the properties are taken in to consideration. Further addition of reclaimed rubber is not recommended as far as various vulcanisate properties are concerned. Also, it may be noted that the presence of reclaimed EPDM rubber does not adversely affect the ageing properties of vulcanisates. Also, addition of reclaimed rubber did not affect the thermal degradation stability significantly. Mixing large quantities of reclaimed rubber with virgin EPDM results in poor dispersion which eventually give vulcanisates with inferior properties.

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