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Effect of ground tyre rubber devulcanisates on the properties of a passenger car tyre tread formulation

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Abstract. Extensive research on recycling processes, in particular for waste tyre material, is necessary due to increasing raw material costs, diminishing resources and growing awareness of environmental issues. One of the preferred methods is devulcanisation, in which only sulphur cross-links are broken, while the polymer chains remain intact. In this study, optimised processing conditions for the devulcanisation of whole passenger car tyres using diphenyldisulphide (DPDS) as a devulcanisation aid were applied. The devulcanised ground tyre rubber (D-GTR) obtained from the process was blended on top of the original tyre tread formulation at different concentrations. The cure characteristics and mechanical properties of the re-vulcanised blends are evaluated in comparison to the original compound. The results so far indicate that the addition of D-GTR influences the properties of the blend, but to a lesser extent than the commercially used powder or reclaim types do, for which loadings less than 5% can be applied. The main reason for the influence of D-GTR on the rubber properties is inhomogeneities in the D-GTR/virgin rubber blend.

Introduction

Several methods have been developed to reclaim rubber waste, e.g. thermo- and chemo-mechanical processes, microwave regeneration, microbial action, ultrasonic regeneration, and other methods of which a comprehensive summary has been published by Adhikaari *et al* [1] and Myhre *et* al [2]. However, the properties of reclaimed material are relatively poor compared to the original material and hence limiting the applications, especially for high quality products such as tyres. Therefore, extensive research on processes for high-quality recycling materials based on waste tyres is still necessary.

The objective of this work is to devulcanise whole ground passenger car tyre rubber using conditions optimised in an earlier study[3], then add this material on top to the original tyre tread formulation in different concentrations. The curing characteristics and mechanical properties of these compounds were investigated and compared with those of the original compound.

Experimental

Materials. The recycled ground tyre rubber (GTR) from whole passenger car tyres with an average particle size of 0.35 mm obtained from Rubber Resources, Maastricht, the Netherlands, was used in this study. The GTR comprised a 50% rubber content mainly based on synthetic rubber and 28% carbon black, 7% acetone extract and 14% ash. Processing oil used in this study was treated distillate aromatic extract (TDAE) from Hansen & Rosenthal, Germany. Diphenyldisulphide (DPDS) used as devulcanisation aid was obtained from Sigma-Aldrich, Germany.

Preparation of the De-vulcanisates. The devulcanisate was prepared batchwise using the optimised conditions as elaborated by Sitisayidah [3]. The devulcanisation was done in an internal mixer (Brabender Plasticorder PL-2000), having a mixing chamber volume of 50 cm^3 and a cam-type rotor.

Re-vulcanisation of Devulcanised Rubber. The devulcanisates were then incorporated at 10, 30 and 50 phr on top of original tyre tread compound, with and without adjustment of the carbon black and curing system as depicted in Table 1. The adjustment of carbon black was based on equal hardness of the materials. Mixing was done in an internal mixer Brabender Plasticorder PL-2000 with a mixing chamber volume of 50 cm^3 and chamber temperature of 75˚C. A fill factor of 0.7 and constant rotor speed of 60 rpm were used. The mixing time was about 4 minutes and then the compound was sheeted using a two-roll mill.

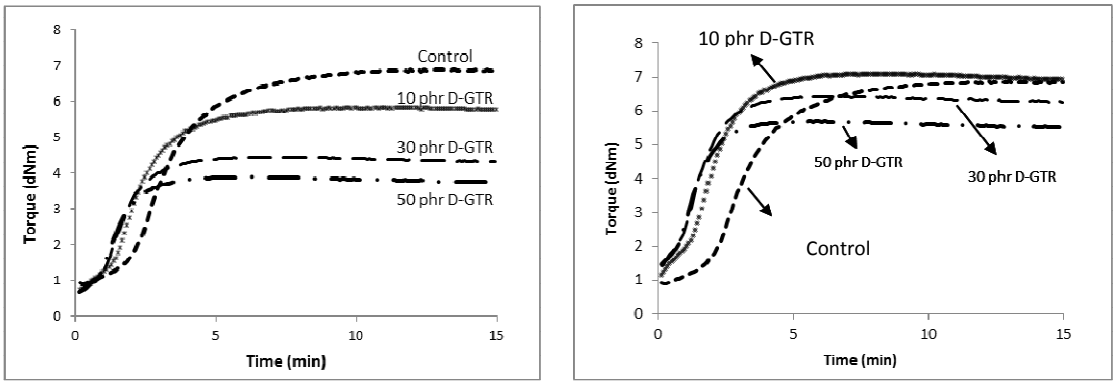
Table 1. Tyre tread compound containing various contents of D-GTR with adjustment of carbon black and curing system.

	1	**2**	**3**	**4**	**5**	**6**	**7**
Tread-supplied*	100	100	100	100	100	100	100
D-GTR	0	10	30	50	10	30	50
Carbon black	0	0	0	0	8	15	20
Sulphur	0	0.25	0.75	1.25	0.25	0.75	1.25
TBBS	0	0.15	0.45	0.75	0.15	0.45	0.75

*100 phr of new rubber

Results and Discussion

Cure Characteristics. Fig. 1 (a) – (b) shows the rheograph of compounds containing 10, 30 and 50 phr of devulcanised ground tyre rubber (D-GTR) on top of the tyre tread formulation with and without the addition of additional carbon black. A significant decrease in cure and scorch time is observed with the addition of extra carbon black. The minimum torque, which is a measure of stock viscosity, increases with additional carbon black; similar to the observation made by Maridass *et.al* [4]. The addition of extra carbon black resulted in a higher maximum torque compared to the compounds before adjustment; however, for 30 and 50 phr D-GTR the maximum torque is still lower compared to the original compound.



(a) Without adjustment (b) With adjustment of carbon black

Figure 1. Cure curves of tyre tread formulation with various contents of D-GTR: a) without adjustment and b) with adjustment of carbon black

Physical Properties. Fig. 2 and 3 show the hardness and modulus (M100% and M300%) results of the compounds containing various contents of D-GTR with and without carbon black adjustment respectively. Modulus is a measure of the stiffness of the material. It can be seen that addition of D-GTR without further adjustments tends to decrease hardness and modulus at 100 % and 300 % elongation. Incorporation of additional carbon black increases the modulus and hardness values due to the lower elastomer content in the matrix.

Tensile strength marginally increases whereas elongation at break decreases with adjustment of the carbon black content (Fig. 4). The additional carbon black present in the D-GTR/virgin blend compounds inhibits the mobility of the rubber chains as they are partly bound to the carbon black surface, thus reducing the flexibility of the material. The deterioration of the tensile properties by the addition of D-GTR compared to the original compound, independent of the adjustment of carbon black, can be attributed to several facts:

- Weak spots from remaining cross-links of the D-GTR [1];
- The presence of inhomogeneities in the blend, e.g. of the modulus across the interphase between the matrix and the D-GTR phase;
- Migration of curatives between the blend phases which could affect the formation of the new crosslinks in spite of the fact that the cure system was adjusted;
- Deterioration of the tyre material due to aging during the service life;
- Contaminations in the whole tyre material.

Further developments of the devulcanisation process after this work have shown that the addition of 40% of devulcanised rubber to a passenger car tyre tread compound is feasible.

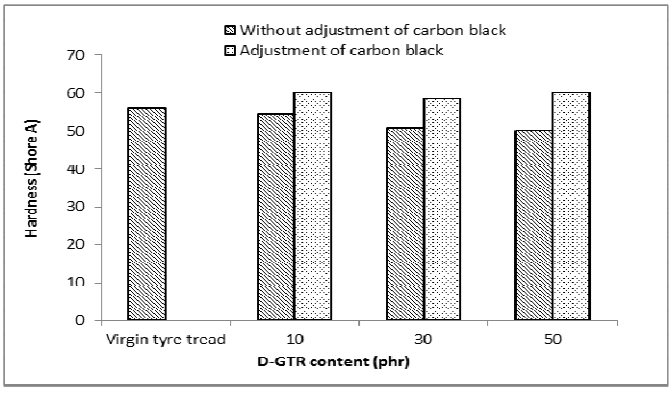


Figure 2: Hardness of typical tyre tread compound with various contents of D-GTR before and after carbon black adjustment

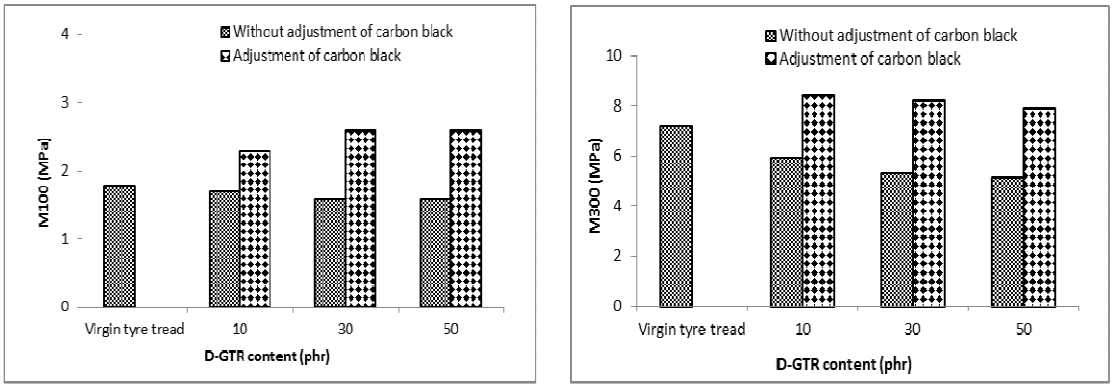
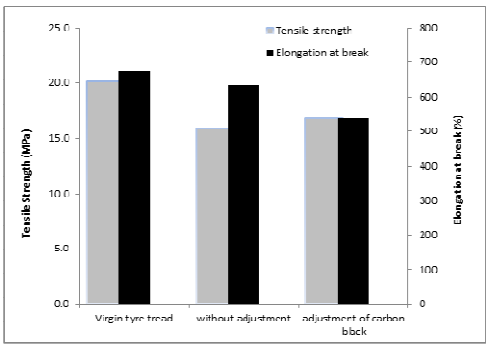
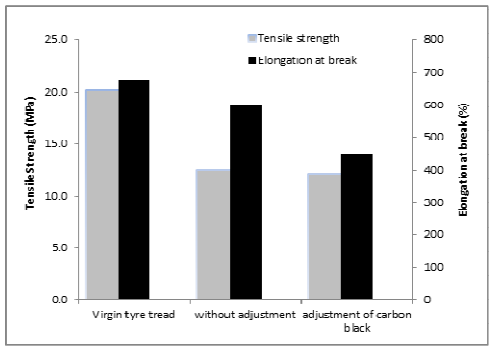


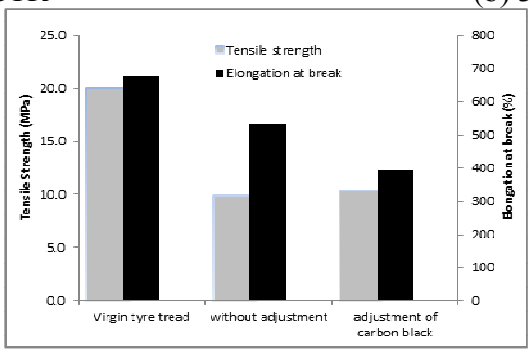
Figure 3: Moduli M100 and M300 of a typical tyre tread compound with various contents of D-GTR before and after carbon black adjustment



(a) 10 phr D-GTR



(b) 30 phr D-GTR



(c) 50 phr D-GTR

Figure 4: Tensile strength and elongation at break of a typical tyre tread compound with various contents of D-GTR before and after carbon black adjustment

Summary

The increasing content of D-GTR of up to 50 phr leads to a decrease in elongation at break, maximum torque values of the revulcanisates and some reduction in tensile strength of a passenger car tyre tread compound. The main reasons for the influence on the mechanical properties are inhomogeneities in the blend, migration of curatives and contaminations in the feedstock for the D-GTR. If these problems are overcome, a considerable amount of D-GTR can be added to the original compound without significant loss in properties. This has actually been proven in the meantime by further devulcanisation process optimisation. This shows the significant benefit of thermochemical devulcanisation over the commonly used powder or reclaim, for which loadings of less than 5% can be applied.

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