Journal of Engineering and Applied Sciences 12 (2): 232-238, 2017
ISSN: 1816-949X

Effect of Crumb Rubber Content and Particle Size on the Mechanical and Rheological Properties of Passenger Tyre Tread Composite

[1]Adnan A. Alshukri, [1]A.A. Faieza, [1, 2]S.M. Sapuan, [1]A.A. Nuraini, [3]Mohammed Al-Maamori
and [4]Dheaa S. Zageer
[1]Department of Mechanical and Manufacturing Engineering, Faculty of Engineering,
Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
[2]Laboratory of Biocomposite Technology,
Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia,
43400 UPM Serdang, Selangor, Malaysia
[3]Department of Rubber Material, Material Engineering, University of Babylon, Babylon, Iraq
[4]Forensic DNA Center for Training and Research, Al-Nahrain University, Baghdad, Iraq

Abstract: The purpose of this study is to determine the effects of the content and particle size of Crumb Rubber (CR) on the mechanical and rheological characteristics of the passenger tyre tread layer. Crumb rubber from ambient grinding of the whole ground tyre was used after it had been subjected to sieve analysis to produce six size categories 40, 150, 180, 250, 425 and 600 μm. Each category was incorporated in different fractions 20, 40 and 60 phr as a filler in a virgin Styrene Butadiene Rubber (SBR) matrix to examine the effect of CR loading on the properties of the blends produced. The results revealed that the mechanical and curing properties of the tread blend were improved as the CR particle size reduced while all the mechanical and rheological properties were reduced in a different manner when the CR content was increased. The findings of the investigation could be used as a handy tool and offer some insight into the material design stage to decide the amount and the grade of CR suitable for certain passenger tyre tread according to the performance required.

Key words: Recycling, crumb rubber, tyre tread, particle size, passenger

INTRODUCTION

The number of vehicles on the road grows rapidly each year over the world, leading to a similarly rapid increase in the worn tyres. According to a previous estimation, around 800 mln scrap tyres have been disposed of around the globe and this amount is expected to increase by approximately 2% each year (Beukering and Janssen, 2001) . The existence of a high cross-linking chemical structure causes end-of-life tyres to be non-biodegradable as they are made from non-environmentally materials (Wang *et al*., 2010). The huge amount of discarded tyers over the world has become a great ecological challenge. A common industrial process prior to any form of recycling comprises of shredding and grinding the tyres to extract a powder called Crumb Rubber (CR) in addition to the steel and textile content (Amari *et al*., 1999). CR has been proposed as a suitable reinforcement filler for composite materials as a feasible way to reduce the number of scrap tyres in added value applications (Meysami and Tzoganakis, 2010). Many technologies have been developed to introduce CR as a toughening agent for thermoplastic, thermoset polymers (Luo and Isayev, 1998), asphalt modifier (Mull *et al*., 2002) and many other applications. However, one of the main drawbacks for using CR as a filler is its low compatibility with most of the matrix materials used. This is because of the high molecular and crosslinking bonds (Yehia *et al*., 2004a, b). Several attempts have been made to utilize the CR by either chemical or physical methods. Most of these methods have succeeded, up to a certain limit, to incorporate the CR with different matrix materials with acceptable performance and mechanical properties (Colom *et al*., 2011). One of most important recycling fields for CR is the polymer matrix. CR can be used with all of the three groups of polymers, thermoplastic (Zhang *et al*., 2011), thermoset (Fan and Lu, 2012) and elastomer. For all of the three groups, the interface between the CR particles and the polymer matrix is the main issue which governs the final properties of the material (Ramarad *et al*., 2015). In general, CR is incompatible with the polymer matrix. This

Corresponding Author: Adnan A. Alshukri, Department of Mechanical and Manufacturing Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

deficiency can be reduced either by using chemical or physical processes to increase the CR surface activity or by particle size reduction to achieve finer dispersion of the CR in the polymer matrix (Mangaraj, 2004). The energy consumed to reduce the CR particle size is very high due to the energy spent on heat dissipation during the process. However, the CR fraction and particle size effects on the Synthetic Rubber Matrix (SBR) are arguable issues according to previous studies. Some researchers claim improvement in terms of mechanical properties with the incorporation of CR (Sreeja and Kutty, 2003, Li *et al*., 2012) and some claim deterioration (Han and Han, 2002). Therefore, for a particular application, a suitable CR content and particle size are important to achieve better performance at a low cost.

In this work, different particle sizes and fractions of CR have been incorporated in a Styrene Butadiene Rubber (SBR) matrix, in a recipe designed for manufacturing passenger tyre tread layer, to investigate the effect of each parameter on the mechanical properties and curing characteristics of the produced composite. A comparison has been made between three weight ratios of CR content, for each of the particle size categories to examine the ability of size reduction and CR content in replacement of virgin rubber material.

MATERIALS AND METHODS

The recipe used in manufacturing the passenger tyre tread layer has been utilized in this work, to produce a group of blends having same the composite elements with different CR particle sizes. Three sets of blends have been prepared, with each set having different particle size grades with the same amount of CR content, 20, 40 and 60 phr. The basic characteristics of the materials are given in Table 1, the rubber chemicals used were such as zinc oxide, stearic acid, sulphur, N-cyclhexyle-2-benzothiazyl sulphenamide (CBS), permanax (6PPD), flectol (TMQ) and phthalimide (CTP-100) were of commercial grade.

Mixing procedure: Rubber compounds with different CR particle size categories and amounts were prepared on a two roll mixing mill, having a friction ratio of 1:1.4. Three tread layer recipes were used to produce blends with 20, 40 and 60 phr CR content for each of the size categories in Table 1. The compound material specifications of the blends are given in Table 2. All the batch and master batch mixing processes were carried out in a laboratory with an open two roll mill (150 mm diameter and 400 mm working length). The temperature of the rolls was controlled at around 60°C by flushing water circulated through the hollow rollers of the mixer according to ASTM D 3182 (ASTM International, 2001a, b). During the first stage of mixing (master batch), the SBR rubber was mixed with reclaimed rubber, zinc oxide, stearic acid, carbon N375, process oil, TMQ, 6PPD and crumb rubber. The blends were left for eight hours before starting the final mixing process. The other rubber chemical materials were added during the second stage (batch) which included CBS as an accelerator, CTP-100 as a retardant and sulphur as a vulcanizing agent. The rubber mixes obtained were sheeted and prepared for the test and curing process.

Vulcanization process: After the rubber compounds were mixed, the blends were pressed and cured by using different molds for each particular mechanical property test:

- Tensile strength, elongation and tensile modulus tests were done by pressing and curing a square rubber sheet of (150×150×2 mm) at a temperature of 145±2°C and pressure 3.44 MPa for 45 min curing time
- Abrasion resistance test was done by producing a rubber disc (64Ø×12.00) mm at a temperature of 160±2°C and a pressure of 3.44 MPa for 20 min curing time
- A rubber disc (45Ø×3) mm cured at a temperature of 160±2°C, for a 15 min curing time was used for the hardness and specific gravity tests

All blends were cured in an electrically heated hydraulic press according to the condition and curing time for each test.

Cure characteristics: An Oscillating Disc Rheometer (ODR) was used at 185°C for 6 min to investigate the rheological properties of the blends. A rheometer model ODR2000 E from Monsanto-manufactured in the United Kingdom was used. The procedure was started by adding a piece of the uncured blend on the device die cavity when the device was working, the torque began to rise. The plotted curve showed the torque development rate accrued as the sample cured. The measured parameters were: maximum torque M_H, minimum torque M_L, time to two units of torque increase above minimum t_{s2}, time to torque at 90% of full torque M_{c90} development (cured time) t_{c90} and viscosity. The measurements were done according to ASTM 2084 (ASTM International, 2003).

Mechanical testing: Tensile strength, elongation and the modulus of the produced sheet were determined by a tensile testing machine type 10-T10 Monsanto, United Kingdom. A specimen cutting press was used to cut out test pieces (dumbbells) from the 2 mm thickness cured sheet. Three dumbbells were used for testing each property and the average value over the three samples was adopted. The across head speed was 500 mm min^{-1} and 25 mm length of the dumbbells was assigned as the

Table 1: The recipe used for 20, 40 and 60 phr CR filled rubber blend

Ingredients	Descriptions	Amount (phr) 10% wt	Amount (phr) 20% wt	Amount (phr) 20% wt
SBR 1502	Copolymer of styrene and Butadiene	100	100	100
Reclaim	The whole tyre reclaimed tyre compound	4	4	4
Carbon black	Carbon black N375	61	61	61
Process oil	Naphthenic	18	18	18
CBS	N-Cyclohexyl-2-Benzothizole Sulphonamide	1.6	1.6	1.6
Zinc oxide	Zinc oxide nodular (0.25% maximum lead)	1.5	1.5	1.5
Stearic acid	Fatty acids of carbon chain length predominantly C16-C18	1	1	1
6PPD	N-(1,3-dimethylbutyl)-N-phenyl-p- phenylenediamine	1	1	1
TMQ	Polymerised 2, 2, 4-trimethyl-1, 2-dihydroquinoline	0.5	0.5	0.5
Sulphur	Rhombic sulphur	2	2	2
CTP-100	N-(cyclohexylthio) phthalimide	0.1	0.1	0.1
Crumb rubber	Whole tyrecrumb 250-500 µm	20	40	60

Table 2: Details and sources of materials compounds

Materials	Characteristics	Values	Sources
SBR 1502	Viscosity	ML (1+4) 100°C	52±3
	Bound styrene	23.5±1.0 maximum	
	Specific gravity typical	0.95	Kumho, Korea
Reclaim	Mooney viscosity	ML (1+4)100°C 30-80	
	Specific gravity typical	1.16	Babylon, Iraq
Carbon black	Iodine adsorption value	92±5	
	Specific gravity typical	1.8	Doda Sanati,Iran
CBS	Melting point	95-107°C	
	Specific gravity (typical) 1.34	Flexsys, Belgium	
Zinc oxide	Zinc oxide content	99.0 minimum	
	Specific gravity (typical) 5.50	Nocil, India	
Stearic acid	Iodine value	max23	
	Specific gravity (typical)	0.85	Nocil , India
6PPD	Melting point	44-50°C	
	Specific gravity (typical)	1.00	Flexsys, Belgium
Sulphur	Sulphur content	99.0 Minimum	
	Specific gravity (typical)	2.10	Flexsys, Belgium
CTP-100	Melting point	89-95°C	
	Specific gravity (typical)	1.30	Bayer, Germany
Process oil (Naphthenic)	Specific gravity (typical)	0.87-0.89	
	Viscosity at 20°C redwood No. 1	320-400	Aldora refinery, Iraq
Crumb rubber	Specific gravity (typical)	1.13	Babylon factory, Iraq

primary testing length. The tests were performed according to ASTM D-412 (ASTM International, 2003). An abrasion resistance test was performed by a machine from Wallace, England. The test was achieved by applying 500 revolutions for the cured disc with dimensions and cure conditions as mentioned above according to ASTM D 5963 (ASTM International, 2001).

Hardness measurements: Hardness test data was collected using a rubber disc (45Ø×3) mm cured at a temperature of 160±2°C, for 15 min curing time. The measurements were carried out according to ASTM D 1415 using a durometer from Wallace, England. The unit of hardness was expressed in International Rubber Hardness Degree (IRHD).

RESULTS AND DISCUSSION

Mechanical properties: Crumb rubber CR produced from the whole tyre was subjected to sieving analysis to classify six categories of particle size as shown in Table 3. Each size was incorporated with rubber compounds according to the recipes mentioned in Table 1 to obtain the laboratory blends. All blends had the same mixing and curing procedures but differed in CR particle size and amount. The mechanical properties were measured and shown in Table 4.

Table 3: Particle size categories

Size name	Size in mesh	Size (µm)
A	<350	<40
B	350-100	40-150
C	100-80	150-180
D	80-60	180-250
E	60-40	250-425
F	40-20	425-600

It can be seen that when the CR particle size reduced, the tensile strength increased. This was true for all amounts 10, 20 and 30%. These results were expected because a small CR particle size gives better dispersion and a smooth curative material migration as compared to the larger size. In addition, a reduction of filler particle size increased the interfacial surface area and enhanced the rate of bonding and crosslinking (Colom *et al*., 2009). Howerve, Fig. 1 reveals that the reduction trend was approximately linear and there was no significant increase in tensile strength <150

Table 4: Mechanical properties

	Tensile strength (Mpa)			Elongation at break			Abrasion (%)			Hardness (IRHD)		
Particle size	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
A	15.82	13.80	12.184	529.0	485.00	433.0	0.49	0.676	0.85	61	53	56
B	15.60	13.69	11.900	512.5	455.00	425.0	0.49	1.021	0.88	60	52	55
C	15.35	12.99	11.690	480.0	442.00	412.0	0.52	1.023	0.90	59	51	50
D	14.22	12.09	10.820	472.0	426.50	408.0	0.53	1.119	0.94	59	53	53
E	13.17	10.64	10.070	414.0	399.00	390.0	0.58	1.134	1.02	58	52	52
F	11.38	10.06	7.5200	452.5	380.25	336.5	0.62	1.188	1.06	56	52	54

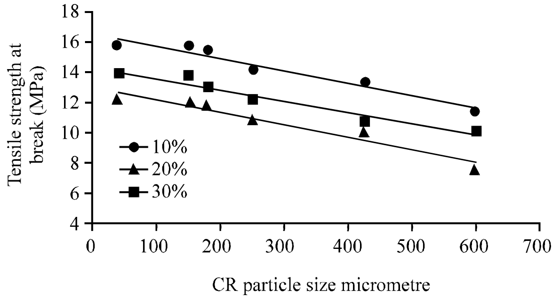

Fig. 1: Effect of particle size on tensile strength of tyre tread layer at diffrent CR loading

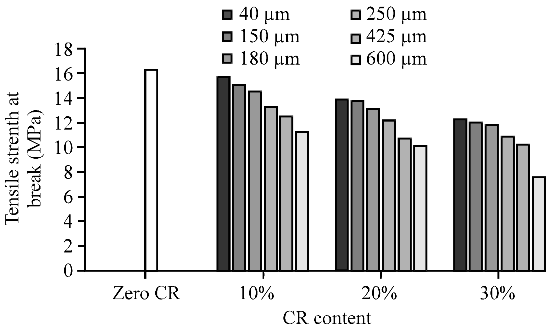

Fig. 2: Effect of particle size and CR content on the tensile at break

micrometre CR categories. The trend was in good agreement with previous profiles investigated for EPDM (ethylene propylene diene monomer) by Klingensmith and Baranwal, 1998). Figure 2 shows a comparison between tensile at the break for a blend with no CR content and multi-CR loading with different particle sizes. It is clear from this figure that the lowest particle sizes gives a better value for all CR content in comparison with the zero CR content blend. Figure 3 shows there has been a gradual decrease in elongation with an increase in particle size of CR. These results were expected, because elongation was proportional to the crosslinking density and smooth rubber molecular slip, while the existence of a large particle size means, less surface area, less filler-matrix bonding, rough slip movement of rubber molecules and a high probability of crack growth. These results agree strongly with work done by Seung Cheol (Han and Han, 2002).

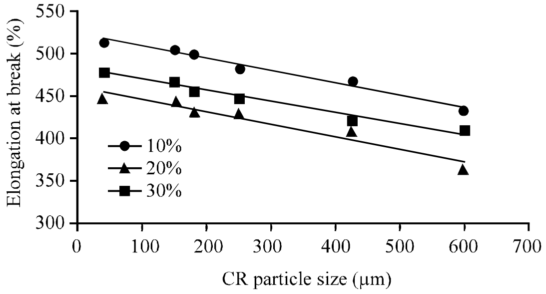

Fig. 3: Effect of partilce size on the elongation at break

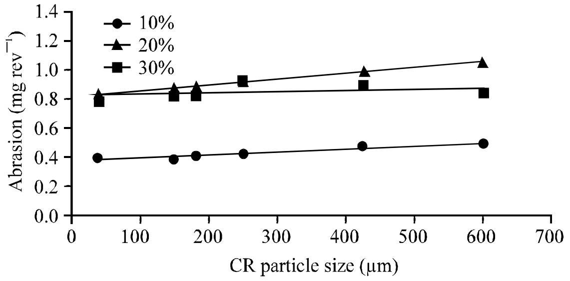

Fig. 4: Effect of CR loading and particle size on abrasion resistance

The most important mechanical property for the tyre tread layer is abrasion resistance. This property is more dependent on the abrading surface and slightly on loading level than on the particle size. As shown in Fig. 4, there was no significant change with CR particle size reduction but there was an increase in abrading rate with an increase in CR content. This could be attributed to the reduction in crosslinking rate due to the accumulation of CR particles. This result agreed with the investigation carried out by Li *et al.* (2004).

The plot of hardness versus CR particle size and the multi-CR is shown in Fig. 5. The hardness reduced slightly as the CR content increased. On the other hand, the CR particle size did not appear to have a clear effect. This could be because of the hardness is dependent on the crosslink density which was affected by an increase in the CR loading. The shortest fragment and lack of sulphur led to a low crosslink density (Li *et al.*, 2012).

Table 5: Curing behaviors

	Max. torque			Viscosity			Scorch time			Cure time (IRHD)		
Particle size	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
A	41.69	32.32	37.35	21.06	21.249	22.815	0.977	0.866	0.933	2.200	2.177	2.277
B	37.47	30.75	35.00	20.46	20.709	23.120	0.990	0.900	0.900	2.155	2.230	2.380
C	37.94	30.23	32.75	19.93	23.166	23.598	1.010	0.900	0.940	2.260	2.310	2.320
D	37.99	29.68	33.39	18.82	23.220	25.866	1.030	0.910	1.020	2.210	2.280	2.380
E	35.69	29.67	35.45	18.49	24.003	26.487	1.010	0.960	1.040	2.170	2.480	2.490
F	36.66	29.38	37.91	17.25	25.461	29.241	1.030	0.970	1.030	2.200	2.430	2.450

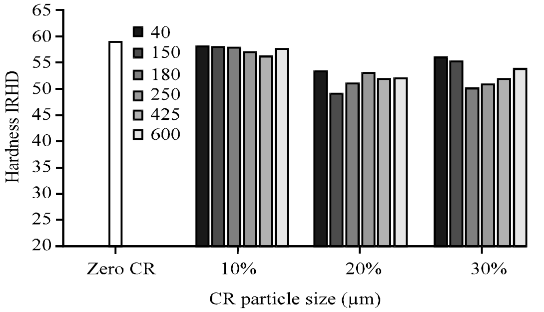

Fig. 5: Effect of CR loading and particle size on hardness

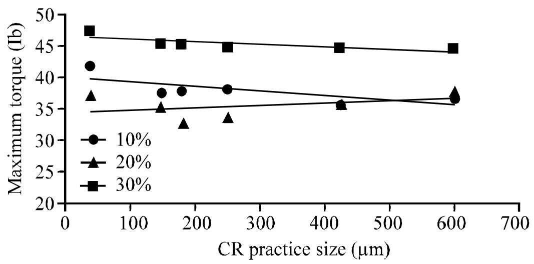

Fig. 6: Maximum torque versus CR particle size and content

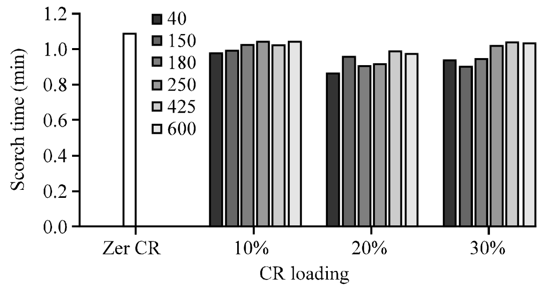

Fig. 7: Scorch time vs CR loading and practicle size

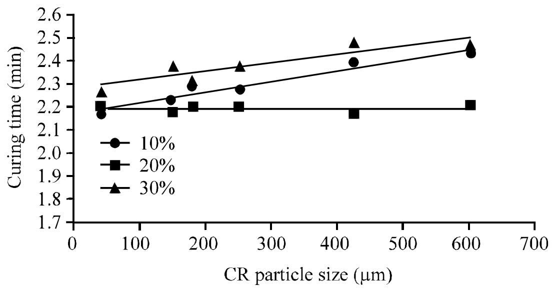

Fig. 8: Curing time vs CR particle size

Curing behaviors: The curing characteristics are listed in Table 5 for all the blends filled with different CR particle size categories and content. The results show that there was a reduction in maximum torque as the particle size increased as shown in Fig. 6. A possible explanation for these results could be the reduction in bonding rate between the CR particles and the rubber matrix due to reducing the overall CR surface area. The results also showed that while the CR loading increased, the maximum torque is reduced. These results agreed well with the previous investigations and could be explained by the migration of sulphur from the matrix rubber to the CR and by increasing of the CR concentration in the compound mixture (Gibala and Hamed, 1994).

The effect of CR particle size versus scorch time is shown in Fig. 7. The scorch time represents a point where the rubber compound starts to crosslink. From this graph it can be seen that as the CR loading increase, the scorch time was reduced and the same trend occurred when the particle size was reduced. There could be several possible explanations for this phenomenon. The presence of a crosslink precursor and unreacted curatives in the CR particles or the existence of active crosslinking sites in the CR which led to an increase in the crosslinking rate upon heating (Sreeja and Kutty, 2002). The decrease in scorch time indicated that the cross-linking process started early and the scorch safety was low which would not be a desirable property in the industry.

The effect of CR particle size and content on curing time is presented in Fig. 8. The results showed that the curing time increased with an increase in both CR particle size and content. This result was expected because less CR loading means an increase in curing agent concentration and content and a small particles size gives a better distribution of curative material (Lamminmaki *et al*., 2006).

Viscosity is measure of process ease. It can be seen from Fig. 9 that the viscosity increased with the loading of CR. This was related to the molecular slip of the CR as

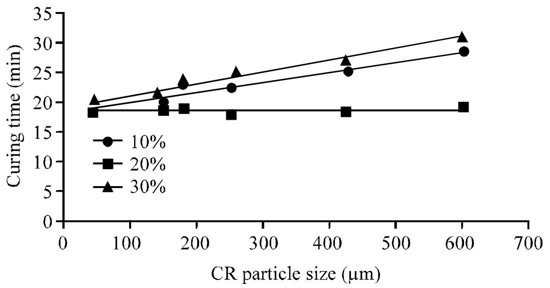

Fig. 9: Viscosity vs CR particle size

vulcanised material which does not flow and agglomerates in the rubber matrix led to an increase in the compound viscosity. This effect is tended to lessen when the particle size was reduced which caused better mobility and slip of the CR particles (Sreeja and Kutty, 2000).

CONCLUSION

The present study was designed to determine the effects of crumb rubber produced by ambient grinding methods on the properties of tyre tread blend. The study comprised of the incorporation of CR as a filler in passenger tyre tread blends for different particle sizes and content. The properties studied were mechanical and process characteristics. The results showed that there were clear enhancements in the mechanical properties as the CR particle size reduced. On the other hand, there was deterioration in these properties with an increase in the CR loading. The experiments confirmed that the CR particle size reduction positively affected the properties through a reduction in viscosity and curing time with an increase in maximum torque and negatively by a reduction in the scorch time which meant less safety in the scorch process. Furthermore, an increase in the CR content produced undesirable process characteristics such as reduced maximum torque and scorch time with the increase of viscosity and curing time. In conclusion, an optimization study based on CR particle size reduction energy cost, with virgin rubber cost could be undertaken to achieve better utilization of the CR content and particle size which could be convenient to use in the manufacturing of the tyre tread layer.

ACKNOWLEDGEMENT

The researchers are grateful to the State Company for Tyre Industry, Laboratory and Technology Department, Iraq, for providing full support to carry out this research.

REFERENCES

ASTM International, 2001a. Standard practice for rubber materials equipment and procedures for mixing standard compounds and preparing standard vulcanized sheets. ASTM International Voluntary Organization, Conshohocken, Pennsylvania.

ASTM International, 2001b. Standard test method for rubber property abrasion resistance rotary drum abrader. ASTM International Voluntary Organization, Conshohocken, Pennsylvania.

ASTM International, 2003. Standard test methods for vulcanized rubber and thermoplastic elastomers tension. ASTM International Voluntary Organization, Conshohocken, Pennsylvania.

Amari, T., N.J. Themelis and I.K. Wernick, 1999. Resource recovery from used rubber tires. Resour. Policy, 25: 179-188.

Beukering, P.J.V. and M.A. Janssen, 2001. Trade and recycling of used tyres in Western and Eastern Europe. Resour. Conserv. Recycl., 33: 235-265.

Colom, X., F. Carrillo, C. Pilar and C. Xavier, 2011. New Routes to Recycle Scrap Tyres. Intech Open Access Publisher, USA.,.

Colom, X., J. Canavate, F. Carrillo and J.J. Sunol, 2009. Effect of the particle size and acid pretreatments on compatibility and properties of recycled HDPE plastic bottles filled with ground tyre powder. J. Appl. Polym. Sci., 112: 1882-1890.

Fan, P. and C. Lu, 2012. Grafting of hyperbranched poly (amidoamine) onto waste tire rubber powder and its potential application as the curing agent for epoxy resin. Polym. Adv. Technol., 23: 48-56.

Gibala, D. and G.R. Hamed, 1994. Cure and mechanical behavior of rubber compounds containing ground vulcanizates. Rubber Chem. Technol., 67: 636-648.

Han, S.C. and M.H. Han, 2002. Fracture behavior of NR and SBR vulcanizates filled with ground rubber having uniform particle size. J. Appl. Polym. Sci., 85: 2491-2500.

Klingensmith, W. and K. Baranwal, 1998. Recycling of rubber: An overview. Rubber World, 218: 41-46.

Lamminmaki, J., S. Li and K. Hanhi, 2006. Feasible incorporation of devulcanized rubber waste in virgin natural rubber. J. Mater. Sci., 41: 8301-8307.

Li, S., J. Lamminmaki and K. Hanhi, 2004. Effect of ground rubber powder on properties of natural rubber. Macromol. Symp., 216: 209-216.

Li, Y., S. Zhao and Y. Wang, 2012. Microbial desulfurization of ground tire rubber by Sphingomonas sp: A novel technology for crumb rubber composites. J. Polym. Environ., 20: 372-380.

Luo, T. and A.I. Isayev, 1998. Rubber-plastic blends based on devulcanized ground tire rubber. J. Elastomers Plast., 30: 133-160.

Mangaraj, D., 2004. Rubber Recycling by Blending with Plastics. In: Rubber Recycling, Sadhan, K.D., I.I. Avraam and K. Khait (Eds.). Taylor & Francis, New York, USA., pp: 247-298.

Meysami, S.Z.M. and C. Tzoganakis, 2010. Effect of Incorporation of Devulcanized Rubber. Antec Publisher, New York, USA.

Mull, M.A., K. Stuart and A. Yehia, 2002. Fracture resistance characterization of chemically modified crumb rubber asphalt pavement. J. Mater. Sci., 37: 557-566.

Ramarad, S., M. Khalid, C.T. Ratnam, A.L. Chuah and W. Rashmi, 2015. Waste tire rubber in polymer blends: A review on the evolution, properties and future. Progress Mater. Sci., 72: 100-140.

Sreeja, T.D. and S.K.N. Kutty, 2000. Cure characteristics and mechanical properties of natural rubber-reclaimed rubber blends. Polym. Plast. Technol. Eng., 39: 501-512.

Sreeja, T.D. and S.K.N. Kutty, 2002. Studies on acrylonitrile butadiene rubber-reclaimed rubber blends. J. Elastomers Plast., 34: 145-155.

Sreeja, T.D. and S.K.N. Kutty, 2003. Styrene butadiene rubber-reclaimed rubber blends. Int. J. Polym. Mater., 52: 599-609.

Wang, H.Z., X.U. He and X.J. Xuan, 2010. Review of waste tire reuse and recycling in China-current situation, problems and countermeasures. Adv. Natural Sci., 2: 31-39.

Yehia, A.A., M.A. Mull, M.N. Ismail, Y.A. Hefny and A.E.M. Bary, 2004a. Effect of chemically modified waste rubber powder as a filler in natural rubber vulcanizates. J. Appl. Polym. Sci., 93: 30-36.

Yehia, A.A., M.N. Ismail, Y.A. Hefny, A.E.M. Bary and M.A. Mull, 2004b. Mechano-chemical reclamationof waste rubber powder andits effect on the performanceof NR and SBR Vulcanizates. J. Elastomers Plast., 36: 109-123.

Zhang, S.L., Z.X. Zhang and J.K. Kim, 2011. Study on Thermoplastic Elastomers (TPEs) of waste polypropylene-waste ground rubber tire powder. J. Macromol. Sci. Phys., 50: 762-768.