

PAPER • OPEN ACCESS

## Study and comparison on mechanical properties of various polymers reinforced with ground tires rubber (GTR)

To cite this article: M Marín-Genescà *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **542** 012027

View the [article online](#) for updates and enhancements.



**IOP | ebooks™**

Bringing you innovative digital publishing with leading voices to create your essential collection of books in STEM research.

Start exploring the collection - download the first chapter of every title for free.

# Study and comparison on mechanical properties of various polymers reinforced with ground tires rubber (GTR)

M Marín-Genescà<sup>1</sup>, J García-Amorós<sup>2</sup>, J Bordes-Arroyo; R Mujal-Rosas<sup>1</sup>, LI Massagués Vidal<sup>2</sup>

<sup>1</sup>Mechanical Engineering Department, ETSEQ-URV, Països Catalans, 26, Tarragona – 43007 Spain

<sup>2</sup>Electrical Engineering Department, ETSE-URV, Països Catalans, 26, Tarragona – 43007 Spain

<sup>3</sup>Electrical Engineering Department, EET-UPC, Colom, 1 Terrassa - 08222 Spain

E-mail: [marc.marin@urv.cat](mailto:marc.marin@urv.cat)

**Abstract.** The recycling of worn tires is a big challenge nowadays and it is necessary for environmental and economic reasons: mass production of tires, as well as the difficult storage or elimination is a real environmental problem. Various methods for recycling tires are currently used, such as mechanical grinding, which puts vulcanized rubber, steel and fibers apart. The rubber may be used in several industrial applications as flooring, insulations, footwear, etc. The aim of this paper focuses on finding a new application for the old used tires (GTR). Tires dust and 7 different polymers have been mixed, and we have checked the maximum accepted values of GTR that can be admitted while keeping mechanical properties within acceptable values, as well as initial polymer microstructure. This would allow including GTR in industrial applications of recycled polymers. The recycled tire dust which results from the industrial milling processes has been divided by categories according to the size of the particles. This has been mixed with different polymers, in different GTR concentrations in order to establish its conduct through mechanical test. With the present study it is intended to give a second outlet to the tires out of use, demonstrating the feasibility of the mechanical properties analyzed.

## 1. Introduction

The important issue of the accumulation of used tires [1-3] has driven the efforts of the international scientific community to seek solutions for recovery and reuse. Many plastic materials include elastomers to improve its toughness. In general, a thermoplastic or thermosetting polymer acts as a matrix and the elastomer acts as a dispersed phase [4-6]. Moreover, as in other two-phase polymer blends [7-8], the interfacial compatibility between the components is important for achieving the desired properties. In the case of recycled elastomers, expected compatibility is low. One way to increase the compatibility between components is to reduce the degree of cross-linked GTR by devulcanization [9-11]. Significant changes in properties are observed when we change the size of the reinforcement particles [12]. The size of particles is restricted because of the pulverization technical procedures, so we have chosen a simple and cheap way to obtain the classification in the three desired particle sizes (<200 $\mu\text{m}$ , 200 $\mu\text{m}$ -500 $\mu\text{m}$  and >500 $\mu\text{m}$ ) such as screening, although this method has the disadvantage of using only a part of the initial amount of GTR. It is, therefore, to determine what percentage of GTR can be added to different polymers matrix (PVC, EVA, HDPE, PP, PA, ABS and PS), keeping mechanical properties, as well as the polymer initial microstructure [13-14] within an acceptable range of mechanical values. This would allow to add GTR to various industrial processes. To this end, we have analyzed various concentrations of polymer/GTR (from 0% to 70% of GTR), with the three indicated particle sizes. Finally, we will give some applications that could have these compounds analyzed, there will be a valuation of the residual GTR for applications that may be interesting, and thus give out a waste like the GTR, which today represents a very important problem in developed societies.

## 2. Methodology

### 2.1 Materials

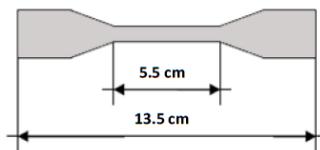
The polyvinyl chloride (PVC) with a fluidity index of 1.35 g/min and a density of 1.225 kg/m<sup>3</sup>. High density polyethylene (HDPE) recycled, with a melt flow index of 1.35 g/min and density of 960 kg/m<sup>3</sup> was used in this study. The EVA (Etylene Vinyl Acetate) copolymer, with 18% of vinyl acetate and 82% ethylene, melt flow rate of 0.2 g/min and a density of 937 kg/m<sup>3</sup>. Polypropylene (PP) with a melting



temperature of 165 °C, a flow rate of 0.55 g/min, and a density of 902 kg/m<sup>3</sup>. ABS it is made up of 30% acrylonitrile, 20% butadiene, and 50% styrene, its melting temperature is 230 °C and its density is 1050 kg/m<sup>3</sup>. Polyamide 6 (PA), also known as Nylon 6, its melting temperature is 220 °C, its melt flow rate is 1.55 g/min and its density is 1130 kg/m<sup>3</sup>. Polystyrene, PS, (styrene-butadiene-styrene), its melting temperature is 180°C, its melting index is 1.45 g/min and its density is 1050 kg / m<sup>3</sup>. On the other hand the old used tire (GTR), with a particle size less than 700 μm, was verified by TGA analysis that carbon black content was about 35%. The original GTR was separated by sieving into three categories of size particles: <200 μm, 200-500 μm and >500 μm.

## 2.2 Preparation of the compound and mechanical analysis

The recycled tire powder was dried in an oven at 100°C for 24h. Five samples of Polymer/GTR compound, varying the composition (5%, 10%, 20%, 40%, 50% and 70% of GTR), were prepared for each particle size. The mixing process was done with a *Brabender* mixer machine, at different temperatures. Polymer/GTR laminates were obtained by using a hot plates press at constant pressure 200 bar and different temperatures for 10 min depending on the polymer to be treated. Samples for testing were properly set up according to the specifications of ASTM-D-638 type V. A sample of the pure polymer was also prepared with the same requirements in order to obtain comparable results. Stress–strain tests were carried out with an *Instron 3366–10kN* universal machine, as the specifications of ASTM-D-638 type V. The test speed was 20 mm/min. The test temperature was 23±2 °C, and the relative humidity was of 50±5%. The study of mechanical properties, according to the GTR concentration in the matrix and the particle sizes include the Young's modulus, tensile stress, elongation at break and toughness. Five specimens for testing (Fig 1) were used.

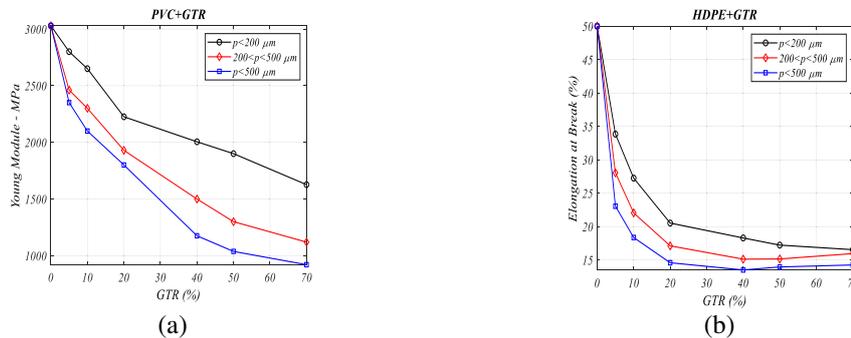


**Figure 1.** Obtained samples for testing.

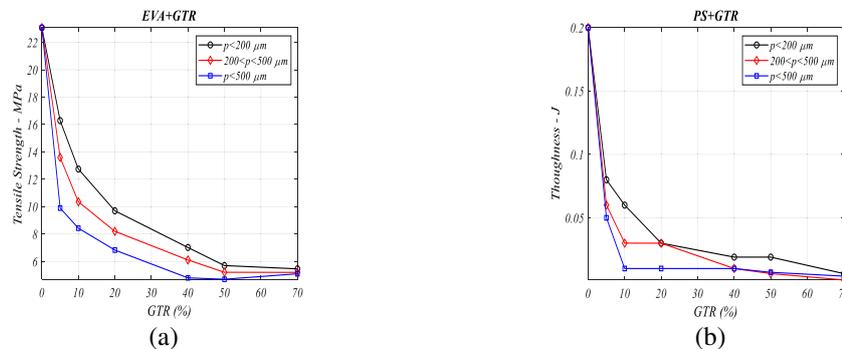
## 3. Results and discussion

### 3.1 Mechanical Properties

Figure 2 and 3 shows the results by testing the stress-strain mechanical properties of different polymers analyzed. These properties were analyzed with different GTR concentrations and the three categories of sizes of reinforcing particles in the Polymer matrix [15-19]. Figure 2-a shows the Young's modulus of the composite, the rigidity for small GTR concentrations (>10%) decreases significantly regarding the original PVC, from a value of 2800 MPa (5% GTR) to 1627 MPa (70% GTR) for small particle sizes (<200 μm). We can also see how rigidity decreases with the other particle sizes, from original Young's modulus value of 2461 MPa (5% GTR) to 1120 MPa (70% GTR) for particle size of 200-500μm, and to 921 MPa for particle size of >500μm for the same GTR concentration. This is due to the fact that large particles have higher probability of cracks, and they also crowd during the melting process. When the content of GTR continues to increase, interfacial adhesion is weakened, and it causes a diminution of stiffness in all the cases. Thus, for concentrations of 40% and 50% GTR, values are 2005 MPa and 1900 MPa (28.4% and 32.14% lower than those of the EVA/GTR-5% compound for <200μm particles), reaching slightly higher values than those of pure PVC for 70% GTR concentrations, no matter the size of the particles is (1627MPa, 1120MPa and 921MPa, for the three particle sizes 200μm, 200-500μm and >500μm, respectively). In the HDPE composites with GTR, can check that the decrease is so high for the 5% GTR compounds (figure 2-b), for HDPE/GTR-5%, we have this values 33.84% for 200μm size of GTR particles, 28.04% for 200-500μm of size, and 23.05% >500μm, this probes that the decrease is 18% and 32% respectively in function of the particle size, is really proved that the particle size is a very important fact for the mechanical behavior [20-21].



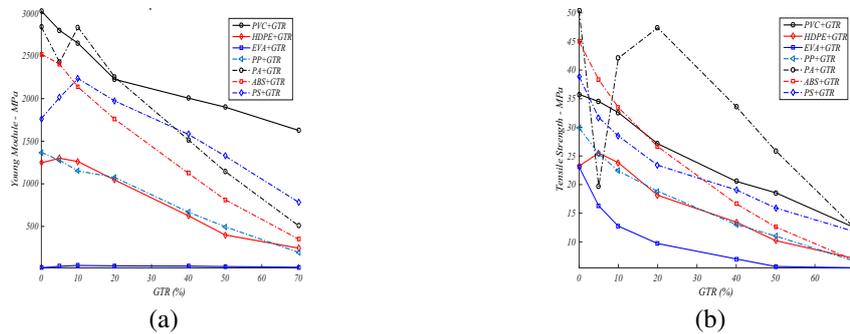
**Figure 2.** a) Young's modulus (MPa) for different concentrations of PVC/GTR and particles sizes. b) Elongation at break (%) for different concentrations of HDPE/GTR and particles sizes.



**Figure 3.** a) Tensile Stress (MPa) for different concentrations of EVA/GTR and particles sizes. b) Toughness (J) for different concentrations of PS/GTR and particles sizes.

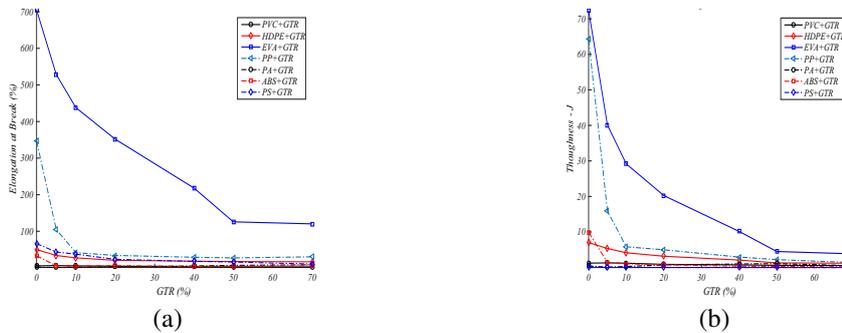
Figure 3-a shows the tensile stress, which differs from the stiffness, showing a decrease since low GTR concentrations. Thus, for 5-10% GTR concentrations, the decrease of tensile stress compared to pure EVA are quite significant, from 23MPa to 16.2MPa and to 12.7MPa (28% and 44% respectively for particles size <200μm). These decreases are more significant for larger particle sizes, reaching 13.6MPa (41%) and 10.4MPa (54%) respectively for particles sizes between 200μm-500μm, and 9.9MPa (57%) and 8.4MPa (63%) respectively for >500μm particles. From 20% GTR, values fall sharply regardless of particle size or GTR concentration in the matrix. Values are 3.5 - 4 times lower than the original EVA. It shows poor compatibility between components when the percentage of reinforcement is increased. We can see similar trends in the figure 3b) for a PS composites analyzed. The optimum particle size is the p<200μm. The addition of GTR decrease the mechanical properties analyzed [22].

As we can see in the figures 2 a) and b) and 3 a) and b) analyzed, can assure that the size particle of p<200μm is the optimum particle size analyze. With this condition are analyzed the 7 polymers mixed with GTR generated in the next figures 4 a) and b) and 5 a) and b). The addition of old used tires particles in all compounds produces a decrease in the four mechanical properties analyzed in general like young modulus (Fig 4-a) and tensile strength (Fig 4-b) elongation at break (Fig. 5-a) and toughness (Fig. 5-b).



**Figure 4.** a) Young Module (MPa), b) Tensile Strength (MPa), for seven polymers/GTR concentrations and particle size <200µm.

For the Young Module analysis, we can see how the PVC and EVA compounds has better behavior with the addition of the GTR percent in the composite, and in the Tensile Strength analyzed we can see how the PA and PVC compounds has better behaviors with the addition of quantities of GTR (near 70%).



**Figure 5.** a) Elongation at Break (%), b) Toughness (J), for seven polymers/GTR concentrations and particle size <200µm.

For particle sizes <200µm elongation at break of the GTR compounds [23], the optimum behavior is for EVA compounds, that goes from 704% to 528%, 437% and 351%, which means decreases of 29%, 43% and 50% for GTR concentrations of 5%, 10% and 20%. From 40% concentration, the values are always lower than 3.5 times those of pure EVA. The reduction of deformation capacity of the elastomer influences the decrease in elongation and, subsequently, the decrease of hardness, and we have similar behaviors of the Toughness property for the composites with EVA/GTR. Regarding toughness, these drops are even greater. Actually, regarding the compound with the smallest particle size (<200µm), the toughness goes from 72.3J for pure EVA to 40J or 29.2J for 5% and 10% GTR concentrations. It will decrease even more for larger particle sizes. For concentrations above 20%, considerations on the particles sizes or the percentage of GTR are not important at all, since even 10J of energy are not reached in any case, and it means that these values are seven times lower than the original EVA. The decrease in elongation at break is related to the imperfect interfacial adhesion between components. The rest of polymers are so low values for the elongation at break and toughness properties. The incidence of poor adhesion between phases is a particularly important result [23].

**4. Conclusions**

Firstly, it can be clearly seen how the particle size decisively influences the properties analyzed, it is observed as GTR particle of lower sizes (<200µm) have better mechanical behavior in the figures 2 a) and b) and 3 a) and b), so, the mechanical properties analyzed have a better behavior with a GTR minimum particle size (<200µm), this is due to the imperfect interfacial adhesion between components is better for a low size of particle than a highest size of particle (p>500µm). Stress-strain tests show how, by adding the GTR in the matrix up to 10%, and especially for small particle sizes, the Young's modulus of the compound increases, although other mechanical properties decrease. With the EVA,

PVC and HDPE composites. This behavior may be due to the fact that reinforcement-matrix is correct for these formulations, and therefore, some mechanical properties such as stiffness improve. However, for GTR concentrations above 10%, with any particles size, all mechanical properties decrease, and the larger the size of the particle is, the more significant the decrease is. The results obtained from the analysis of these compounds, only obtained from recycled polymers, show that that at 10-20% GTR concentration is the limit concentration value for keeping acceptable values of mechanical properties of the compound, considering exclusively the GTR particle size as a variable, and without any previous pre-treatment considered. Other methods such as prior devulcanization of GTR should be tested, to see if values could be increased up to 20-25% of GTR. This would allow its use in various fields of industry, as for example in pipes for waste in construction works and conduits for electrical cables, as well as for waste deposits in construction works, through these applications we could introduce recycled GTR in industrial applications, and so we could give also an exits to quantities of GTR in disuse that nowadays represent a problem both its storage and its recycling.

## 5. References

- [1] European Tyre Recycling Association (ETRA). Available at: [www.etra-eu.org](http://www.etra-eu.org).
- [2] Used Tyre Working Group (UTWG). *Tyre Recycling*; Department of Trade and Industry: London, UK. Available at: [www.tyredisposal.co.uk](http://www.tyredisposal.co.uk), November, 2006.
- [3] Liu HS, Richard CP, Mead JL and Stacer RG. *Development of Novel Applications for Using Recycled Rubber in Thermoplastics*; Technical Research Program, Chelsea Center for Recycling and Economic Development, University of Massachusetts: Lowell, 2000.
- [4] Figovslq O, Beilin D, Blank N, Potapo J and Chernyshe V. *Development of polymer concrete with polybutadiene matrix. Cem Concr Compos* 1996, 18, 437-444.
- [5] Hernandez-Olivares F, Barluenga G, Bollatib M and Witoszekc B. *Static and dynamic behaviour of recycled tyre rubber-filled concrete. Cem Concr Compos* 2002, 32, 1587-96.
- [6] Goncharuk GP, Knunyants MI, Kryuchkov AN and Obolonkova ES. *Effect of the specific surface area and the shape of rubber crumb on the mechanical properties of rubber-filled plastics. J Polym Sci Part B: Polym Chem* 1998, 40, 166-169.
- [7] Dierkes WK. *Rubber recycling*. In Recent research developments in macromolecules, Pandalai SG, Ed.; Trivandrum: Research Signpost, 2003, vol. 7, pp. 265–292.
- [8] Radeshkumar C and Karger-Kocsis J. *Thermoplastic dynamic vulcanisates containing LDPE, rubber, and thermochemically reclaimed ground tyre rubber. Plast Rubber Compos* 2002, 31,99-105.
- [9] Yehia A, Mull MA, Ismail MN, Hefny YA and Abdel-Bary EM. *Effect of chemically modified waste rubber powder as a filler in natural rubber vulcanizates. J Appl Polym Sci* 2004, 93, 30-36.
- [10] Colom X, Andreu-Mateu F, Cañavate FJ, Mujal R and Carrillo F. *Study of the influence of IPPD on thermo-oxidation process of elastomeric hose. J Appl Polym Sci* 2009, 5, 2011-2018.
- [11] Cepeda-Jimenez CM, Pastor-Blas MM, Ferrándiz-Gómez TP and Martín-Martínez JM. *Surface Characterization of vulcanized Rubber treated with sulphuric acid and its adhesion to polyurethane adhesive. J Adhesion* 2000, 73, 135-160.
- [12] Mujal R, Marín-Genescà M, Orrit J, Rahhali A, Colom X, *Dielectric, mechanical, and thermal characterization of high-density polyethylene composites with ground tire rubber. Journal of thermoplastic composite materials*, 2012, vol. 25, núm. 5, p. 537-559
- [13] Nakason C, Kaesaman A and Supasanthitkul P. *The grafting of maleic anhydride onto natural rubber. Polym Test* 2004, 23, 35-41.
- [14] La Mantia FP, Lo Verso S and Tzankova Dintcheva N. *EVA Copolymer Based Nanocomposites. Macro Mat Eng* 2002, 287, 12, 909-914.
- [15] Kim JI, Ryu SH and Chang YW. *Mechanical and dynamic mechanical properties of waste rubber powder/HDPE composite. J Appl Polym Sci* 2000, 77, 2595-2602.
- [16] Colom X, Cañavate J, Carrillo F, Velasco JI, Pages P, Mujal R and Nogues F. *Structural and mechanical studies on modified reused tyres composites. Eur Polym J* 2006, 42, 2369-2378.
- [17] Ganesh B and Unnikrishnan GJ. *Cure characteristics, morphology, mechanical properties, and aging characteristics of silicone rubber/ethylene vinyl acetate blends. Appl Polym Sci* 2006, 99, 3, 1069-1082.
- [18] Tang LW, Tam KC, Yue CY, Hu X, Lam YC and Li L. *Rheological and mechanical properties of compatibilized polystyrene/ethylene vinyl acetate blends. J Appl Polym Sci* 2004, 94, 5, 2071-2082.

- [19] Shih-Kai C, Po-Tsun C, Cheng-Chien W and Chuh-Yung C. *Mechanical properties of ethylene-vinyl acetate/ polystyrene blends studied by in situ polymerization*. Journal of Applied Polymer Science. *J Appl Polym Sci* 2003, 88, 3, 699-705.
- [20] Mujal R, Orrit-Prat J, Ramis-Juan X, Marin-Genesca M, Ahmed Rahhali. *Dielectric, Thermal, and Mechanical Properties of Acrylonitrile Butadiene Styrene Reinforced with Used Tires*. Advances in Polymer Technology 32:E399-E415. Wiley 2013. 0730-6679
- [21] Mujal R, Orrit-Prat J, Ramis-Juan X, Marin-Genesca M. *Electrical application of polyamide reinforced with old tire rubber (ground tire rubber): Dielectric, thermal, mechanical and structural properties*. Journal of thermoplastic composite materials. 27(9):1209-1231. 2014. 0892-7057
- [22] El-Nashar DE, Eid MAM, Abou Aiad TH and Abd-El-Messieh SL. *Electrical and Mechanical Investigations on Polyvinyl Chloride Filled with HAF Black*. *J Reinforc Plast Compos* 2009, 28, 14, 1763-1773.
- [23] Saad ALG, Aziz HA and Dimitry OIH. *Studies of Electrical and Mechanical Properties of Poly(vinyl chloride) Mixed with Electrically Conductive Additives*. *J Appl Polym Sci* 2004, 91, 1590-1598.

#### **Acknowledgements**

This work was funded by the Ministerio de Economía y Competitividad de España ENE2015-64117-C5-3-R (MINECO/FEDER).