Giving a second opportunity to tire waste: an alternative path for the development of sustainable self-healing styrene-butadiene rubber compounds overcoming the magic triangle of tires

Javier Araujo-Morera; Marianella Hernández Santana*; Raquel Verdejo; Miguel Angel López-Manchado

Institute of Polymer Science and Technology (ICTP-CSIC), Juan de la Cierva 3, Madrid 28006, Spain. jaraujo@ictp.csic.es (J.A.M.); marherna@ictp.csic.es (M.H.S.); rverdejo@ictp.csic.es (R.V.); lmanchado@ictp.csic.es (M.L.M.)

* Correspondence: marherna@ictp.csic.es (M. H. S.)

S1. Cryo-grinding protocol



Figure S1-1. Schematic representation of grinding and cooling cycles.

S2. Tensile curves of virgin and repaired SBR/GTR compounds



Figure S2-1. Stress-strain curves of the SBR/GTR compounds in virgin (V) and repaired (R) state.

| Table S2-1. Mechanical | properties | of pristine | and repaired | SBR compounds. |
|------------------------|------------|-------------|--------------|----------------|
|------------------------|------------|-------------|--------------|----------------|

| | Compound | | | | | |
|---|-----------------|-----------------|-------------------|-------------------|-------------------|--|
| | SBR | GTR | SBR/10GTR | SBR/20GTR | SBR/30GTR | |
| PRISTINE | | | | | | |
| Tensile stress at 50% strain, σ_{50} (MPa) | 0.54 ± 0.02 | 1.27 ± 0.05 | $0.65 {\pm} 0.02$ | 0.66 ± 0.02 | 0.59 ± 0.04 | |
| Tensile stress at 100% strain, σ_{100} (MPa) | 0.65 ± 0.02 | 2.25 ± 0.05 | $0.83 {\pm} 0.03$ | 0.86 ± 0.03 | 0.77 ± 0.04 | |
| Tensile stress at 300% strain, σ_{300} (MPa) | 0.75 ± 0.02 | - | 1.42 ± 0.06 | 1.64 ± 0.07 | 1.58 ± 0.03 | |
| Tensile stress at 500% strain, σ_{500} (MPa) | 0.88 ± 0.02 | - | $2.40{\pm}0.10$ | 2.79 ± 0.09 | 2.61 ± 0.07 | |
| Tensile strength, σ_b (MPa) | 1.33 ± 0.08 | 4.8 ± 0.2 | $2.60{\pm}0.20$ | 2.90 ± 0.10 | 3.30 ± 0.10 | |
| Elongation at break, ε_b (%) | 846±34 | 198±9 | 550±24 | 546±18 | 639±24 | |
| Crosslink density, v x 10^{-5} (mol/g) | 1.46 ± 0.02 | 30.1±0.3 | 4.83±0.06 | 4.35±0.08 | 3.24 ± 0.04 | |
| REPAIRED | | | | | | |
| Tensile stress at 50% strain, σ_{50} (MPa) | $0.54{\pm}0.04$ | - | $0.68 {\pm} 0.02$ | 0.65 ± 0.03 | 0.66 ± 0.01 | |
| Tensile stress at 100% strain, σ_{100} (MPa) | 0.67 ± 0.05 | - | $0.86{\pm}0.03$ | 0.85 ± 0.03 | 0.83 ± 0.01 | |
| Tensile stress at 300% strain, σ_{300} (MPa) | - | - | - | - | - | |
| Tensile stress at 500% strain, σ_{500} (MPa) | - | - | - | - | - | |
| Tensile strength, σ_b (MPa) | 0.75 ± 0.05 | - | $0.80{\pm}0.10$ | $0.97 {\pm} 0.08$ | $0.91 {\pm} 0.02$ | |
| Elongation at break, ε_b (%) | 177±14 | - | 98±9 | 180±55 | 228±66 | |
| Healing efficiency, η (%) | 56±7 | _ | 31±5 | 33±3 | 28±1 | |



Figure S2-2. The S2p core spectrum of as received GTR and cryo grounded GTR.



S3. Fracture surface of SBR/GTR compounds.

Figure S3-1. Scanning electron microscope (SEM) images of fracture surface of SBR/GTR compounds.



S4. Dielectric properties of SBR/GTR compounds.

Figure S4-1. Dielectric loss (ε ") as a function of the frequency of: a) SBR/20GTR; b) SBR/30GTR, in the temperature range from -45 to -5 °C.



Figure S4-2. Electrical conductivity (σ') as a function of frequency of SBR/GTR compounds at 25 °C.