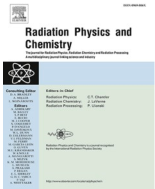
Radiation Physics and Chemistry 201 (2022) 110414

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem



Synergistic effects of gamma pre-irradiation and additional vulcanizing agent in case of ground tire rubber containing vulcanizates

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ARTICLE INFO

Keywords:
Gamma irradiation
Recycling
Ground tire rubber
Elastomers

ABSTRACT

In this study, we investigated gamma radiation treated ground tire rubber (GTR) at different doses and measured the soluble content and the cross-link density of the samples. These results have been evaluated with Horikx's diagram. The results showed that at low doses, the selective chain-scission of the sulphur cross-links is dominant; however, at higher doses, the degradation of the polymer backbone comes to the fore. Natural rubber (NR) based mixtures were prepared in an internal mixer using conventional and two-step mixing, then the samples were vulcanized. Synergistic effects were observed when using radiation treatment and two-step mixing, resulting in improved mechanical properties. In materials where only one treatment was applied, the properties did not change significantly. The morphology of the mixtures was observed using scanning electron micrographs, which showed that the GTR could integrate well into the matrix.

1. Introduction

Nowadays, rubber waste is an important issue, as it is generated in enormous quantities and can cause serious environmental problems. A lot of harmful substances can dissolve from them and they can easily catch fire, which is almost impossible to control and can cause serious respiratory health concerns. The most significant source of rubber waste is end-of-life tires, which cannot be recycled easily due to their cross-linked structure. These tires are often ground (ground tire rubber, GTR) and then mixed with other polymeric materials. One of the biggest problems is that the GTR and the matrices are often incompatible, so additional modifying methods are required to increase the compatibility; thus, the produced materials can meet the requirements of engineering applications (Karger-Kocsis et al., 2013; Paleri et al., 2021; Simon and Bárány, 2021).

In order to improve adhesion, several methods have already been used. Most commonly grafting reactions are used on the GTR or even on the matrix, thus bringing the properties of the two phases closer together. The particle size (Kiss et al., 2022) also plays a significant role in establishing a good connection between the phases, smaller GTR particles results in better mechanical properties. Another possibility is adding reactive compatibilizers to the system, which may form covalent bonds at the interface. Often it is enough to oxidize the surface of the GTR with reactive gases, acids, or gamma radiation in the air atmosphere (Colom et al., 2007; Karger-Kocsis et al., 2013; Naskar et al., 2002; Ramarad et al., 2015; Tolstov et al., 2007).

Ionizing radiation is an efficient way to improve the compatibility of different polymers. Good results have been obtained with this method, and mostly thermoplastic matrix has been used (Mészáros et al., 2012; Sonnier et al., 2006). In most GTR-containing compounds, the final product is irradiated, resulting in a cross-linked structure. Although the properties of the resulting materials are favorable, recycling the product is difficult. Nevertheless, it would be useful to use ionizing radiation treatment before the mixing step (Ratnam et al., 2013). In this case, only the GTR needs to be treated, which means less volume to be irradiated. Not only the economic significance of it is important, but this way, the produced raw material can be used for different processing technologies and products.

Several reactions can occur during the interaction of ionizing radiation and polymers, such as cross-linking, chain scission, oxidation, and grafting. The dominant reaction depends on many parameters, such as the structure of the polymer, the conditions (atmosphere, temperature), the dose rate, and the amount of dose absorbed (Keizo and Song, 2012).

Based on some studies (Şen et al., 2003; Sonnier et al., 2007), it can

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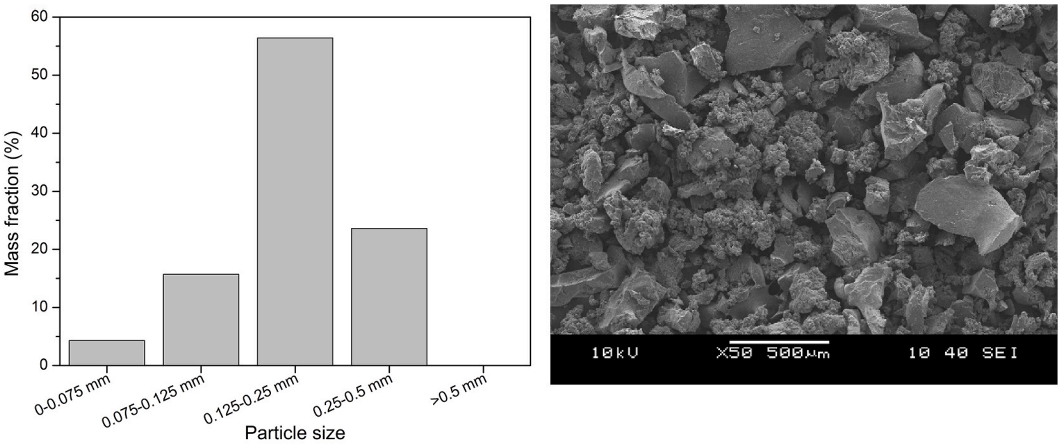
https://doi.org/10.1016/j.radphyschem.2022.110414
Received 17 May 2022; Received in revised form 8 July 2022; Accepted 12 July 2022



a) b)

Fig. 1. The particle size distribution a) and the morphology b) of the used GTR.

be seen that at low doses (<100 kGy), oxygen-containing groups appear on the surface of the rubber when irradiated in air atmosphere. The formed functional groups make the surface more hydrophilic, which may promote better compatibility in the case of polar matrices. Among these, the various peroxides are particularly important. They can decompose during processing, thus creating radicals that can attack the double bonds of the rubber and, as a final result, form covalent bonds between the phases that greatly improve compatibility (Cataldo et al., 2010).

Although the use of GTR in thermoplastic matrices has been more commonly researched, its use in virgin rubbers is a promising approach for tire recycling (low cost, negligible environmental impact). Yehia et al. (2004) studied the effect of chemically modified GTR in natural rubber (NR) vulcanizates, where they used nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) to activate the surface of the GTR. They recorded the infrared spectrum of the materials, and bands associated with carbonyl groups were found in the case of GTR treated with oxidants. They also found that the treatment helped to improve the tensile strength of the vulcanizates.

To compensate for life-cycle and radiation-induced degradation, adding an additional vulcanizing agent to the system is advisable, which is most effective when mixed directly with the GTR and then placed in the fresh, unvulcanized rubber. This technique is called two-step mixing. This method was developed by Simon et al. (2020b). They achieved improved mechanical properties in the case of NR/GTR vulcanizates. Gopi Sathi et al. (Gopi Sathi et al., 2021) also highlighted the importance of the vulcanization system on the final properties of rubber compounds.

A combination of gamma radiation and two-step mixing can be beneficial. The former can create active sites on the surface, which can form strong covalent bonds between the two phases. The latter can compensate for the degradation that occurred during the tires' lifetime and by irradiation and also may help the active sites to form the cross-links. The combined effect of these methods has not yet been studied but seems to be promising.

In this study, we investigate the effects of gamma radiation, two-step mixing, and their combination in NR-based vulcanizates. In order to determine which parameters are the most suitable for the radiation treatment, GTRs with different absorbed doses and their use in NR vulcanizates were investigated. To evaluate the treatments' effectiveness and explore the synergistic effects between the two methods, the GTR containing vulcanizates were subjected to various mechanical and morphological tests.

2. Experimental

Waterjet-milled ground tire rubber from the tread of truck tires was kindly provided by Aquajet Ltd. (Budapest, Hungary). The particle-size distribution of the used GTR is shown in Fig. 1a. We determined the particle size distribution of the GTR using a BA 200N type sieve shaker (CISA Cedaceria Industrial, Barcelona, Spain) with 75, 125, 250 and 500 μm sieves. A scanning electron micrograph of the used GTR is presented in Fig. 1b., where the complex morphology of the particles can be seen.

Based on former research (Simon et al., 2020a), the composition of the GTR was the following: 50–55 phr of NR, 45–50 phr of synthetic rubber, 33–37 phr of carbon black, 7.5 phr of residual additives, and 4–6 phr of oil (the results were determined using thermogravimetric analysis).

For the irradiation of the GTR, a panoramic SLL-01 type ^{60}Co radiation source at Institute of Isotopes Ltd. was used. Irradiation was carried out at room temperature and in an air atmosphere. The dose rate was 2 kGy/h, and the room's air was changed six times an hour to always have enough oxygen to create functional groups on the surface. The examined doses based on literature data were: 20, 40, 60, 80, 100, 250, 500, 750, and 1000 kGy. The treated GTR samples are abbreviated as GTR_IR*, where the * stands for the absorbed dose; for instance, the 100 kGy sample is denoted as GTR_IR100.

We measured the sol fractions of the GTRs with Soxhlet extraction according to Eq. (1). Toluene was used as a solvent; the extraction lasted for 16 h, after which the samples were dried to constant weight. We weighed the samples twice: before the extraction and after drying.

$$Sol\,fraction\ (\%) = \left(1 - \frac{\mathrm{m_f}}{\mathrm{m_i}}\right)\cdot 100, \tag{1}$$

where m_i and m_f stand for the mass of GTRs before and after the extraction.

We determined the cross-link density of the GTRs via swelling tests according to ASTM D 6814-02, using the Flory-Rehner Eq. (2) (Flory and Rehner, 1943). The swelling was carried out in toluene for 72 h, and then the samples were dried at 80 °C to constant weight.

$$\nu_e = \frac{-\left[\ln(1 - V_r) + V_r + \chi_1\cdot V_r^{2}\right]}{\left[V_1\cdot\left(V_r^{\frac{1}{3}} - V_r\right)\Big/ 2\right]}, \tag{2}$$

where ν_e is the cross-link density in mol/cm^3; V_1 is the molar volume of the solvent (in the case of toluene: 106.13 cm^3/mol); χ_1 is the rubber-solvent interaction parameter (0.39), and V_r is the volume fraction of

Table 1
Additives used in the rubber mixtures.

Material	Manufacturer	Trade name
Stearic acid	Oleon (Ertvelde, Belgium)	Radia0444
Zinc oxide (ZnO)	Werco Metal (Zlatna, Romania)	ZnO WZ-1
Paraffin oil	Hansen und Rosenthal (Hamburg, Germany)	Tudalen 3036
N-cyclohexyl-2-benzothiazole sulfonamide (CBS)	Rhein Chemie (Mannaheim, Germany)	Rhenogran® CBS-80
Tetramethyl thiuram disulfde (TMTD)	Lanxess (Cologne, Germany)	Rhenogran® TMTD-70
N772 carbon black	Omsk Carbon Group (Omsk, Russia)	N-772 OMSK
Sulphur	Ningbo Actmix Polymer (Ningbo, China)	Curekind Sulphur

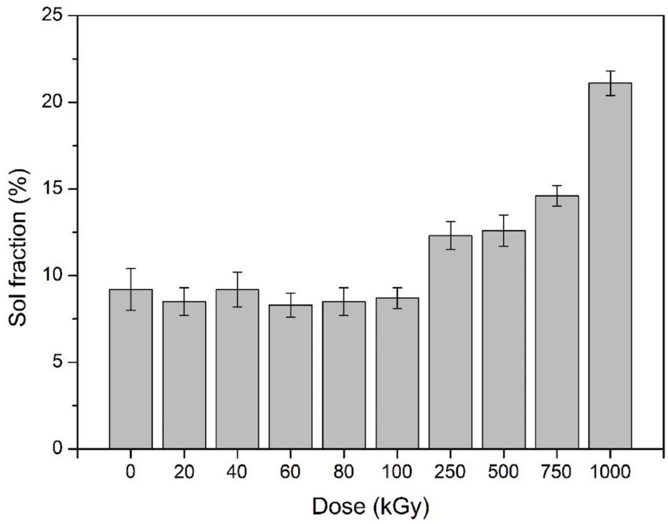


Fig. 2. The sol fractions of the examined GTRs.

the rubber in the swollen sample, which can be calculated with the Ellis and Welding Eq. (3) (Ellis and Welding, 1964).

$$V_r = \frac{\frac{m_r}{\rho_r}}{\frac{m_r}{\rho_r} + \frac{m_s}{\rho_s}}, \tag{3}$$

where m_r is the mass of the dry sample in grams; m_s is the mass of the swollen sample in grams; ρ_s is the density of the solvent (0.8669 g/cm^3), and ρ_r is the density of the GTR (1.20 g/cm^3).

The decrease in cross-link density, i.e., the degree of devulcanization (Dev (%)), can be determined using Eq. (4).

$$Dev\ (\%) = \left(1 - \frac{\nu_f}{\nu_i}\right) \cdot 100, \tag{4}$$

where ν_f is the cross-link density of the irradiated GTR and ν_i is the cross-link density of the untreated GTR.

In order to gain a deeper understanding of the changes that took place in the GTR due to radiation, we performed a Horikx analysis using the results of Soxhlet extraction and swelling tests. This analysis can be used to determine the ratio of chain-scission and the selective cross-link break in a degraded cross-linked polymer (Horikx, 1956).

One extreme case is when only the main chain scission occurs. In this scenario, the relative decrease in the soluble fraction and cross-linking density can be described by Eq. (5), which describes the random scission of the main chain.

$$1 - \frac{\nu_f}{\nu_i} = 1 - \frac{\left(1 - \sqrt{s_f}\right)^2}{\left(1 - \sqrt{s_i}\right)^2}, \tag{5}$$

where ν_i stands for the initial cross-link density, ν_f stands for the cross-link density after the irradiation; s_i stands for the initial sol fraction of the GTR, and s_f stands for the sol fraction of the GTR after the radiation treatment.

The other scenario involves only cross-link cleavage, with no scission of the polymer backbone, in which case Eq. (6) can be written.

$$1 - \frac{\nu_f}{\nu_i} = 1 - \frac{\gamma_f\left(1 - \sqrt{s_f}\right)^2}{\gamma_i\left(1 - \sqrt{s_i}\right)^2}, \tag{6}$$

where γ_i and γ_f stands for the initial and final cross-linking index, respectively.

In the case of Horikx analysis, we corrected the results of the Soxhlet extractions (relative soluble fraction), by subtracting the sol content of the untreated GTR from every sample.

After the characterization of the GTR, we prepared the mixtures. The natural rubber used was the NR TSR 10 type (Sud Comoe Caoutchuc, Aboisso, Ivory Coast), while the additives are summarized in Table 1.

In this study, we examined two mixture types: NR-based and two-step mixing. The former is based on a general tire tread recipe, while the latter differs in that it contains excess vulcanizing agent (which will be described in detail later).

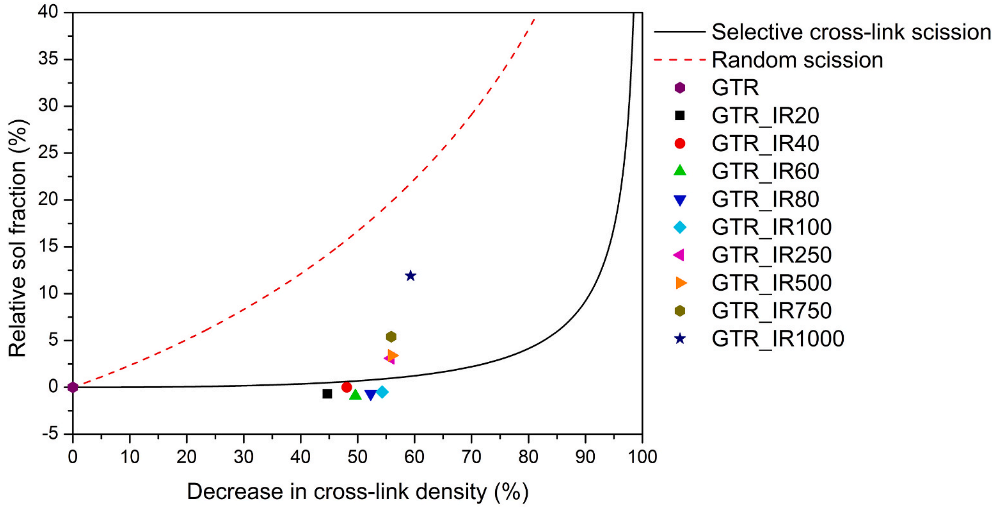


Fig. 3. Horikx plot of the irradiated GTRs.

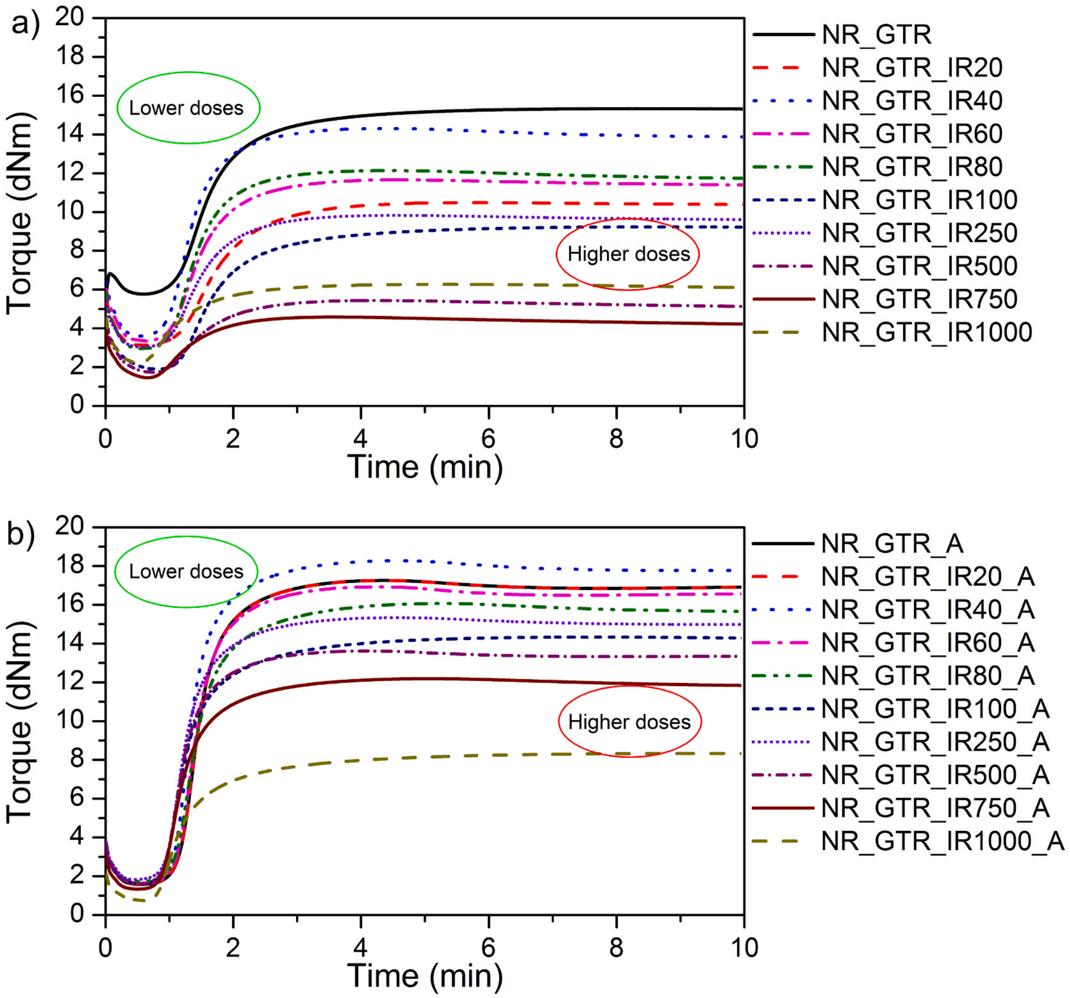


Fig. 4. Vulcanization curves for **a)** NR-based compounds **b)** for two-step compounds.

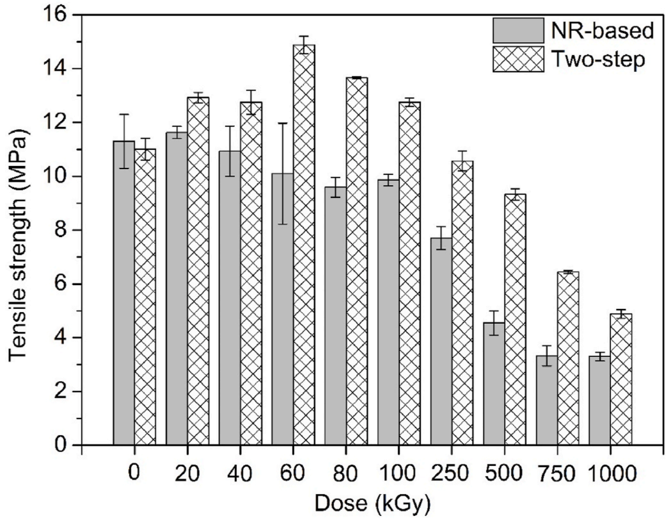


Fig. 5. Tensile strength of the GTR containing rubber mixtures as a function of gamma radiation dose.

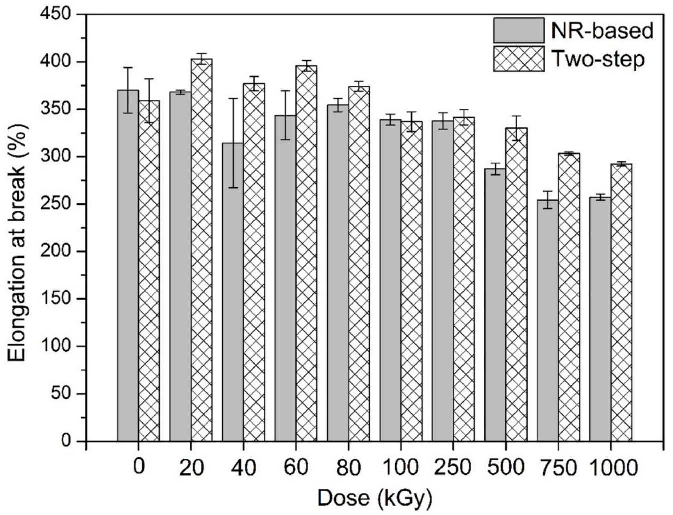


Fig. 6. Elongation at break values of the GTR containing rubber mixtures as a function of gamma radiation dose.

First, we prepared the NR-based mixtures using the treated and untreated GTR (as reference); the recipe was: 100 phr NR, 60 phr CB, 10 phr ZnO, 2 phr stearic acid, 100 phr GTR, 10 phr paraffin oil, 1.25 phr CBS, 0.6 phr TMTD, 0.6 phr sulphur. The nomination of the reference (untreated GTR) was NR_GTR, while the mixtures containing irradiated GTR were marked as NR_GTR_IR*, where * stands for the absorbed dose; thus, the sample containing 100 kGy absorbed GTR is denoted as

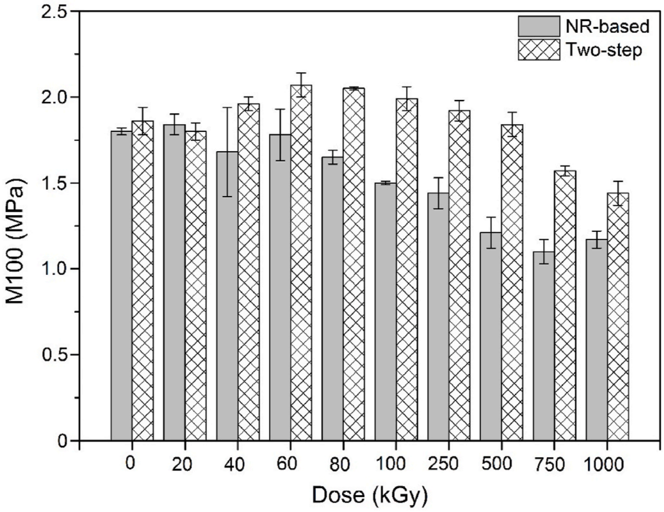


Fig. 7. Modulus at 100% of elongation of the GTR containing mixtures as a function of gamma radiation dose.

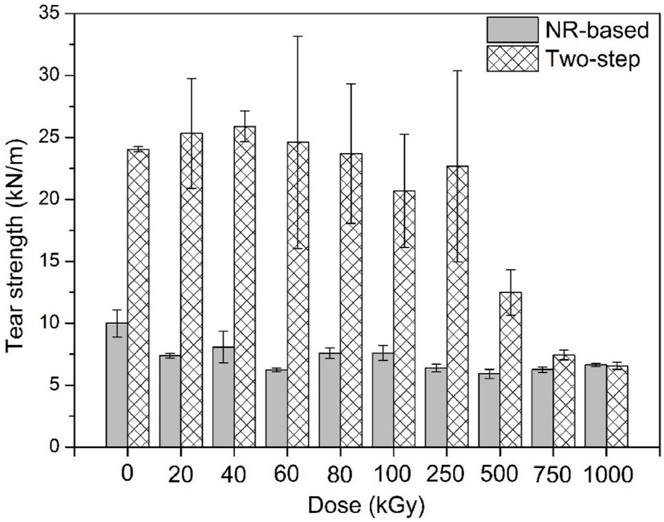


Fig. 8. Tear strength of the GTR containing rubber mixtures as a function of gamma radiation dose.

NR_GTR_IR100.

To include the excess vulcanizing agent in the two-step mixtures, we made GTR_IR based pre-mixtures. These differed from the composition of the NR-based mixtures (mentioned above) in that they did not contain NR and CB, so the recipe was: 10 phr ZnO, 2 phr stearic acid, 100 phr GTR, 10 phr paraffin oil, 1.25 phr CBS, 0.6 phr TMTD and 0.6 phr sulphur.

Lastly, we produced the two-step mixtures. Their formulation was identical to the NR-based mixtures presented above, except that instead of 100 phr GTR, they contained 100 phr of the corresponding GTR_IR pre-mixture. The nomination of the reference was NR_GTR_A, which contained the untreated GTR containing pre-mixture. The materials containing GTR_IR pre-mixtures were abbreviated as NR_GTR_IR*_A, where * stands for the absorbed dose; thus, the sample containing 100 kGy absorbed GTR pre-mixture is denoted as NR_GTR_IR100_A.

We prepared the mixtures in a Brabender Lab-Station internal mixer (Brabender GmbH & Co. KG, Duisburg, Germany) at a temperature of 50 °C and a rotor speed of 40 rpm. The order of appearance for the components written above reflects the order of mixing.

To determine the curing characteristics of the compounds, we recorded the vulcanization curves of the mixtures with a MonTech D-RPA 3000 moving die rheometer (MonTech Werkstofprüfmaschinen GmbH, Buchen, Germany). The measurements were carried out at 160 °C with an amplitude of 1° and a frequency of 1.67 Hz for 20 min.

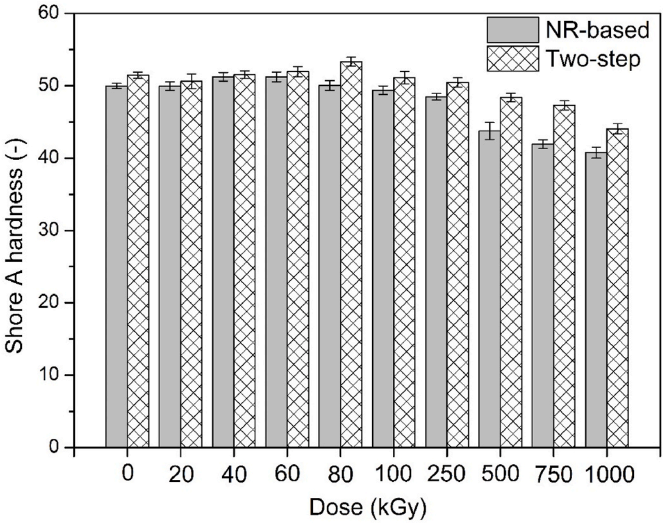


Fig. 9. Shore A hardness of the GTR containing rubber mixtures as a function of gamma radiation dose.

We vulcanized the compounds in a Teach-Line Platen Press 200E (Dr. Collin GmbH, Munich, Germany) hydraulic press. The applied pressure was 2.8 MPa, while the temperature was 160 °C. Each compound was pressed into 200 mm x 200 mm x 1 mm sheets, and the curing lasted for t_{90}, which is the time needed for 90% vulcanization. These times were acquired from the rheometer measurements.

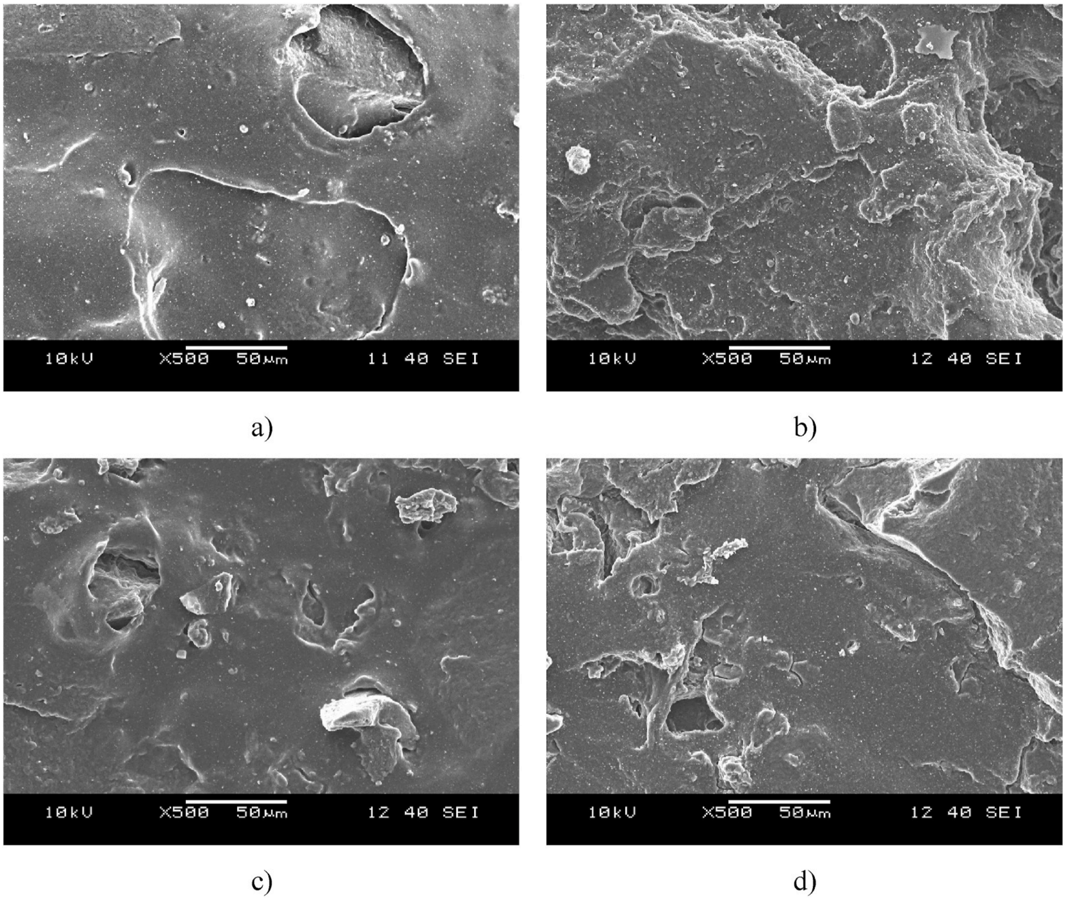
We revealed the mechanical properties of the compounds using a Zwick-Z005 universal testing machine (Zwick GmbH., Ulm, Germany) at room temperature. Tensile tests were performed according to the DIN 53504 standard, using Type 1 specimens with a clamping length of 60 mm and a crosshead speed of 500 mm/min. Tear tests were performed according to the ASTM D624 standard, using type C specimens and the same test speed of 500 mm/min with a clamping distance of 56 mm. The average and standard deviation values of the tensile strength, tear strength, and elongation at break were determined for each compound. We also calculated the M100 values using the results of the tensile tests, which is the stress value at 100% elongation.

We determined the hardness of the cured compounds according to the ISO 48–4:2018 Shore A method on a Zwick H04.3150.000 hardness tester (Zwick GmbH., Ulm, Germany). For each material, ten tests were performed, then the average and standard deviation values were calculated.

We studied the morphology of the GTR and the fracture surfaces of the tensile specimens using scanning electron micrographs, which were taken with a JEOL JSM 6380LA (Jeol Ltd., Tokyo, Japan) scanning electron microscope after sputtering with gold.

3. Results and discussion

The results of the Soxhlet extractions are shown in Fig. 2. At low doses (≤100 kGy), the sol content did not change significantly, suggesting that in this range, small molecular weight parts have not yet appeared; the GTR was relatively stable; however, at high doses, the sol content significantly increased. Ashfaq et al. studied the processes that take place during the irradiation of polymers at different doses and dose rates. They found out that at low dose rates cross-linking and chain-scission are two competing processes. At low doses, these two are able to compensate each other, which is consistent with our results (soluble content did not change), but at higher doses the scission of low



a) b) c) d)

Fig. 10. SEM micrographs of the fracture surfaces of the a) NR_GTR; b) NR_GTR_A; c) NR_GTR_IR1000, and c) NR_GTR_IR1000_A materials.

molecular weight parts take precedence. The longer irradiation time favours the chain-scission of the polymer backbone, which is consistent with our results (sol content increased significantly) (Ashfaq et al., 2020).

Fig. 3 Shows the results of the Horikx analysis. The diagram clearly shows that up to a dose of 100 kGy, the samples follow each other near the curve of the selective cross-linking, which means that the sulphur cross-links were selectively broken. The cross-link density decreases as the absorbed dose increases while the soluble content remains unchanged. One can see that above 100 kGy, the calculated values follow roughly a vertical line, which suggests that the cross-links breaks were less dominant, but chain-scission become to the fore.

The vulcanization curves of the compounds are represented in Fig. 4. In the case of NR-based materials (Fig. 4a.), the torque decreased with the increase of the absorbed dose. One can see that the compounds containing GTR that absorbed high doses ($\geq$500 kGy) showed much lower maximum torque values. Fig. 4b displays the vulcanization curves of the two-step mixtures in Fig. 4b. In this case, the maximum torque values are in a higher range than in the case of NR-based mixtures. This can be attributed to the fact that in these recipes more sulphur was added to the system, as a result of which more cross-links can form in the material. The torques for the same doses were in a larger range, and the minimum torque values did not decrease with the higher absorbed doses compared to the NR-based mixtures, which suggests a better connection between the phases in the case of two-step mixtures.

Fig. 5 shows the tensile strength of the materials. In the case of NR-based compounds, the gamma radiation treatment alone was not sufficient to improve tensile strength. However, the combined use of radiation and excess vulcanizing agent brought significant improvements regarding the reference materials, in the range of 20–80 kGy. The best result was obtained with the 60 kGy sample, where the tensile strength improved by almost 35% compared to the NR_GTR reference material. The increased strength can be explained by the better adhesion between the phases, which correlates with the results of the vulcanization curves. It means that a synergistic effect awakes when using the two methods together. At higher doses the tensile strength decreases compared to the references, the interfacial adhesion between the GTR and the matrix was inadequate. Also, the radiation induced degradation of the polymer backbone of the GTR leads to a decrease in the tensile strength, which also has an adverse effect on the mixture.

The elongation at break values are represented in Fig. 6. In the case of NR-based compounds, the elongation at break slightly decreased; however, even at high doses, it proved to be over 300%, which is especially good. Based on the results of the two-step mixtures, the elongation at break values improved in the 20–80 kGy range due to the better adhesion between the phases. Gamma radiation treatment and including excess vulcanizing agent in a mixture have a synergistic effect, as the reference values show that two-step mixing alone did not improve the tensile strength and elongation at break values.

Fig. 7 shows the stress at 100% elongation (M100) of the mixtures. The modulus values are higher for two-step mixtures in most cases than NR-based mixtures—a better interfacial adhesion formed between the phases, which is in line with the results. This can be attributed to the increased cross-link density in the materials (similar results can be seen for the hardness values). The M100 values did not decrease significantly. The absorbed dose and the materials with the highest modulus were in the 40–80 kGy range.

Fig. 8 shows the tear strength of the investigated materials. The tear strength more than doubled if there was the additional vulcanizing agent in the system. During the irradiation peroxides appeared on the surface of the GTR, which were able to react with the additional vulcanizing agent, then with the unsaturated bonds of the fresh rubber, resulting in a better interfacial adhesion between the phases. The radiation treatment had no significant effect alone, and the tear strength values nearly remained the same. In the case of combining the two treatments, there is an improvement in the low dose range, and the best result was obtained at 40 kGy.

The results of the hardness tests are represented in Fig. 9. Based on the results, a modest increase can be seen, which is larger in the case of the two-step mixtures since hardness is roughly related to the number of cross-links in the material. At higher doses, the chain-scission caused by high dosage radiation could not be compensated by the formulating cross-links; consequently, the hardness decreased.

In order to examine the morphology, SEM micrographs were taken of the fracture surfaces of the tensile specimens of some materials, as shown in Fig. 1. Both treated and untreated GTR were able to integrate properly into the matrix. Homogenous structures were formed, which was not affected by the absorbed dose. Even at higher doses (Fig. 10c and Fig. 10d) the morphology remained the same, despite the significant changes in the mechanical properties. Similar conclusions can be drawn in the case of two-step mixing, which did not affect the morphology.

4. Conclusions

In this study, we treated ground tire rubber with ionizing radiation and found, that at low doses, the selective scission of sulphur cross-links was the dominant process; however, at higher doses (>100 kGy) the chains-scission of the polymer backbone became to the fore. We produced different GTR containing mixtures using conventional and two-step mixing techniques. The combined use of gamma radiation treatment and two-step mixing resulted in better adhesion between the phases in the mixtures. During irradiation the unsaturated bond of the GTR can react with the oxygen in the air, resulting in peroxides on the surface. These reactive functional groups might have reacted with the additional vulcanizing agent and then with the unsaturated bond of the fresh rubber phase, thus the better connection between the phases. As a result, the mechanical properties of the materials (tensile strength, elongation at break, tear strength) improved. When the two treatments were applied separately, no significant improvement in the properties of the mixtures was observed, so a synergistic effect awakes when the two are combined.

Author contribution statement

Lóránt Kiss: manuscript writing, results evaluation.
Dániel Ábel Simon: experimental work.
Tamás Bárány: experimental design, reviewing the manuscript.
László Mészáros: supervision, reviewing the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

The research reported in this paper is part of project no. BME-NVA-02, implemented with the support provided by the Ministry of Innovation and Technology of Hungary from the National Research, Development and Innovation Fund, financed under the TKP2021 funding scheme. This work was supported by the National Research, Development and Innovation Office, Hungary (K128268). László Mészáros is thankful for János Bolyai Research Scholarship of the Hungarian Academy of Sciences, and for the ÚNKP-21-5 New National Excellence Program of the Ministry for Innovation and Technology. The authors also extend their acknowledgment to the International Atomic Energy Agency (IAEA) for financial support under the umbrella of CRP (Coordinated Research Project). The authors are grateful to Professor Erzsébet Takács for her valuable advices.

References

Ashfaq, A., Clochard, M.-C., Coqueret, X., Dispenza, C., Driscoll, M.S., Ulański, P., Al-Sheikhly, M., 2020. Polymerization reactions and modifications of polymers by ionizing radiation. Polymers 12, 2877.

Cataldo, F., Ursini, O., Angelini, G., 2010. Surface oxidation of rubber crumb with ozone. Polym. Degrad. Stabil. 95, 803–810.

Colom, X., Carrillo, F., Cañavate, J., 2007. Composites reinforced with reused tyres: Surface oxidant treatment to improve the interfacial compatibility. Compos. Appl. Sci. Manuf. 38, 44–50.

Ellis, B., Welding, G.N., 1964. Estimation, from swelling, of the structural contribution of chemical reactions to the vulcanization of natural rubber. Part II. Estimation of equilibrium degree of swelling. Rubber Chem. Technol. 37, 571–575.

Flory, P.J., Rehner, J.J., 1943. Statistical mechanics of cross-linked polymer networks II. Swelling. J. Chem. Phys. 11, 521–526.

Gopi Sathi, S., Harea, E., Machu, A., Stocek, R., 2021. Facilitating high-temperature curing of natural rubber with a conventional accelerated-sulphur system using a synergistic combination of bismaleimides. Express Polym. Lett. 15, 16–27.

Horikx, M.M., 1956. Chain scissions in a polymer network. J. Polym. Sci. 19, 445–454.

Karger-Kocsis, J., Mészáros, L., Bárány, T., 2013. Ground tyre rubber (GTR) in thermoplastics, thermosets, and rubbers. J. Mater. Sci. 48, 1–38.

Keizo, M., Song, C., 2012. Basic concepts of radiation processing. In: Keizo, M., Song, C. (Eds.), Radiation Processing of Polymer Materials and its Industrial Applications. John Wiley & Sons, Hoboken, USA, pp. 1–25.

Kiss, L., Simon, D.Á., Petrény, R., Kocsis, D., Bárány, T., Mészáros, L., 2022. Ground tire rubber filled low-density polyethylene: The effect of particle size. Adv. Ind. Eng. Polym. Res. 5, 12–17.

Mészáros, L., Bárány, T., Czvikovszky, T., 2012. EB-promoted recycling of waste tire rubber with polyolefins. Radiat. Phys. Chem. 81, 1357–1360.

Naskar, A.K., De, S.K., Bhowmick, A.K., 2002. Thermoplastic elastomeric composition based on maleic anhydride-grafted ground rubber tire. J. Appl. Polym. Sci. 84, 370–378.

Paleri, D.M., Rodriguez-Uribe, A., Misra, M., Mohanty, A.K., 2021. Preparation and characterization of eco-friendly hybrid biocomposites from natural rubber, biocarbon, and carbon black. Express Polym. Lett. 15, 236–249.

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

Ratnam, C., Ramarad, S., Khalid, M., Noraini, N., 2013. Effect of pre-irradiation of waste tire dust on the properties of ethylene vinyl acetate/waste tire dust blend (EVA/WTD) blends. Journal of J. Compos. Biodegradable Polym. Polymers 1, 16–22.

Şen, M., Uzun, C., Kantoğlu, Ö., Erdoğan, S.M., Deniz, V., Güven, O., 2003. Effect of gamma irradiation conditions on the radiation-induced degradation of isobutylene–isoprene rubber. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 208, 480–484.

Simon, D.Á., Bárány, T., 2021. Effective thermomechanical devulcanization of ground tire rubber with a co-rotating twin-screw extruder. Polym. Degrad. Stabil. 190, 109626.

Simon, D.Á., Pirityi, D., Tamás-Bényei, P., Bárány, T., 2020a. Microwave devulcanization of ground tire rubber and applicability in SBR compounds. J. Appl. Polym. Sci. 137, 48351.

Simon, D.Á., Pirityi, D.Z., Bárány, T., 2020b. Devulcanization of ground tire rubber: microwave and thermomechanical approaches. Sci. Rep. 10, 16587.

Sonnier, R., Leroy, E., Clerc, L., Bergeret, A., Lopez-Cuesta, J.M., 2006. Compatibilisation of polyethylene/ground tyre rubber blends by γ irradiation. Polym. Degrad. Stabil. 91, 2375–2379.

Sonnier, R., Leroy, E., Clerc, L., Bergeret, A., Lopez-Cuesta, J.M., 2007. Polyethylene/ground tyre rubber blends: influence of particle morphology and oxidation on mechanical properties. Polym. Test. 26, 274–281.

Tolstov, A., Grigoryeva, O., Fainleib, A., Danilenko, I., Spanoudaki, A., Pissis, P., Grenet, J., 2007. Reactive compatibilization of polyethylene/ground tire rubber inhomogeneous blends via interactions of pre-functionalized polymers in interface. Macromol. Symp. 254, 226–232.

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