RESEARCH ARTICLE



Sustainable thermoplastic elastomers based on thermoplastic polyurethane and ground tire rubber

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Funding information

National Research, Development and Innovation Office, Grant/Award Numbers: K 128268, K 146085; National Research, Development and Innovation Fund, Grant/Award Numbers: TKP-6-6/ PALY-2021, TKP2021-NVA

Abstract

In this research, blends are prepared of thermoplastic polyurethane (TPU), ground tire rubber (GTR) and its devulcanized version (dGTR). The primary objective is to produce thermoplastic elastomers wherein TPU is partially substituted by GTR or dGTR obtained from used tires, thereby forming a blend with a favorable cost/value ratio and a smaller environmental footprint. Throughout the experiment, the rubber content of the blends is varied between 0 and 50 wt% and the effect on mechanical properties is investigated. The blends are compounded with a twin-screw extruder, after which sheet samples are produced by injection molding. With a view to a possible future industrial application, it is important that both the compounding and the injection molding of the specimens are easy to perform, even with a 50 wt% filler content. Increasing the amount of rubber phase reduce the tensile strength and elongation at break of the blends. Unfortunately, devulcanization did not significantly improve the properties of the blends. Overall, even at a (d)GTR content of 50 wt%, an elongation at break of 300% is achieved, which allows the use of the blends as thermoplastic elastomers. In addition, dynamic tests show that the rubber phase increases the damping capacity of the samples.

K E Y W O R D S

circular economy, ground tire rubber, recycled rubber, thermoplastic elastomers

1 | INTRODUCTION

The production and consumption of cross-linked elastomers or rubbers continues to grow due to their beneficial properties. This means that more and more rubber waste needs to be managed worldwide every year. Unlike thermoplastic polymers, the three-dimensional cross-linked structure of rubber does not allow it to be recycled in a molten state. As a result, recycling used rubber products is a major challenge and is a serious environmental issue. More than 60% of rubber waste comes from various types of tires. It is estimated that 1.5 billion end-of-life tires (ELTs) are produced worldwide each year, of which more than 40% by weight is made up of various synthetic and natural rubber components. Tires are highly complex products with multiple reinforcing structures (like beads, steel, and textile belts) and up to 10–15 different rubber compounds, which further complicates their recycling.^{1–5}

For many decades, used tires were dumped in landfills or burned in incinerators, and their chemical energy

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was recovered in the form of heat. However, dumping in landfills poses significant environmental risks and is now regulated by strict legislation. Under EU Directive 1999/31/EC, whole tires have been banned from landfills since 2003 and shredded tires since $2006.^{6-8}$

In addition to legal requirements, the aim is to recycle ELTs in their material for economic and environmental reasons. The elastomer parts are usually ground with various methods (mechanical, cryogenic, or water jet milling). The resulting ground tire rubber (GTR), from which the reinforcing structures have been removed, is commercially available in sufficient quality and quantity. GTR is mostly used to provide the surface of, for example, sports fields or playgrounds, but this is only a small proportion of the amount produced, so new methods and applications need to be found. An obvious solution would be to reuse the rubber regrind in new rubber compounds. In low quantities (<10 wt%), it does not produce a significant degradation of properties.⁹

An important application for rubber regrind could be the production of toughened thermoplastics or multiple recyclable thermoplastic vulcanizates (TPVs), when the regrind is combined with thermoplastic polymers. TPVs belong to the family of high-performance thermoplastic elastomers (TPEs) and they are gaining in importance today.⁸ TPVs are two-phase systems, consisting of a continuous thermoplastic polymer matrix with dispersed vulcanized rubber particles.¹⁰ TPEs are a bridge between thermoplastic polymers and conventional cross-linked elastomers. Their great advantage is that they can be processed with conventional thermoplastic processing techniques, they are recyclable in the molten state, and have the mechanical properties of rubbers.^{11,12} Thanks to their advantageous properties, they are replacing traditional rubber in many applications, such as pipes, seals, shoe soles, and so forth.¹³⁻¹⁵

Recently, numerous studies have been published in which various thermoplastic polymers were blended with GTR, for example, polyolefins like polyethylene $^{16-18}$ and polypropylene,^{19–21} and also polystyrene,²² polylactic acid,²³ ethylene vinyl acetate,^{24,25} acrylonitrile-butadienestvrene²⁶ and even thermoplastic polyurethane (TPU).^{27,28} However, the adhesion between the two phases is often weak. Several modification methods are available to improve the connection between the GTR and the thermoplastic polymer matrix, including devulcanization.²⁹ It means the recovery of plasticity by breaking down the 3D cross-linked structure of the rubber by selectively chopping the intermolecular sulfur-sulfur or carbon-sulfur bonds. There are various methods of rubber devulcanization, such as microwave, thermochemical, mechanochemical, biological, and thermomechanical devulcanization.^{30,31} Taking into account productivity

and technological and economic considerations, thermomechanical devulcanization seems to be an advantageous option. Using heat and shear forces generated by a twinscrew extruder to break the cross-linked structure provides a continuous process with high efficiency, short treatment time, and good product quality.³²

TPUs are a popular group of TPEs. These TPUs are block copolymers whose molecular chains consist of hard and soft segments. These materials have excellent properties such as high elasticity, wear resistance, and chemical resistance, and robust low-temperature resistance.^{12,15,33} However, the price of TPUs is relatively high. Incorporating rubber compounds^{34,35} or GTR into a TPU is an effective approach to optimizing the cost of these materials in everyday life while retaining their elastomeric properties. This process can result in a thermoplastic elastomer with balanced properties and a small environmental footprint.²⁷ The resulting TPU/GTR blends can be used as FDM 3D printing filaments that can be used to make rubber-like (color, smell and feel) products.

Our goal was to develop sustainable TPEs in which the TPU is partially replaced by GTR from used tires. During the experiment, we investigated the effect of the amount of rubber phase on the properties of the blends. We would like to improve adhesion between the phases by devulcanizing the GTR.

2 | EXPERIMENTAL

2.1 | Materials

A polyester-based TPU, Elastollan C 78 A 15 (BASF, Mannheim, Germany) was used as the matrix in our blends. The GTR was kindly provided by AquaJet Ltd (Budapest, Hungary). The GTR was produced by high-pressure water-jet milling, with a particle size of less than 400 μ m, from the tread part of truck tires.

2.2 | Sample preparation

The thermomechanical devulcanization of the GTR was performed with a Labtech Scientific LTE-26-44 corotating, intermeshing twin-screw extruder (Labtech Engineering Co., Ltd., Samutprakarn, Thailand). Based on our previous research, we set all heating zones of the equipment to 190°C and screw speed to 60 rpm. These parameters were chosen because, based on Horikx's analysis, these settings provide a good approximation to the theoretical curve for selective cross-link scission.³²

In all cases, the TPU, the GTR and the blends were dried at 80° C for 4 h in a Binder VD 53 vacuum drying

oven (Binder GmbH, Tuttlingen, Germany) equipped with a Vacuubrand MZ 2 NT vacuum pump (Vacuubrand GmbH, Wertheim, Germany) prior to processing.

The blends were also produced with the twin-screw extruder described above. The first step was to produce dry blends of TPU and GTR or devulcanized GTR (dGTR). The composition and name of the samples are listed in Table 1. The filaments exiting the die were air cooled with a fan-assisted conveyor belt and then granulated with a Labtech LZ 120/VS (Labtech Engineering Co., Ltd., Samutprakarn, Thailand) granulator. The temperature profile used during extrusion (from the feed section to the die) was 185-185-190-195-195-200-200-205-205-205°C and screw speed was 120 rpm.

We injection molded 2 mm thick 80×80 mm sheets from the prepared blends using an Arburg Allrounder Advance 270S 400–170 injection molding machine (Arburg GmbH, Lossburg, Germany). Table 2 contains the injection molding parameters. The specimens for the tests were cut from these injection molded specimens.

TABLE 1 Composition	of TPU/(d)GTR blends.
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	Amount of components (wt%)		
Sample	TPU	GTR	dGTR
TPU	100	—	—
TPU/GTR _{90/10}	90	10	_
TPU/GTR _{80/20}	80	20	_
TPU/GTR _{70/30}	70	30	_
TPU/GTR _{60/40}	60	40	_
TPU/GTR _{50/40}	50	50	_
TPU/dGTR _{90/10}	90	_	10
TPU/dGTR _{80/20}	80	_	20
TPU/dGTR _{70/30}	70	_	30
TPU/dGTR _{60/40}	60	_	40
TPU/dGTR _{50/50}	50	—	50

Abbreviations: dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane.

TABLE 2 Injection molding parameters.

Parameter	Value
Melt temperature (°C)	210
Injection speed ($cm^3 \cdot s^{-1}$)	50
Dose volume (cm ³)	45
Holding pressure (bar)	450
Holding time (s)	10
Residual cooling time (s)	50
Mold temperature (°C)	30

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2.3 | Test methods

The mechanical properties of the blends were determined with a Zwick Z005 universal testing machine (Zwick GmbH, Ulm, Germany) equipped with a 5 kN load cell. The specimens were tensile tested, and tear tested at room temperature with a crosshead speed of $500 \text{ mm} \cdot \text{min}^{-1}$, and a grip length of 45 mm. The tensile test was carried out on Type 2 dumbbell specimens according to ISO 37:2017. The tear test was carried out according to the ISO 34-1:2015, Method B, on angle test pieces with a nick. In each case, 5 tests were performed and the results were averaged.

The Shore A hardness of the injection molded plates was determined with a Zwick H04.3150.000 hardness tester (Zwick GmbH, Ulm, Germany) according to ISO 48-4:2018, at 10 points per sample. The thickness of the samples was approximately 6 mm (3 plates stacked).

The compression set of the samples was determined according to ISO 815-1:2019, Method A. Type B circular specimens (13 mm diameter, 6 mm thickness) were compressed by 25% of their original thickness and placed in a drying oven (Baxter DN-63, Baxter, International, Deerfield, USA) at 70°C for 24 h. The "pressure device" was then removed from the drying oven, the pressure was released, and the samples were placed on a wooden board, where they relaxed for 30 min, then the thickness of the samples was measured again. The compression set was the average of the results at least 5 samples in each case.

The viscoelastic properties of the blends were analyzed by oscillating shear with a MonTech D-RPA 3000 rubber process analyzer (RPA) (MonTech Werkstoffprüfmaschinen GmbH, Buchen, Germany). First, we performed an amplitude sweep at 10 Hz at 30° C from 0.01° to 1.50° . We determined the limits of the linear viscoelasticity of the materials based on 30 measurement points at each amplitude and chose an appropriate amplitude for the frequency sweep. The frequency sweep was performed on the specimens at 30° C from 0.1 to 10 Hz, at an amplitude of 0.05° (at least 5 points at each frequency).

The falling weight impact test of the samples was performed with a Ceast 9350 impact tester (Instron, Massachusetts, USA). The specimen was placed on a support with a hole and positioned with a clamping ring. The injection molded plates were used for the test. The total mass of the dart was 20.036 kg and height was 1.0 m, therefore impact energy was 196.5 J. The diameter of the hemispherical dart tip was 20 mm and the diameter of the support ring was 40 mm. In each case, the mechanical properties determined are the average of the results of 10 tests.

The morphology of the samples was examined with a JEOL JSM 6380LA scanning electron microscope (SEM)

(Jeol Ltd., Tokyo, Japan). Based on our previous experience, it was not possible to clearly distinguish the two phases (TPU and the rubber phase) in the images of the cryogenically fractured surface of the samples. Therefore, the cross-section of the severed dumbbell specimens was studied. Before electron microscopy, these surfaces were coated with a thin layer of gold.

3 | RESULTS

35

30

25

20

15

10

5

0

0

Stress (MPa)

3.1 | Mechanical properties

TPU

100

200

TPU/GTR_{90/10}

TPU/GTR_{80/20}

FPU/GTR

TPU/GTR_{60/40}

TPU/GTR.

The tensile test results (Figure 1, Table 3) indicate that the tensile strength of the specimens and the stress for a given elongation (a modulus-like characteristic) decreased with the addition of GTR and dGTR. In contrast, the elongation at break of the specimens increased slightly with 10 wt% dGTR, but decreased if more rubber was added.¹⁸ It was probably due to the small amount of rubber phase that includes longer, shorter free rubber chains, which facilitated the mobility of the molecular chains of the matrix polymer. So a small amount of dGTR acts as a toughening agent that improves the elongation at break of the blends. The rubber particles in the TPU matrix can absorb and dissipate energy, allowing the material to stretch further before breaking.³⁶ However, when the amount was further increased, it acted as a failure site due to the weak bond between the two phases.

The stiffness of the samples (Table 3) decreased as the rubber phase was increased, and the devulcanization had

FIGURE 1 Typical tensile curves of the TPU matrix and blends: (a) TPU/GTR, (b) TPU/dGTR. dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane. [Color figure can be viewed at wileyonlinelibrary.com]

(a)

700

35

30

25

20

15

10

5

0

0

Stress (MPa)

TPU

100

TPU/dGTR_{90/10}

TPU/dGTR_{80/20} TPU/dGTR_{70/30}

TPU/dGTR 60/40

TPU/dGTR 50/50

200

300

400

Strain (%)

500

600

TABLE 3	The mechanical	properties of the	matrix and the blends.
INDLUJ	The incentation	properties of the	matrix and the orenus.

300

400

Strain (%)

500

600

Sample	Tensile strength (MPa)	Elongation at break (%)	M50 (MPa)	M100 (MPa)	M300 (MPa)	Tear strength (N∙mm ⁻¹)
TPU	33.0 ± 0.8	670 ± 21	4.5 ± 0.0	5.5 ± 0.0	10.5 ± 0.2	63.4 ± 1.4
TPU/GTR _{90/10}	24.2 ± 0.9	681 ± 16	4.1 ± 0.0	5.2 ± 0.0	9.6 ± 0.1	53.8 ± 1.9
TPU/GTR _{80/20}	15.3 ± 0.7	572 ± 18	3.8 ± 0.0	5.0 ± 0.1	9.3 ± 0.2	52.7 ± 1.0
TPU/GTR _{70/30}	11.2 ± 0.5	485 ± 23	3.6 ± 0.1	4.9 ± 0.1	9.4 ± 0.1	46.2 ± 0.9
TPU/GTR _{60/40}	9.2 ± 0.2	367 ± 13	3.3 ± 0.0	4.6 ± 0.0	9.1 ± 0.1	41.5 ± 1.1
TPU/GTR _{50/50}	6.8 ± 0.2	304 ± 17	2.7 ± 0.0	4.0 ± 0.1	6.5 ± 0.3	30.7 ± 0.9
TPU/dGTR _{90/10}	29.1 ± 1.1	714 ± 18	4.2 ± 0.0	5.3 ± 0.0	9.6 ± 0.1	53.4 ± 1.1
TPU/dGTR _{80/20}	19.5 ± 0.8	600 ± 24	3.8 ± 0.1	5.1 ± 0.1	9.2 ± 0.2	48.7 ± 1.8
TPU/dGTR _{70/30}	13.1 ± 0.3	500 ± 11	3.5 ± 0.0	4.9 ± 0.0	8.6 ± 0.1	47.6 ± 1.5
TPU/dGTR _{60/40}	9.4 ± 0.9	442 ± 28	3.2 ± 0.1	4.6 ± 0.1	7.4 ± 0.3	41.6 ± 0.8
TPU/dGTR _{50/50}	5.4 ± 0.1	305 ± 31	2.6 ± 0.0	3.9 ± 0.1	5.1 ± 0.2	32.1 ± 0.6

Abbreviations: dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane.

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(b)

700

TABLE 4 The Shore A hardness and compression set of the samples.

Sample	Shore A hardness	Compression set (24 h, 70°C) (%)
TPU	85.7 ± 0.3	57 ± 1.8
TPU/GTR _{90/10}	83.6 ± 0.4	56 ± 2.0
TPU/GTR _{80/20}	81.8 ± 0.4	61 ± 1.4
TPU/GTR70/30	80.0 ± 0.6	62 ± 1.3
TPU/GTR _{60/40}	77.8 ± 0.5	59 <u>±</u> 2.0
TPU/GTR _{50/50}	71.4 ± 0.4	70 ± 1.6
TPU/dGTR _{90/10}	83.5 ± 0.3	67 ± 1.2
TPU/dGTR _{80/20}	81.3 ± 0.7	67 ± 2.5
TPU/dGTR _{70/30}	79.1 ± 0.7	70 ± 1.4
TPU/dGTR _{60/40}	77.1 ± 0.6	67 ± 2.2
TPU/dGTR _{50/50}	70.2 ± 0.7	75 ± 0.6

Abbreviations: dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane.

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no significant effect. This corresponds to the change in Shore A hardness (Table 4) of the samples.

Figure 2 clearly shows that both tensile strength and elongation at break were higher for samples containing dGTR. This can be attributed to the fact that during devulcanization process, some of the cross-links and/or molecular chains in the rubber were broken. As a result, the polymer chains of the rubber phase become more mobile and were able to deform to a greater extent under load and the mechanical compatibility of the two phases was improved by devulcanization.

The rubber phase reduced the tear strength of the samples, but devulcanization had no significant effect on the resistance to crack propagation (Table 3). The reduction in tear strength is also related to the fact that cracks propagate rapidly through the surface due to the weaker contact between the phases.



FIGURE 2 (a) tensile strength (b) elongation at break, and (c) Shore A hardness of the samples as a function of (d)GTR content. dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane. [Color figure can be viewed at wileyonlinelibrary.com]

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The Shore A hardness of the samples (Figure 2c, Table 4) decreased with an increasing amount of (d)GTR compared to the TPU matrix. In addition, we found that the devulcanization of GTR did not affect the hardness of the blends.

We investigated the compression set of the samples at 70°C. No significant change was observed with GTR, whereas the compression set of the samples containing dGTR increased by almost 20%. This decrease is associated with a decrease in cross-link density due to the devulcanization of the rubber phase. The residual deformation of the rubber phase, and therefore the residual deformation of the entire sample, increases as the cross-link density decreases.

3.2 | Dynamic properties

The response of the samples to dynamic loading was investigated with a RPA. The results show (Figure 3) that

as the amount of rubber phase in the samples increased, their storage modulus (G') decreased. This decrease was more pronounced for samples containing dGTR. The decrease in storage modulus is consistent with the change in modulus measured during static mechanical testing.

The behavior of the polymers depends on the relationship between the frequency of the load and the rate of molecular rearrangement. As the frequency of the stress increases, highly elastic deformation can develop less and less in the time available, so that after a certain limit, mechanical vitrification of the material occurs. Increasing the loading frequency increased storage modulus, and at the same time the loss factor ($\tan \delta$) decreased. The results show that the energy absorbed by the samples, that is their damping capacity, increased with rubber content. This improvement can be beneficial in certain applications (e.g. shoe soles).



FIGURE 3 The storage modulus (*G*') and loss factor $(\tan \delta)$ of samples containing (a, b) GTR and (c, d) dGTR. dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane. [Color figure can be viewed at wileyonlinelibrary.com]

3.3 | Falling weight impact testing

The behavior of the specimens under a dynamic load was investigated with a falling weight impact tester. In all cases, the dart penetrated the 2 mm thick injection molded sheets. The results (Figure 4, Table 5) show that increasing the amount of rubber phase in the blends reduces perforation energy. Figure 4 clearly shows that the maximum load and the displacement measured during the perforation of the specimen also changed accordingly.

At low rubber content, the load-displacement curves of the specimens indicated a tough character, and as the amount of the rubber phase increased, the shape of the material's fractogram indicates brittle behavior. This change may be due to the fact that crack propagation is much faster than the plastic deformation of the TPU matrix due to the weak connection between the two phases. This is also consistent with the tear strength (Table 3) of the samples.

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TABLE 5 Perforation energy of the samples.

Sample	Perforation energy (J mm ⁻¹)
TPU	64.1 ± 4.1
TPU/GTR _{90/10}	55.2 ± 3.3
TPU/GTR _{80/20}	46.7 ± 1.0
TPU/GTR _{70/30}	41.5 ± 0.9
TPU/GTR _{60/40}	34.1 ± 1.4
TPU/GTR _{50/50}	21.4 ± 0.7
TPU/dGTR _{90/10}	52.0 ± 1.7
TPU/dGTR _{80/20}	41.0 ± 1.4
TPU/dGTR _{70/30}	34.1 ± 0.9
TPU/dGTR _{60/40}	28.5 ± 1.0
TPU/dGTR _{50/50}	19.2 ± 0.7

Abbreviations: dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane.



FIGURE 4 Load-displacement curves of the samples: (a) TPU/GTR, (b) TPU/dGTR, and (c) perforation energy as a function of the amount of rubber. dGTR, devulcanized ground tire rubber; GTR, ground tire rubber; TPU, thermoplastic polyurethane. [Color figure can be viewed at wileyonlinelibrary.com]

3.4 | Morphology of the blends

The tensile-fractured surface of the samples was examined by SEM. The rubber phase caused the fragmentation of the surface of the samples (Figure 5). As the amount of the GTR in the blends was increased, there were more and larger particles in the images. In general, samples containing GTR have sharper grain boundaries than



TPU/GTR50/50

TPU/dGTR50/50

FIGURE 5 $100 \times$ magnification SEM images of the cross-section of the samples.

those containing d GTR. This can be explained by the fact that devulcanization typically takes place on the surface of the GTR grains, breaking the 3D cross-linked structure, making the grain surface more fragmented and therefore less sharp grain boundaries visible in the SEM image.^{37,38}

4 | CONCLUSION

The aim of our research was to produce TPEs in which some of the original TPU was replaced by recycled GTR. Above 10 wt% rubber content, the tensile strength and elongation at break of the samples were reduced. However, the mechanical properties (like tensile strength and elongation at break) of the samples somewhat improved due to the devulcanization of the GTR. The RPA tests show that the damping of the samples in the range between 0.1 and 10 Hz is increased by the rubber phase. Devulcanization further improved this capability. The falling weight impact test showed that the perforation energy of the samples decreased as a result of rubber content, which could be related to the weak coupling between the phases.

In summary, we have succeeded in producing a thermoplastic elastomer that contains a large amount of recycled rubber regrind to replace some of the TPU. This elastomer has a good price/performance ratio and better damping properties than TPU. In addition, this material can be used to 3D print realistic (smell and feel) rubberlike products.

AUTHOR CONTRIBUTIONS

Andrea Kohári: Conceptualization (equal); data curation (lead); formal analysis (lead); investigation (lead); methodology (equal); visualization (lead); writing – original draft (lead). **Tamás Bárány:** Conceptualization (equal); funding acquisition (lead); methodology (equal); project administration (lead); supervision (lead); writing – review and editing (lead).

ACKNOWLEDGMENTS

This research was funded by the National Research, Development and Innovation Office, Hungary (K 128268 and K 146085). Project no. TKP-6-6/PALY-2021 has been implemented with the support provided by the Ministry of Culture and Innovation of Hungary from the National Research, Development and Innovation Fund, financed under the TKP2021-NVA funding scheme.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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How to cite this article: A. Kohári, T. Bárány, *J. Appl. Polym. Sci.* **2024**, *141*(44), e56157. <u>https://doi.org/10.1002/app.56157</u>