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Improving the mechanical properties of vulcanizates containing ground tire rubber: Recipe optimization with the Taguchi method

Lóránt Kiss[1] | Márk János Molnár[1] | László Mészáros[1,2]

[1]Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Budapest, Hungary
[2]HUN-REN-BME Research Group for Composite Science and Technology, Budapest, Hungary

Correspondence
László Mészáros, Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3, H-1111, Budapest, Hungary.
Email: meszaros.laszlo@gpk.bme.hu

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Abstract
In this study, we optimized the recipe of ground tire rubber (GTR) containing natural rubber–based vulcanizates using the Taguchi method. We examined the effect of the amount of different components on the mechanical properties of natural rubber–based vulcanizates containing GTR. We determined the individual impact of each component on the tensile strength, elongation at break, modulus, tear strength, and hardness of the vulcanizates. However, when multiple components are changed simultaneously, understanding the underlying processes can be challenging. Nevertheless, using the Taguchi method, we were able to decide which component quantity we needed to modify to enhance a specific mechanical property. With the Taguchi method, we determined how and to what extent each component influences the tensile strength of vulcanizates in a sulfur-based vulcanization system. Finally, based on the results, we formulated a recipe to produce a vulcanizate with approximately 43% higher tensile strength compared to the reference.

KEYWORDS
ground tire rubber, natural rubber, recipe optimization, recycling, rubber waste, Taguchi method

1 | INTRODUCTION

Elastomers are cross-linked materials, which are capable of significant reversible deformation at room temperature, which makes them very popular. The greatest amount is used by the automotive industry for various purposes, such as vibration dampers, seals, and tires, with the latter constituting the majority of rubber usage.[1] This is reflected later in the waste stream: waste tires are a major part of elastomer waste.[2–4] These materials are difficult to recycle due to their cross-linked structure, making it impossible to process them with traditional mass production technologies (extrusion, injection molding).[5,6] Therefore, new solutions need to be developed for recycling.

Nowadays, waste tires are most commonly shredded into ground tire rubber (GTR), which is then used in a new matrix. The most common practice is to mix them into fresh rubber.[1,7–10] However, in most cases, there is poor compatibility between the matrix and the GTR in these new mixtures, resulting in materials that are not suitable for their intended applications.[5,11,12] To address this issue, researchers have developed several compatibility-improving procedures in the past, but these can often be costly (devulcanization)[13] and have a significant environmental impact (chemical treatments).[14] When the GTR is mixed into fresh rubber, optimizing the recipe presents a good, easier, and cost-effective way to adjust various mechanical parameters,[15] which is our goal in this paper.

Different rubber vulcanizates consist mostly of rubber and a vulcanizing system, which manufacturers use to achieve the desired mechanical properties of the product.[8,16] These vulcanizing systems are most commonly sulfur-based,[17,18] but they also contain many other important components, whose proportions and quantities are often not known.[19] Naturally, these factors depend on the specific properties required for the product. For instance, in applications where higher strength is needed, it is advisable to use a system that can form more cross-links, while in applications requiring significant deformation, this may not be the appropriate approach.[20]

A typical rubber recipe can consist of more than 10, or even over 20 components. They always include some form of vulcanizing agent (most commonly sulfur or peroxides), which is responsible for creating covalent bonds between the polymer chains.[21] Another crucial additive is accelerators, which are needed to increase the rate of cross-linking and can also enhance cross-link density. In practice, accelerator systems are used rather than a single compound, which has several advantages: it makes the process more controlled, reduces energy consumption, and allows for a higher cross-link density.[19]

Activators[22] can increase the number of cross-links that form with a given amount of sulfur. The most important activators are various metal oxides (zinc oxide, magnesium oxide) and stearic acid.[23,24] Antiaging agents are also important, as they make the final product more durable over time.[25,26] The use of various filler and reinforcing materials is essential, primarily for improving traction and reducing costs. However, nowadays, they can also influence the modulus, hardness, strength, and other properties of the rubber compound. The most common fillers used in the industry are carbon black, silica, and talc.[27] Of course, there are many other important additives as well (such as plasticizers and processing aids).[28]

Another major problem with rubber compounds containing GTR is that the GTR itself can alter the vulcanization process, thereby significantly influencing the properties of the final compound.[29] This is mostly because GTR contains various active groups on its surface, as well as unreacted additives (accelerators, activators, etc.), which may participate in vulcanization.[16] During the production of GTR, it undergoes degradation, during which sulfur bridges can break,[30] allowing the GTR to potentially withdraw sulfur from the system when used in a new mixture, thus altering the properties of the vulcanizate. However, it is difficult to predict precisely how GTR affects vulcanization and how it impacts the fresh rubber system. Exploring this highly complex process in detail is quite challenging.[5] However, it is possible to determine, using the Taguchi method, how to develop the formulation when we aim to improve specific mechanical properties.

As can be seen from the above, the preparation of rubber compounds requires multiple components, and these components typically influence more than one specific property. Interactions between the components are also important when there are a large number of components in rubber compounds. The introduction of GTR into the system further complicates the situation, as it can participate in vulcanization, hence general assumptions (e.g., sulfur and activators enhance strength) may not necessarily be dominant. It is very difficult to explore these processes (between the GTR and additives), but with the help of the Taguchi method (developed by Genichi Taguchi), it is possible to determine how the amount of different components should be changed when we wish to enhance a specific mechanical property in such a mixture.

The method is essentially based on fractional factorial experimental design, which is almost a standardized experimental design method. First, an appropriate orthogonal matrix needs to be selected, then the corresponding column–factor pairs need to be created according to specified rules. With this the degree of the effects of the individual factors can be determined, and, if applicable, also the degree of interaction between factors. Overall, the method may prove ideal for the development of recipes containing GTR in vulcanized compounds, because it is simple to apply and clear (especially when graphical representation is used). The application of the Taguchi method is justified and effective in cases where the optimization of the combined effects of multiple factors is the goal.[31–33] The purpose of this study is to examine the effects of individual components on the mechanical properties of natural rubber–based vulcanizates containing GTR and develop a formulation using the Taguchi method that has outstanding tensile strength.

2 | MATERIALS AND METHODS

2.1 | Materials

GTR with an average particle size of 0.2 mm from the tread and side wall of truck tires produced by waterjet milling, was kindly provided by Aquajet Ltd. (Budapest, Hungary). The composition of the GTR (Table 1) was determined by Simon et al.[34]

The matrix and other additives used for the preparation of the mixtures are summarized in Table 2.

2.2 | Recipes and vulcanization

For the optimization process, we used a general, natural rubber–based tire model recipe (NR_GTR_REF), the composition of which can be found in Table 3. In the table, from top to bottom, the order of materials also represents the mixing sequence.

The mixtures were prepared 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, while the fill factor was 0.8.

We recorded the vulcanization curves using a MonTech D-RPA 3000 moving die rheometer (MonTech Werkstofprüfmaschinen GmbH, Buchen, Germany) in order to determine the curing characteristics of the mixtures. The 20-min tests were conducted at a temperature of 160°C, with a 1° amplitude and a frequency of 1.67 Hz.

The compounds were vulcanized in a Teach-Line Platen Press 200E hydraulic press (Dr. Collin GmbH, Munich, Germany) at a

TABLE 1 The composition of the ground tire rubber used.[34]

Component	Amount (phr)
Natural rubber (NR)	50–55
Synthetic rubber	45–50
Carbon black (CB)	33–37
Residual additives	7.5
Oil	4–6

TABLE 2 Matrix and other additives used for the preparation of the mixtures.

Function	Material	Brand name
Matrix	Natural rubber	CV-60
Activators	Zinc oxide (ZnO)	ZnO WZ-1
	Stearic acid (StAc)	Radial 0444
Oil	Parraffin oil	Tudalen 3036
Accelerators	*N*-cyclohexyl-2-benzothiazol sulfenamide (CBS)	Rhenogran CBS-80
	Tetramethylthiuram disulfide (TMTD)	Rhenogran TMTD-70
Filler	Carbon black	N-772 OMSK
Cross-linking agent	Sulfur (S)	Curekind sulfur

TABLE 3 The composition of the initial and model recipe.

Component	Amount (phr)
NR	100
CB	60
ZnO	10
Stearic acid	2
GTR	100
Paraffin oil	10
CBS	1.25
TMTD	0.6
Sulfur	0.6

Abbreviations: CB, carbon black; CBS, *N*-cyclohexyl-2-benzothiazol sulfenamide; GTR, ground tire rubber; NR, natural rubber; TMTD, tetramethylthiuram disulfide; ZnO, zinc oxide.

temperature of 160°C. Pressure was 2.8 MPa and each mixture was pressed into 200 mm × 200 mm × 2 mm sheets. We maintained the pressure for the duration of the vulcanization time obtained from the rheometer measurements.

The mixing and vulcanization parameters were the same for each mixture; the only difference was in the recipes.

After producing the reference material according to the original recipe, we began the recipe optimization process with the goal of maximizing tensile strength. In the first step, we individually increased the quantity of each component by 50%, while the amount of the other components remained unchanged (Table 3). This was a relatively simple approach, and the results obtained here could serve as a good starting point for more complex systems in the future. It was also important to verify whether such changes in quantity had an impact on the mechanical properties of the vulcanizates. The abbreviation for the mixture containing 50% more stearic acid is NR_GTR_StAc*, and the one containing 50% more zinc oxide is NR_GTR_ZnO*, and so on for the other vulcanizates.

To further develop the recipe and understand how each component affects the properties, we used the Taguchi method. The essence of this method is to simultaneously change multiple variables (components) and examine their effects on the outcome (in our case, mechanical properties). During the investigation, we examined the influence of all components except the rubber matrix and GTR, in two stages. To do this, we applied an L8(2^7) orthogonal experimental design (Table 4). This means that we examined the effects of 7 different components in two different quantities, totaling 8 mixing scenarios. In the table, one represents the quantity of base recipe (Table 3), while two signifies a + 50% quantity.

Based on our results, we formulated the final recipe, which significantly enhances tensile strength. We will discuss this further in the results and discussion section.

2.3 | Testing of the vulcanizates

The mechanical characteristics of the vulcanizates were evaluated under room temperature with a Zwick-Z005 universal testing machine (Zwick GmbH., Ulm, Germany). Tensile properties were determined following the ISO 37:2017 standard, with the use of Type 1 specimens with a clamping distance of 60 mm and a crosshead speed of 500 mm/min. Tear strength tests were conducted in accordance with the ISO 34-1:2015 standard, utilizing Type C specimens with a test speed of 500 mm/min and a 56 mm clamping distance.

3 | RESULTS AND DISCUSSION

3.1 | Results of the first optimization process

Figure 1 shows the vulcanization curves of the mixtures for the first optimization process. The kinetics of the vulcanization curves is the same (similar shape). The mixture with increased oil content had the lowest maximum torque, and the increasing amount of accelerators (TMTD) and cross-linkers pushed the curves to a higher torque range compared to the reference, by increasing cross-link density. There is no significant difference in the minimum torque values, however the increasing amount of TMTD and sulfur accelerated the vulcanization process, whereby it was not affected by the other additives.

During the tensile test, we mostly obtained the expected results (Figure 2). The activators (StAc, ZnO) increased tensile strength to a small extent (Figure 2A), by increasing the cross-link density in the vulcanizates. In this recipe, it seems that ZnO helped in forming more

TABLE 4 The L8(2^7) orthogonal Taguchi experimental design we used.

Abbreviation	StAc	ZnO	Oil	CBS	TMTD	CB	Sulfur
NR_GTR_REF	1	1	1	1	1	1	1
NR_GTR_Tg1	1	1	1	2	2	2	2
NR_GTR_Tg2	1	2	2	1	1	2	2
NR_GTR_Tg3	1	2	2	2	2	1	1
NR_GTR_Tg4	2	1	2	1	2	1	2
NR_GTR_Tg5	2	1	2	2	1	2	1
NR_GTR_Tg6	2	2	1	1	2	2	1
NR_GTR_Tg7	2	2	1	2	1	1	2

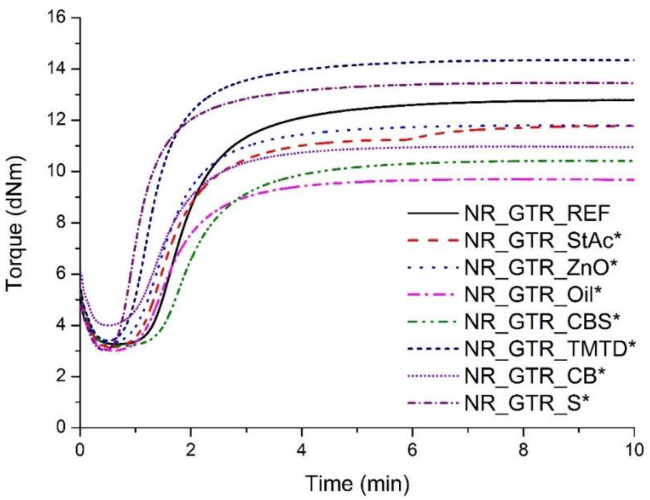

FIGURE 1 The vulcanization curves of the mixtures used in the first optimization process.

covalent bonds between the polymer chains, thus improving strength properties, in contrast to the other activator, stearic acid. Oil reduced tensile strength due to its softening effect, and the polymeric chains were able to move more freely, while accelerators (CBS, TMTD) enhanced strength properties because they can also increase cross-linking density in addition to accelerating the reaction. In this case, carbon black acted as a reinforcing filler, while sulfur had the greatest effect on the tensile strength through increased cross-link density, thereby making it the highest.

Elongation at break (Figure 2B) slightly increased but mostly remained in the same range as that of the reference (the differences not as big as in the tensile strength) except for the vulcanizates with increased CB content. Carbon black is a reinforcing filler, it improved tensile strength because it was present in large quantities. However, because of its high quantity it formed aggregates, which served as starting points for failure, thus reducing elongation at break. Elongation at break is over 300% in all cases, which is very good for NR-based vulcanizates containing GTR. This low variability indicates that the homogeneity of the mixtures was adequate in all cases. In most cases, we were able to increase tensile strength without reducing elongation at break, mainly due to the increased cross-link density in the vulcanizates.

The modulus values are expressed as M100, which is the stress at 100% elongation. Mixtures made with increased activator content were nearly identical to that of the reference (Figure 2C), as it was expected. In these cases, tensile strength did not increase significantly, therefore the resistance to deformation did not improve either. Carbon black significantly increased modulus because it is a very stiff reinforcing agent. The vulcanizates became stiffer, therefore the resistance to deformation became higher due to the increased amount of CB. Accelerators increased the modulus through higher cross-link density. The same mechanism was at work in the case of sulfur. The softener (paraffin oil) reduced the modulus. Due to its softening effect, the polymer chains “slid” more freely, thus reducing the resistance to deformation.

Figure 3 shows the results of the tear strength tests. The values changed slightly compared to the reference, except for mixtures made with increased TMTD, sulfur, and carbon black content. The ability of TMTD (activator) and sulfur (vulcanizing agent) to increase cross-linking density improves the connection between the matrix and the GTR; excellent compatibility between phases is important for great tear strength. The wide variability of filler content reduces the reliability of the results, but carbon black definitely functions as a reinforcing agent. During failure, crack propagation cannot continue without changing direction. The mechanism behind it is that due to the increased carbon black content, crack propagation often has to change direction, which increases the crack paths, resulting in a considerable improvement in tear strength.

The results indicate that a 50% increase in individual components had an impact on the examined mechanical properties. However, with this experimental design, it is challenging to provide clear explanations for the effects of the components on mechanical properties, and it is important to investigate how the components affect mechanical properties if more than one is increased.

3.2 | Results of the Taguchi optimization process

The samples presented in the previous section can provide a picture of the individual effects of the components on mechanical properties, but we have no information about the effect of changing the quantity of multiple components simultaneously. As the final step in recipe

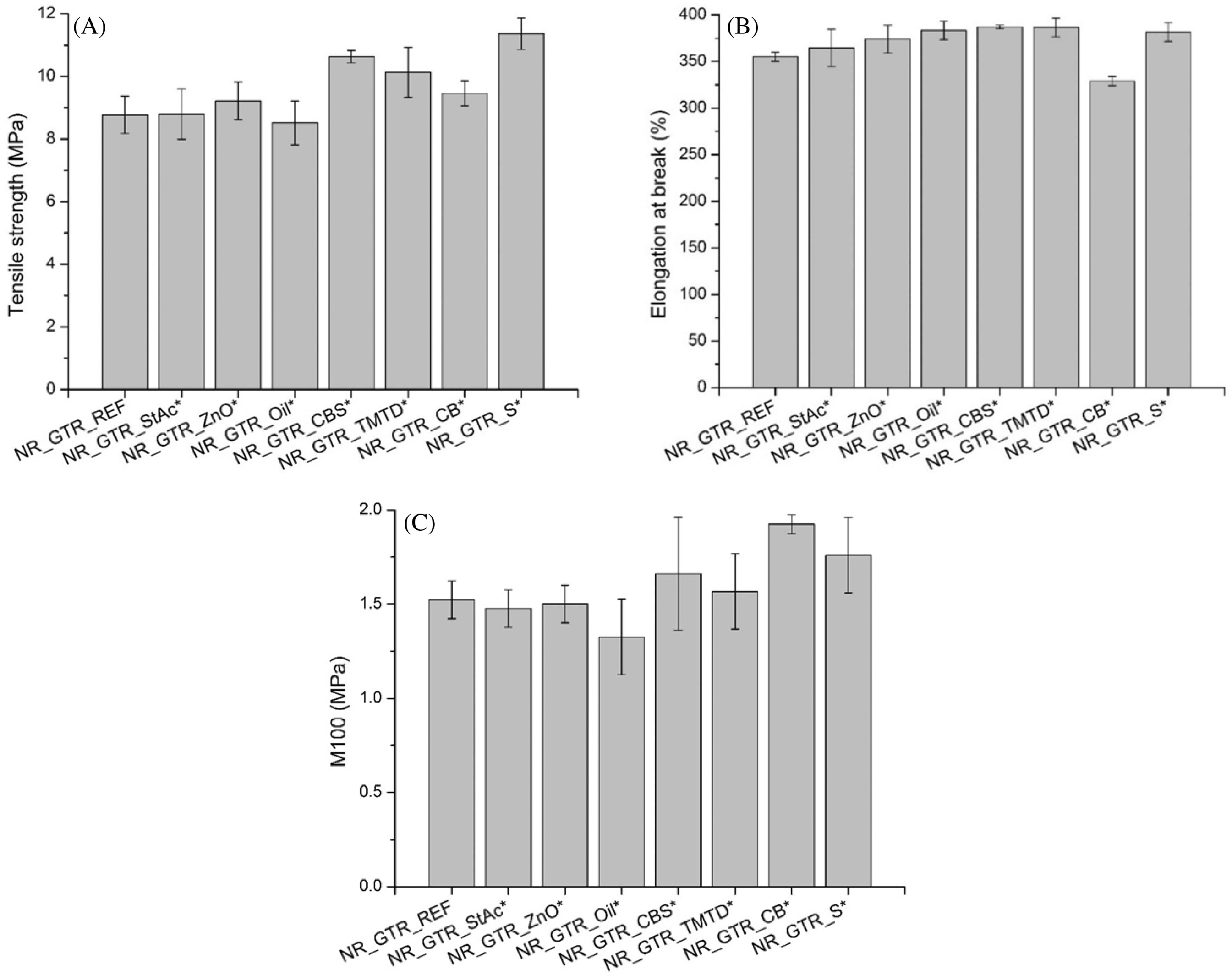

FIGURE 2 The (A) tensile strength, (B) elongation at break, and (C) M100 of the vulcanizates used in the first optimization process.

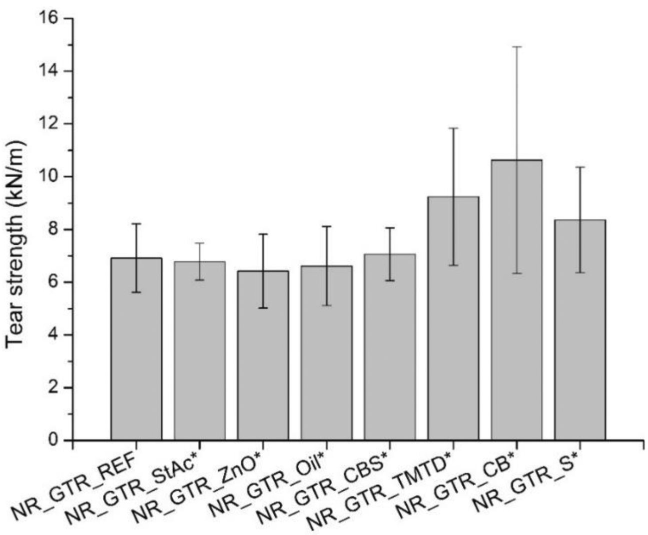

FIGURE 3 The tear strength of the mixtures used in the first optimization process.

development, we applied the Taguchi method. This method allows us to simultaneously change multiple components in a sample, making it easier to determine how specific mechanical properties (such as tensile strength in our case), vary and how we can achieve their highest value with a given system (a larger value is a better type of design). Unlike in the previous section, the following recipes involve changes to multiple components. The Taguchi method can explain how and to what extent the individual components affect the examined property (in our case, tensile strength). With this method, it is also difficult to provide clear materials science explanations due to the complexity of the system (especially with GTR), but we will try to explain the structural changes behind the changes in mechanical properties. However, the method can help determine how the quantity of individual components affects the investigated mechanical properties.

Figure 4 shows the vulcanization curves of the sample series. The results indicate that the kinetics of the curves have not changed here, either. However, NR_GTR_Tg1 (50% more TMTD, and sulfur) and NR_GTR_Tg6 (50% more TMTD and activators) increased the

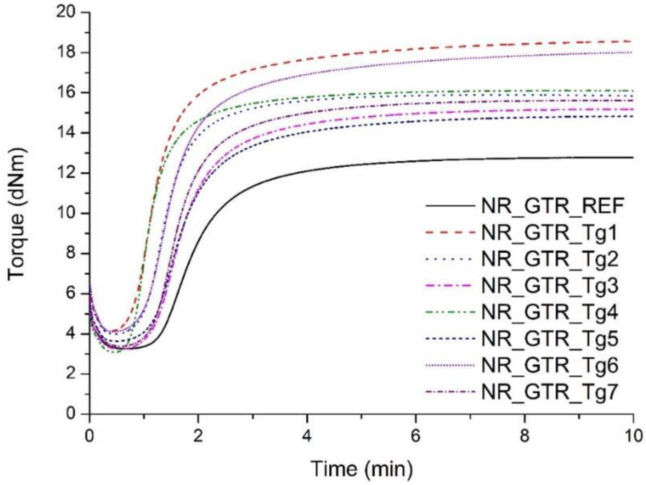

FIGURE 4 The vulcanization curves of the mixtures used for the Taguchi method.

maximum torque to the highest range due to the formation of more cross-links, which can be attributed to the effect of the vulcanizing agent, accelerator (TMTD), and activators. The vulcanization process became faster in these cases (steeper curves). The reference sample had the lowest maximum torque. In this case, fewer cross-links were formed compared to the other samples. There is no significant difference in the minimum torque of the mixtures.

The tensile strength (Figure 5A) of all samples increased compared to the reference. We demonstrated in the previous section that an increased amount of accelerators, sulfur, and activators (mostly ZnO), along with CB can increase tensile strength. If the amount of several components is increased, tensile strength increases even more.

NR_GTR_Tg3 had the highest tensile strength but Tg1, Tg4, and Tg6 are also very close and have improved strength. The tensile strength of Tg2 and Tg7 did not change significantly compared to the reference. The results show that the specimens with enhanced tensile strength contained increased amounts of TMTD, and the best specimen (Tg3) also contained an increased amount of the other accelerator (CBS). The Tg2 sample, although containing increased amounts of the vulcanizing agent, still failed to form a sufficient amount of cross-links. It did not contain increased amounts of accelerators, but it contained an increased amount of oil, which had a softening effect, thus reducing strength. Although the Tg7 sample contained increased amounts of CBS and sulfur, it still failed to increase strength significantly. Therefore, in this system, TMTD increased the number of cross-links in the vulcanizates, thus improving their strength, while the effect of the other accelerator or even the vulcanizing agent was not as significant.

Elongation at break (Figure 5B) mainly decreased or did not change compared to the reference, except for samples Tg3 and Tg4. These samples contained an increased amount of oil, which had a plasticizing effect. This made the polymer chains more flexible so they were able to deform more. In addition, and most importantly, they did not contain increased amounts of carbon black. Although CB greatly improves strength properties, it makes the vulcanizates brittle, which reduces elongation at break. Tg3 and Tg4 both had an increased amount of TMTD in the system so the number of cross-links increased even between phases, which resulted in good elongation at break, as they had a smaller amount of carbon black.

The modulus (Figure 5C) for samples 3, 4, and 7 did not change compared to the reference. These samples contained the same amount of carbon black as the reference. Carbon black is rigid and so it increases strength and makes the vulcanizates brittle, thus increasing their resistance to deformation. The modulus of mixtures containing less CB did not change compared to the reference. The other mixtures had 50% more CB, which caused the increase in M100. In the case of NR_GTR_Tg1, the excess sulfur with an increased amount of accelerators resulted in a higher cross-link density, which resulted in a greater increase of tensile strength (more than 70%) compared to the reference. It is interesting that the effect of the plasticizer (oil) for samples 1, 2, 3, and 4 was overcompensated in all cases by the excess amount of the other components. As a result of the additional amount of plasticizer, the modulus should have decreased. However, the results show that CB has the greatest effect on resistance to deformation (modulus).

Tear strength (Figure 6) increased in all cases compared to the reference sample, the most significant increase was with NR_GTR_Tg1. The change in recipe alone resulted in an increase of 170% in tear strength, which is an extremely good result. This sample contained the most accelerants (both TMTD and CBS), CB, and sulfur. The results so far show that TMTD can greatly increase cross-link density, which can help to create a better connection between the phases. Sulfur has the same effect, but it is not as potent without an increased amount of accelerators. Carbon black can increase the length of the crack propagation path. Since CB is a rigid filler, the cracks need to go around the particles, so the crack will spread over the interface of CB and the matrix, which also increases tear strength. Figure 3 shows that TMTD and carbon black alone can increase tear strength; presumably, they cause an even greater increase together with sulfur (more cross-links due to the increased amount of sulfur and accelerators).

We obtained the effects of the individual components on various mechanical properties using the Taguchi method. With this, we get a clearer understanding of how and to what extent the amount of each component affects mechanical properties. This analysis can be carried out for all the examined properties (tear strength, elongation at break, etc.), but our goal in this paper was to increase the tensile strength of the vulcanizates, so we are only going to examine this property further.

In Figure 7, positive slope indicates that tensile strength increases, and negative slope shows that tensile strength decreases. The value of the slope shows how much the component affects tensile strength (greater slope indicates greater effect). Stearic acid, oil, CBS, and TMTD have a positive impact on tensile strength (as indicated by the steepness of the curves), with TMTD having a particularly significant influence. With an excess amount of TMTD, mechanical properties improved greatly, because the vulcanizing agent formed more cross-links, even in the interphase. In such highly loaded systems (~30 w% GTR), it is mainly

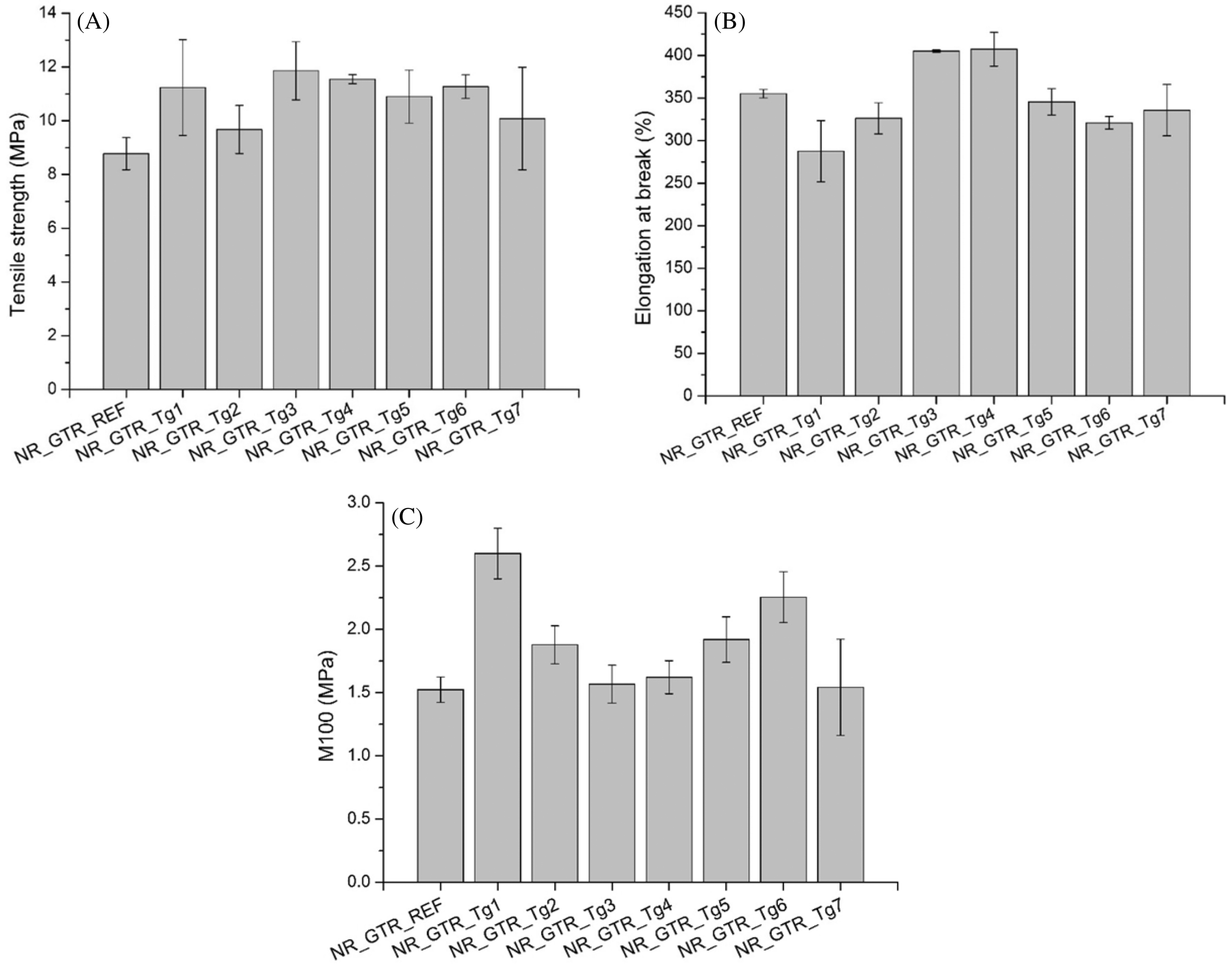

FIGURE 5 The (A) tensile strength, (B) elongation at break, and (C) M100 of the vulcanizates produced with the Taguchi method.

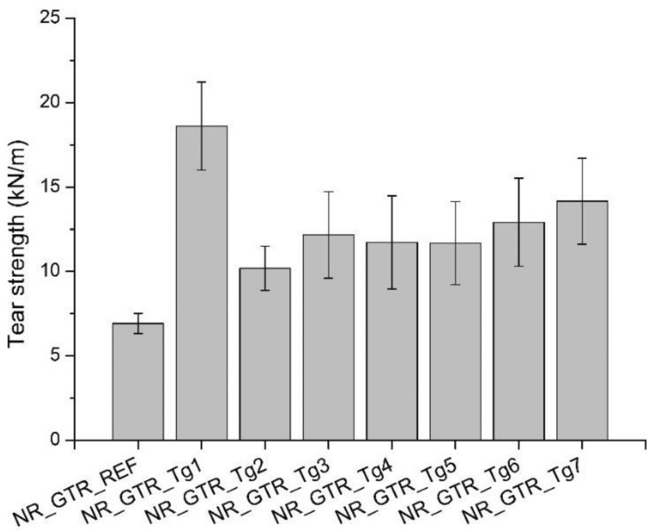

FIGURE 6 The tear strength of the mixtures used with the Taguchi method.

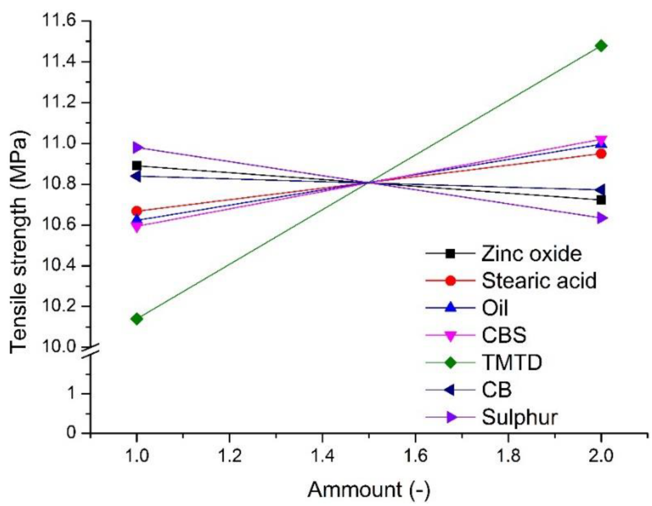

FIGURE 7 The effect of the components on tensile strength with the use of the Taguchi method.

the connection between the phases that mostly determines tensile strength (the weakest point), which TMTD improved due to more covalent bonds. On the other hand, an excess quantity of ZnO (it is a filler in higher contents), carbon black (very rigid filler), and sulfur (without enough accelerator, it cannot produce enough cross-links) has a negative impact on the tensile strength of the vulcanizates, but it is important to point out the slope of these is small, not as significant as the effect of TMTD.

TABLE 5 The final recipe, developed with the Taguchi method.

Component	Amount (phr)
NR	100
CB	60
ZnO	5
Stearic acid	3.5
GTR	100
Paraffin oil	17.5
CBS	2.18
TMTD	1.2
Sulfur	0.6

Abbreviations: CB, carbon black; CBS, *N*-cyclohexyl-2-benzothiazol sulfenamide; GTR, ground tire rubber; NR, natural rubber; TMTD, tetramethylthiuram disulfide; ZnO, zinc oxide.

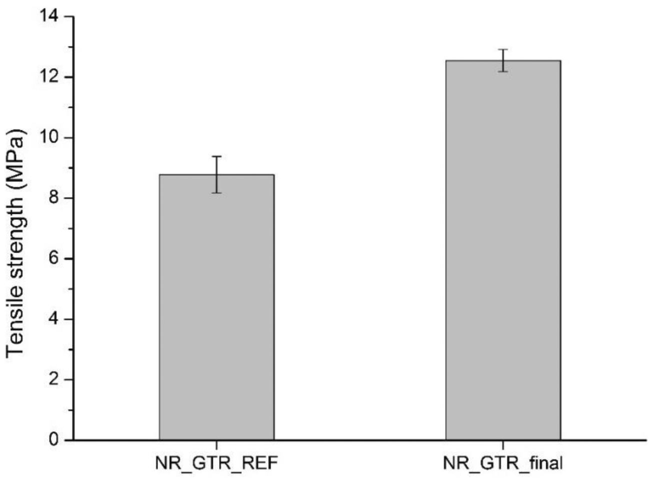

FIGURE 8 The tensile strength of the initial (NR_GTR_REF) and the developed vulcanizate (NR_GTR_final).

Based on the results, we developed the final recipe in order to achieve the highest tensile strength. Since TMTD had the most significant impact on tensile strength, we doubled its quantity, while stearic acid, oil, and CBS also had a positive effect on tensile strength, so we increased their quantities by 75%. We reduced the amount of ZnO by half because it can only increase tensile strength when used in smaller quantities. When too much is used, it acts as a weak filler, which can be the starting point of crack propagation. We did not change the quantities of the other components, as carbon black did not have a significant influence (small negative slope). Also, it can make the vulcanizates very rigid if too much is used. Sulfur is necessary for forming the appropriate amount of cross-links. The final recipe (NR_GTR_final) can be found in Table 5.

With this recipe, tensile strength was the highest in our experiments (Figure 8). The obtained results strongly support the anticipated final outcome based on the Taguchi matrix (meaning that tensile strength increases). Therefore, the Taguchi method is suitable for the development of rubber compound recipes and for gaining a better understanding of the effects of the components. The results are important because in rubber recycling, GTR is often blended into new matrices, but in these, the relationship between the phases is not adequate, which impairs mechanical properties and limits possible applications to a large extent. Different treatments are used to improve mechanical properties (devulcanization, chemical treatments, etc.), but we have shown that a relatively simple method improve and program the mechanical properties of vulcanizates, thus widening their range of applications.

Table 6 shows the tensile strength and the GTR content of the obtained materials and examples from the literature.

Zedler et al.[35] produced natural rubber–based vulcanizates containing 100 phr GTR using two different vulcanization systems. In the case of the sulfur-based system (NR/GTR^S), tensile strength was 8.0 MPa, whereas it was only 3.3 MPa when they used peroxide cross-linking

TABLE 6 Tensile strength of the obtained vulcanizates and some examples from the literature.

Abbreviation	GTR content	Tensile strength (MPa)	Reference
NR_GTR_REF	100	8.8 ± 0.6	This study
NR_GTR_final	100	13.4 ± 0.4	This study
NR/GTR^S	100	8.0 ± 0.4	35
NR/GTR^P	100	3.3 ± 0.7	35
NR/GTR^1	40	10.9	36
NR/GTR^2	50	10.5	36
NR/GTR^3	40	10.4	37
NR/GTR^4	50	9.1	37
NR/GTR^5	60	7.7	37

Abbreviations: GTR, ground tire rubber; NR, natural rubber.

(NR/GTR^P). The reference we produced (NR_GTR_REF) is close to the NR/GTR^S vulcanizate, but the new recipe formulated using the Taguchi method (NR_GTR_final) produced a tensile strength more than 67% higher.

Mandal et al.[36] produced natural rubber–based mixtures containing 40 and 50 phr GTR using a sulfur-based vulcanization system. The results show that these had higher tensile strength than our NR_GTR_REF sample since they produced mixtures containing less GTR. However, as a result of developing the recipe, we were able to increase tensile strength by 27% compared to the NR/GTR^1 sample, with double the amount of GTR.

Debapriya et al.[37] produced natural rubber–based vulcanizates containing 40, 50, and 60 phr GTR using a sulfur-based vulcanization process. The tensile strength of NR_GTR_final obtained with our improved formulation had double the tensile strength of materials containing less GTR, which is an excellent result. We successfully applied the Taguchi model to increase the tensile strength of vulcanizates containing GTR by recipe formulation.

4 | CONCLUSION

In this study, we examined the effect of the amount of different components on the mechanical properties (tensile strength, elongation at break, modulus, and tear strength) of natural rubber–based vulcanizates containing GTR. The samples were made with the use of a sulfur-based vulcanization system. First, we determined how individual components affect mechanical properties, with our primary goal being the improvement of tensile strength. Our results indicate that increasing the quantity of accelerators and sulfur (promoting more cross-linking) enhanced tensile strength, while the other components had a lesser impact, and the plasticizer (oil) reduced it. To assess the changes resulting from the simultaneous modification of multiple components, we utilized the Taguchi method. In this case, TMTD had the most significant impact on tensile strength (likely improving the bonding between the GTR and the matrix), and it also had a positive effect along with stearic acid, oil, and CBS. Using the results, we formulated a mixture that exhibited a 43% higher tensile strength compared to our reference material.

AUTHOR CONTRIBUTIONS

Lóránt Kiss: manuscript writing; experimental work; and results evaluation. **Márk János Molnár:** experimental work and results evaluation. **László Mészáros:** supervision; reviewing the manuscript; and experimental design.

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CONFLICT OF INTEREST STATEMENT

The authors have no conflict of interests related to this publication.

DATA AVAILABILITY STATEMENT

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

ORCID

Lóránt Kiss https://orcid.org/0000-0002-0255-4571

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