INVESTIGATION OF ELASTOMERS RATIO INFLUENCE IN THE COMPOSITES FOR TRUCK TIRES TREADS PRODUCTION

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ABSTRACT

An experimental design of model tread compounds for truck tires treads was carried out to test the possibilities for optimizing the ratio between the combination of elastomers (butadiene styrene rubber, isoprene rubber and butadiene rubber) used in their compositions. The experiment was designed using a Scheffe simplex lattice. The target functions, object of the optimization problem, are the main characteristics of the obtained vulcanizates: abrasion, stress at 100 % elongation, stress at 300 % elongation, tensile strength, elongation at break, residual elongation and Shore A hardness. The optimization was performed in the barycentric factor space with limits of variation of all factors from 0 to 1. Since the seven target functions obtain their extreme values at different points, a generalized criterion for optimality is formulated using the desirability function. The composition of the rubber compound at which this function receives its maximum has been established.

Keywords: truck tire treads, elastomers for tread compounds, compromise optimization.

INTRODUCTION

It is widely known that tire performance is justified by three main properties - rolling resistance, wet grip efficiency, and abrasion resistance, which is regarded as “magic triangle“ in the tire industry [1, 2]. Reducing rolling resistance of tire has gained much attention due to the increase demand for green transportation as tires are responsible for approximately 20 - 30 % of vehicle’s fuel consumption. Wet grip efficiency, or the handling ability of tire on wet road, is also of great importance as it is directly related to driving safety. Abrasion resistance is one of the key parameters to indicate the tire endurance. Many attempts have been made to investigate parameters affecting properties of tire tread compound with the goal of achieving improvements of rolling resistance, wet grip efficiency and abrasion resistance of tire [3 - 6].

On the other hand, it is known that pneumatic tire treads are in most cases produced based on blends of styrene butadiene rubber (SBR), butadiene rubber (BR) and natural (NR) or isoprene (IR), each of these rubbers having a strictly specific effect on the final performance of the pneumatic tire [7 - 12].

It has been reported that wear of a tire tread compound can be improved by the addition of BR, while the dynamic properties depend mainly on natural rubber [13]. Optimizing the ratio between the three types of rubber in the rubber compound used to make the tread is a basic approach to improving the performance of the tire and expanding the magic triangle. Several authors carry out this optimization with the help of experimental design [9, 14], as different experimental designs were used for data collection, described by Dick John [15].

The aim of the present work is to conduct an experimental design and to test the possibilities for optimizing the ratio between the three elastomers in the compositions for truck tires production treads by the method of simplex lattice, to obtain a final product with the most desirable performance. The main advantage of simplex lattice, respectively simplex lattice plans, is that they are particularly suitable for the study of multicomponent systems, whose properties depend on the ratio between the components, but not on the total amount of mixture in a given experiment. The proportions in which each component participates have "n + 1" evenly distributed values in the range from 0 to 1. In this way, the properties of a multicomponent system with each composition can be predicted [16, 17].

EXPERIMENTAL

Butadiene styrene rubber (Bulex 1500, manufactured in Bulgaria), isoprene rubber (SKI-3, manufactured in Russia) and butadiene rubber (SKD-1, manufactured in Russia) were used as starting elastomers for the preparation of tread mixtures. The main characteristics of the elastomers used are the following:

- "Bulex-1500" is a low-temperature unfilled butadiene styrene rubber with a content of 23.5 % bound styrene, Mooney viscosity ML (1 + 4) 100ºC- 46 - 58, with coloring stabilizer, for emulsifier resin-acid soap is used, and for coagulant - salt and acid;
- "SKI-3" is synthesized with the participation of a complex catalyst of Ziegler-Nata and contains: 92 ÷ 99 % cis-1,4-units, 0 - 4 % trans-1,4-units, 0 - 2 % 1,2 - units and 1 - 3 % 3,4-units. The Mooney viscosity ML (1 + 4) 100 ºC is 70 - 80, and its density is 0.92 g cm^{-3};
- "SKD-1" is a stereoregular rubber with a content of 1,4-cis units - 87 - 93 %, Mooney viscosity ML (1 + 4) 100ºC - 45. Titanium is used as a catalyst.

The experiment was designed using a Scheffe simplex lattice.

The main factors (variables) in the experimental plan are the three types of elastomers in the rubber compounds: X_1 - butadiene styrene rubber (SBR), X_2 - isoprene rubber (IR), X_3 - butadiene rubber (BR).

The target functions in the designed experiment are the main characteristics of the obtained vulcanizates, which were studied, namely: Y_1 - abrasion, Y_2 - stress at 100 % elongation, Y_3 - stress at 300 % elongation, Y_4 - tensile strength, Y_5 - relative elongation at break, Y_6 - residual elongation and Y_7 - Shore A hardness. The experimental plan, which contains 6 trials from the simplex lattice and 4 control points, is presented in Table 1.

According to the specified experimental design, 10 rubber compounds (model tread compounds for truck tires) were made based on butadiene styrene, isoprene, and butadiene rubber with the compositions as presented in Table 2.

The production of the rubber compounds is made on an open laboratory two rolls rubber mill, with dimensions: L/D 320x160 mm and friction 1.27. The speed of the slower roller is 25 min^{-1}.

The test specimens are vulcanized in the form of tiles with a thickness of 2 mm in a hydraulic press with electric heating at a temperature of 160°C and pressure of 12.0 MPa in the optimal vulcanization time for each compound, determined by the vulcanization isotherms according to ISO 6502-2: 2018. The vulcanization isotherms of the studied rubber compounds were taken on an oscillating disk vulcameter MDR 2000 (produced by "Alpha Technologies Inc., USA") at a temperature of 160°C.

The following characteristics of the obtained vulcanizates were studied:

Abrasion - by determining the reduction of the volume of a test body of vulcanizate under the influence of friction with a certain type of sandpaper (abrasive). Mass loss of the test piece determination and reduction calculation in volume by the density of the material were done using the formula:

$$\Delta V = \frac{m_0 - m}{\rho . b} .1000 \text{ , mm}^3$$

Table 1. Experimental plan.

No of the trial	X_1 parts	X_2 parts	X_3 parts
1	1	0	0
2	0	1	0
3	0	0	1
4	0.5	0.5	0
5	0.5	0	0.5
6	0	0.5	0.5
7	0.25	0.25	0.5
8	0.5	0.25	0.25
9	0.25	0.5	0.25
10	0.33	0.33	0.33

Table 2. Composition of the rubber compounds used (all ingredients are in phr (parts by weight per 100 parts by weight of rubber)).

Ingredients Compounds code	1	2	3	4	5	6	7	8	9	10
Butadiene styrene rubber	100	0	0	50	50	0	25	50	25	33.3
Isoprene rubber	0	100	0	50	0	50	25	25	50	33.3
Butadiene rubber	0	0	100	0	50	50	50	25	25	33.3
Paraffin	3	3	3	3	3	3	3	3	3	3
Stearic acid	2	2	2	2	2	2	2	2	2	2
Zinc oxide	5	5	5	5	5	5	5	5	5	5
poly(1,2-dihydro-2,2,4-trimethyl-quinoline) (TMQ)	5	5	5	5	5	5	5	5	5	5
Carbon black N220	50	50	50	50	50	50	50	50	50	50
Aromatic oil	15	15	15	15	15	15	15	15	15	15
N-Tertiarybutyl-2-benzothiazole sulfennamide (TBBS)	1	1	1	1	1	1	1	1	1	1
Diphenyl Guanidine (DPG)	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Sulphur	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5

where: ΔV - the reduction of the volume (abrasion) of the working body, mm^3;

m - mass of the working body after abrasion, g;

m_0 - mass of the working body before abrasion, g;

ρ - density of the working body, g/cm^3;

b = 1.1 - sandpaper coefficient.

Strength-elastic properties of vulcanizates in tensile deformation

After 24 hours of vulcanization, dumb-bells are cut from the test specimens using a punch press, which presses a standard-shaped knife onto the sample. The dimensions of the dumb-bells are measured with a micrometer with an accuracy of 0.01 mm. The determination of stresses at 100 and 300 % elongation (M_{100} and M_{300}), tensile strength and elongation at break is performed on a dynamometer at a movable jaw speed of 500 mm / min, according to ISO 37: 2017.

The determination of the residual elongation in tensile deformation is performed by laboratory methods. The measurement of the residual elongation in the working area of the samples shall be performed at least 1 min after rupture. The two parts of the torn blade are joined and the length of the working section is measured. From the experimental results, the residual elongation is calculated by the formula:

$$\varepsilon_{res.} = \{(\ell_2 - \ell_0) / \ell_0\}.100, \%$$

where: $\varepsilon_{res.}$ - residual elongation, %;

ℓ_0 - length of the working section before deformation, cm;

ℓ_2 - length of the working section after the rupture, cm.

Shore A hardness - The Shore A hardness of the test specimens was determined 24 hours after vulcanization using a portable hardness tester (produced by "Mitotoyo", Japan) according to ISO 48-4: 2018. The forced penetration into the material of a special nozzle under certain conditions is measured.

RESULTS AND DISCUSSION

The tread is the part of the outer tire that is in direct contact with the ground, which is why it is constantly subjected to wear by mechanical friction. Therefore, tread vulcanizates must have high tensile strength, high abrasion resistance (i.e., low abrasion), high elasticity, high tear resistance, expansion of cuts and cracks, and resistance to atmospheric ozone, light, and heat aging.

The results from the determination of the studied vulcanizates abrasion are presented in Table 3.

From the results presented in Table 3 it can be seen that the abrasion of the studied vulcanizates varies in the range from 34 to 87 mm^3. Best abrasion resistance, i.e. the lowest abrasion of all considered cases shows the vulcanizate with a content of 100 phr butadiene rubber (sample No 3). This is an expected result, as butadiene

Table 3. Abrasion of the studied vulcanizates.

Samples	Abrasion, mm^3
Compound No 1	87
Compound No 2	87
Compound No 3	34
Compound No 4	86
Compound No 5	55
Compound No 6	58
Compound No 7	66
Compound No 8	65
Compound No 9	74
Compound No 10	53

rubber has the highest abrasion resistance compared to butadiene styrene and isoprene rubber. Vulcanizates based on tread compounds No 5, 6, and 10 also show relatively good abrasion resistance. This is due to the butadiene rubber amount contained in them (50, 50 and 33.3 phr, respectively). Vulcanizates that do not contain butadiene rubber (1, 2 and 4) have the lowest abrasion resistance.

The results of the determination of the physico-mechanical parameters of the studied vulcanizates are presented in Table 4 and their values correspond to the results of other similar studies [18].

The results in Table 4 show that the highest values of stresses at 100 and 300 % of elongation and tensile strength have vulcanizates based on compound No 9 (25 phr SBR, 50 phr IR and 25 phr BR) and No 2 (100 phr IR). These vulcanizates have a relatively high elongation at break, but unfortunately compound No 2 also has the highest residual elongation. In terms of tensile strength, all tested samples show a value of this indicator above 18 MPa, except for vulcanizates based on a compound No 3 (100 phr BR) and a compound No 5 (50 phr SBR and 50 phr SBR), which is due to the absence of isoprene rubber in the elastomeric compound.

All studied vulcanizates have a relatively high elongation (over 520 %), with the highest values reaching this indicator for vulcanizates that contain only butadiene styrene rubber, isoprene rubber or a combination thereof. Vulcanizates based on butadiene rubber and the combination of butadiene styrene rubber and butadiene rubber have the lowest residual elongation, which is a result of the high elasticity given by these rubbers to the vulcanizates containing them. This is also confirmed by the fact that the highest residual elongation has the vulcanizate, which does not contain these rubbers. Shore A hardness for all studied vulcanizates has similar values, ranging from 60 to 64 rel. units. The maximum value of this indicator is in the vulcanizates based on a compound No 9, which correlates with the highest values for the stresses at 100 and 300 % of elongation, which shows this vulcanizate.

After the execution of the experimental design the software products “Minitab” and “Design Expert” were used for statistical processing of the obtained results. The basic statistical analysis is presented in Table 5.

The correlation analysis is presented in Table 6.

For all target functions, a statistical analysis was performed for the possible structure of the mathematical models and based on its results the best possible models were selected according to the available data. Table 7 presents the calculated values of the linear regression coefficients.

All coefficients are significant for which p-value ≤ the selected significance level alpha = 0.05.

The following mathematical models are obtained, giving the dependences of the target functions Y_1 -

Table 4. Physicomechanical parameters of the studied vulcanizates.

Code of the compounds Indicators	M_{100} MPa	M_{300} MPa	σ MPa	ε_{rel} %	ε_{res} %	Shore A rel. units
Compound No 1	1.6	5.7	18.1	632	16	63
Compound No 2	1.5	7.4	22.4	640	26	62
Compound No 3	1.6	6.0	13.1	520	5	62
Compound No 4	1.3	6.7	20.5	617	18	61
Compound No 5	1.4	6.6	16.3	550	8	61
Compound No 6	1.5	7.6	19.4	574	13	63
Compound No 7	1.4	6.0	18.6	602	10	60
Compound No 8	1.4	6.5	19.0	597	10	60
Compound No 9	1.6	8.7	22.5	579	13	64
Compound No10	1.5	7.1	18.9	573	12	62

Table 5. Basic statistical analysis of the obtained results.

Response	Name	Units	Observation count (N)	Min	Max	Mean	Std Dev
R1	Abrassion	mm^3	10	34.17	87.07	66.56	17.28
R2	M 100	MPa	10	1.34	1.62	1.48	0.10
R3	M 300	MPa	10	5.72	8.65	6.82	0.89
R4	Tensile strength	MPa	10	13.06	22.52	18.88	2.77
R5	Relative elongation	%	10	519.80	639.50	588.25	36.90
R6	Residual elongation	%	10	5.00	26.05	13.13	5.94
R7	Shore A Hardness	Rel. units	10	60.00	64.00	61.80	1.32

Table 6. Correlation analysis.

	Run	**A:SBR**	**B:IR**	**C:BR**	**Abrasion**	**M_{100}**	**M_{300}**	**Tensile strength**	**Relative elongation**	**Residual elongation**	**Shore A hardness**
Run	1.000	0.177	-0.088	-0.088	0.008	-0.382	-0.065	-0.097	-0.137	-0.095	-0.362
A:SBR	0.177	1.000	-0.500	-0.500	0.424	-0.212	-0.440	-0.042	0.375	0.004	-0.090
B:IR	-0.088	-0.500	1.000	-0.500	0.514	0.011	-0.648	0.814	0.524	0.811	0.180
C:BR	-0.088	-0.500	-0.500	1.000	-0.938	0.201	-0.208	-0.772	-0.899	-0.815	-0.090
Abrasion	0.088	0.424	0.514	-0.938	1.000	-0.121	0.156	0.760	0.938	0.836	0.115
M_{100}	-0.382	-0.212	0.011	0.201	-0.121	1.000	0.134	-0.121	-0.138	0.081	0.744
M_{300}	-0.065	-0.440	0.648	0.208	0.156	0.134	1.000	0.702	0.001	0.312	0.560
Tensile strength	-0.097	-0.042	0.814	0.772	0.760	-0.121	0.702	1.000	0.698	0.760	0.258
Relative elongation	-0.137	0.375	0.524	-0.899	0.938	-0.138	0.001	0.698	1.000	0.838	-0.056
Residual elongation	-0.095	0.004	0.811	-0.815	0.836	0.081	0.312	0.760	0.838	1.000	0.228
Shore A hardness	-0.362	-0.090	0.180	-0.090	0.115	0.744	0.560	0.258	-0.056	0.228	1.000

Table 7. Regression coefficients (p-value shading: $p < 0.05$ $0.05 \leq p < 0.1$ $p \geq 0.1$).

	A	B	C	AB	AC	BC	A^2BC	AB^2C	ABC^2
Abrasion	86.3178	87.4557	35.2963	-13.0352	-26.8957	-6.5717			
p-values				0.7258	0.4808	0.8587			
M_{100}	1.5905	1.5470	1.6193	**-0.93356**	**-0.8974**	-0.2604	1.0232	**15.9816**	-8.4072
p-values				**0.0264**	**0.0274**	0.0939	0.4537	**0.0352**	0.0667
M_{300}	5.7165	7.3913	5.9495	0.4012	**2.9388**	**3.6380**	**-41.0778**	**174.052**	**-133.436**
p-values				0.1278	**0.0177**	**0.0143**	**0.0289**	**0.0068**	**0.0089**
Tensile strength	18.6266	23.6986	14.3045						
p-values									
Relative elongation	617.831	629.621	517.298						
p-values									
Residual elongation	16.2166	26.0745	5.2484	**-15.8541**	**-14.0106**	-10.4628			
p-values				**0.0217**	**0.0320**	0.0734			
Shore A hardnes	62.9937	61.9937	61.9937	-6.0756	-6.0756	3.9244	-136.593	327.408	-184.592
p-values				0.2950	0.2950	0.4192	0.2992	0.1329	0.2289

Y_7 on the ratio of the three elastomers in the rubber compounds:

$y_1 = 86.3178x_1 + 87.4557x_2 + 35.2963x_3$

$y_2 = 1.59052x_1 + 1.54702x_2 + 1.61932x_3 - 0.933563x_1x_2 - 0.897363x_1x_3 + 15.9816x_1x_2^2x_3$

$y_3 = 5.71653x_1 + 7.39133x_2 + 5.94953x_3 + 2.93877x_1x_3 + 3.63797x_2x_3 - 41.0887x_1^2x_2x_3 + 174.052x_1x_2^2x_3 - 133.436x_1x_2x_3^2$

$y_4 = 18.6266x_1 + 23.6986x_2 + 14.3045x_3$

$y_5 = 617.831x_1 + 629.621x_2 + 517.298x_3$

$y_6 = 16.2166x_1 + 26.0745x_2 + 5.24844x_3 - 15.8541x_1x_2 - 14.0106x_1x_3$

$y_7 = 62.9937x_1 + 61.9937x_2 + 61.9937x_3$

The graphical images of the mathematical models are presented in Fig. 1 - 7.

Since the seven target functions receive their extreme values at different points in the barycentric

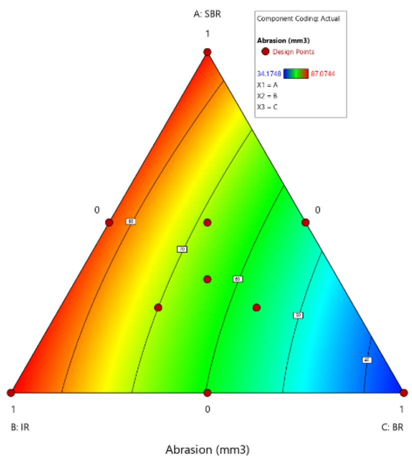

Fig. 1. Dependence of abrasion on the ratio of the three elastomers in the rubber compound.

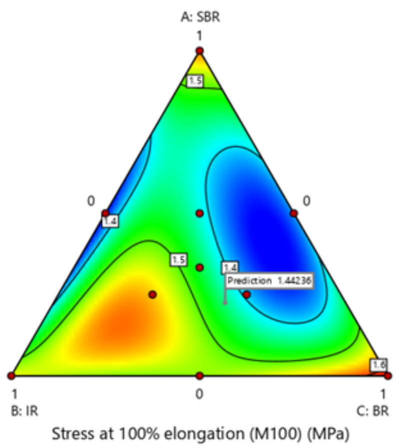

Fig. 2. Dependence of M100 on the ratio of the three elastomers in the rubber compound.

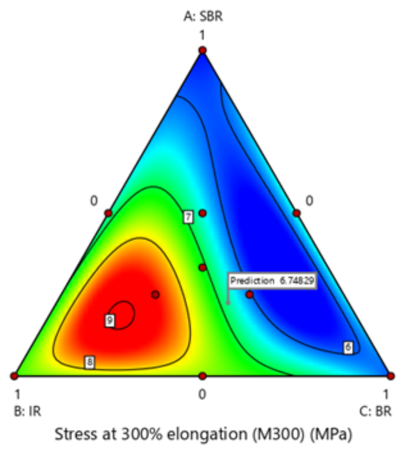

Fig. 3. Dependence of M300 on the ratio of the three elastomers in the rubber compound.

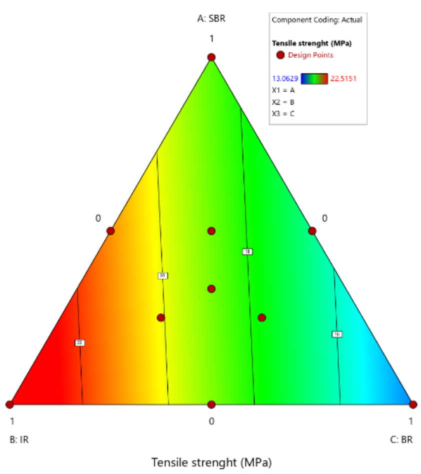

Fig. 4. Dependence of tensile strength on the ratio of the three elastomers in the rubber compound.

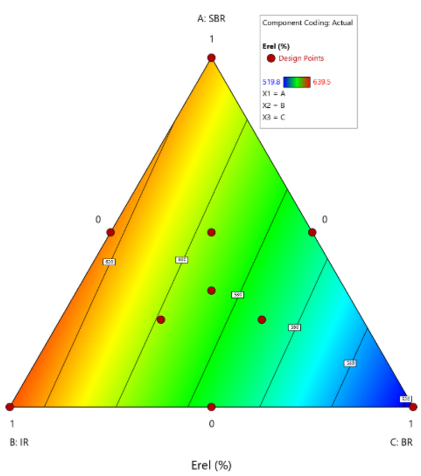

Fig. 5. Dependence of Erel on the ratio of the three elastomers in the rubber compound.

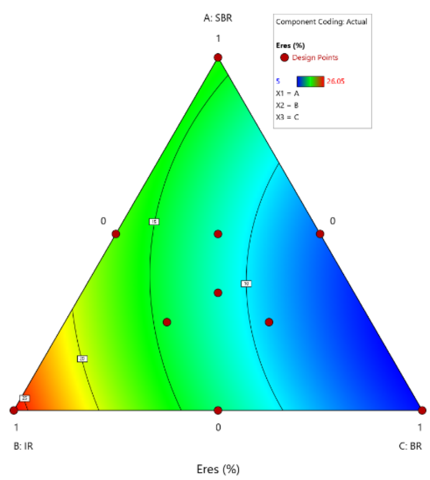

Fig. 6. Dependence of Eres on the ratio of the three elastomers in the rubber compound.

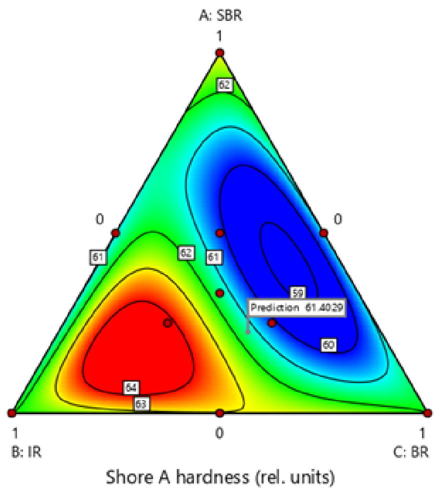

Fig. 7. Dependence of Shore A Hardness on the ratio of the three elastomers in the rubber compound.

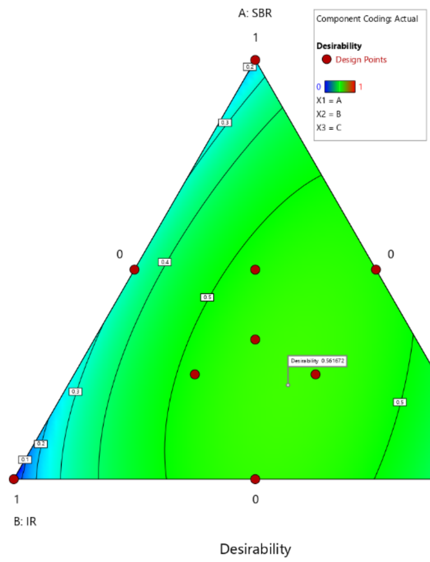

Fig. 8. Contours of the desirability function in the barycentric factor space in 2D-dimension.

space, it is necessary to formulate a generalized criterion for optimality. One of the most commonly used for this purpose is that of Harrington [19, 20], as a built-in function in "Design Expert".

The optimization was performed in the barycentric factor space with limits of variation of all factors from 0 to 1. The conditions and weight coefficients of the functions in the optimization task are defined in Table 8.

Four numerical solutions of the optimization problem are found, which are given in Table 9.

Graphical images of the desirability function in 2D and 3D dimensions are shown in Figs. 8 and 9.

As can be seen from the results presented in Table 9 and Figs. 8 - 9, the desirability function gets its maximum in the compound with the following composition: 22.5 phr butadiene styrene rubber, 32 phr isoprene rubber and

Table 8. Optimization conditions.

Name	Goal	Lower Limit	Upper Limit	Lower Weight	Upper Weight	Importance (5 - max, 1 - min)
A:SBR	is in range	0	1	-	-	-
B:IR	is in range	0	1	-	-	-
C:BR	is in range	0	1	-	-	-
Abrasion	minimize	34.1748	87.0744	1	1	5
Tensile strenght	maximize	13.0629	22.5151	1	1	4
Erel	maximize	519.8	639.5	1	1	3
Eres	minimize	5	26.05	1	1	3
Shore A hardness	none	60	64	1	1	1

Table 9. Solutions.

No	SBR	IR	BR	Abrasion	Tensile strenght	Erel	Eres	Shore A hardness	Desi
1	**0.225**	**0.320**	**0.455**	**58.791**	**18.280**	**575.799**	**10.276**	**61.403**	
2	0.000	0.469	0.531	58.128	18.711	569.990	12.413	62.971	
3	0.542	0.000	0.458	56.273	16.647	571.785	7.715	61.028	
4	0.541	0.459	0.000	83.603	20.953	623.239	16.802	61.026	

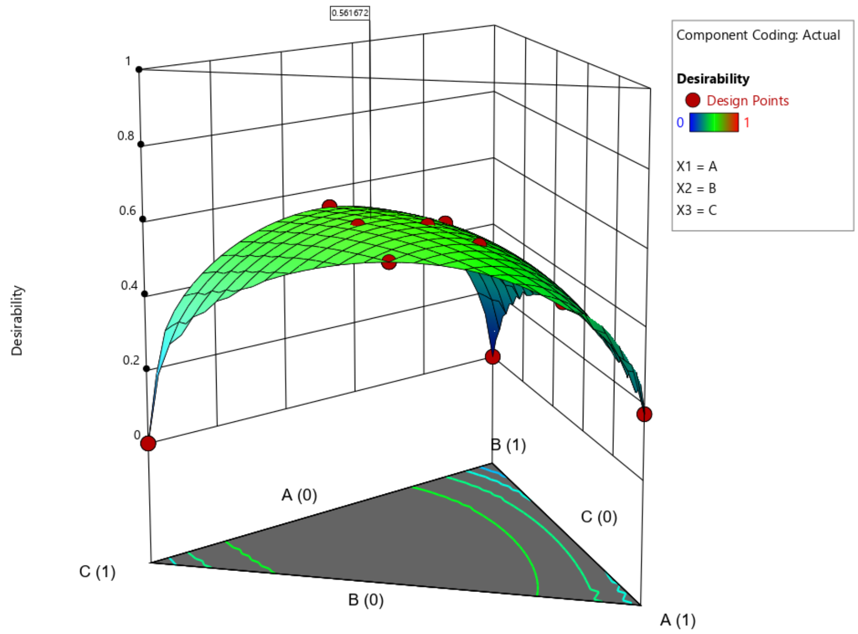

Fig. 9. Surface and contours of the desirability function in the barycentric factor space in 3D-dimension.

45.5 phr butadiene rubber.

The solution is optimally compromised, as the individual indicators receive their optimal values for different mixtures. For example, the abrasion has the lowest value for a mixture No 3 (Table 9), but the tensile strength is the furthest from its optimum. For a mixture No 4 the tensile strength and relative elongation have the highest values, but the abrasion and residual elongation are furthest from their optimum. In mixture No 2 the abrasion and tensile strength are comparable to those in mixture No 1, even slightly superior to them, but the relative elongation is lower and the residual elongation is higher.

CONCLUSIONS

An experimental study was conducted to verify the possibilities for optimizing the ratio between the three elastomers in the compositions of model tread compounds for truck tires by the method of simplex lattice. After the implementation of the experimental plan, statistical analysis was performed using the software products “Minitab” and “Design Expert” for the possible structure of the mathematical models. Based on its results, the best possible models of the seven target functions were selected, representing the dependence of the most important performance characteristics of the studied vulcanizates on the ratio of the three elastomers in their composition.

It was found that:

- The seven target functions receive their extreme values at different points of the barycentric factor space, due to which a generalized criterion for optimality is formulated.
- The desirability function receives its maximum in the compound with the following composition: 22.5 phr butadiene styrene rubber, 32 phr isoprene rubber and 45.5 phr butadiene rubber.

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