



Adhesion optimization of rubber compound on polyester cord to retain physico-mechanical properties

Hossein Roshanaei¹ · Henrik Margharian Pekachaki¹ · Fatemeh Khodkar¹

Received: 13 May 2019 / Accepted: 23 September 2019
© Iran Polymer and Petrochemical Institute 2019

Abstract

The adhesion of the rubber compound to polyester cords is an important parameter in tire production. The adhesion of the rubber compound and the mechanical properties of the rubber compound containing polyester cord could be decreased under environmental conditions. Hence, in this paper, the adhesion of common rubbers like natural rubber (NR)/styrene butadiene rubber (SBR) compounds to polyester cords was optimized by Box–Behnken design, while physico-mechanical properties were retained in desirable values. The effects of factors such as silica, resorcinol (as methylene acceptor), hexamethoxymethyl melamine formaldehyde (HMMM, as methylene donor), *N*-cyclohexyl-2-benzothiazole sulfenamide (CBS) and 2,2-dithiobis(benzothiazole) (MBTS) contents on adhesion and physico-mechanical properties were evaluated. The results have shown that the optimized values for each variable including silica, resorcinol, HMMM and CBS/MBTS were 5.76, 1.17, 1.45 and 0.81/0.19 phr, respectively. As HMMM used in this work includes 30% (wt) inert filler as the carrier, the HMMM/resorcinol ratio is near 1:1. In this formulation, the adhesion value of 15.7 kgf was obtained and tear strength reached 27.4 kgf/cm. The results showed that silica improved the adhesion because of longer time for the reaction of resorcinol with HMMM. To verify the optimized values for each variable, the formulation was again prepared and the results obtained from modeling data and experimental results showed the proper fitting of the modeling data with the experimental results.

Keywords Box–behnken · Physico-mechanical properties · Resorcinol · Silica · Styrene–butadiene rubber

Introduction

Using different cords and fibers as rubber reinforcement in different products, such as tires, hoses, belts, and diaphragms, is very common. The main determinant in performance characteristics of products is the strength of interfacial adhesion between the cord and rubber matrix [1]. In general, mechanical interlocking, electrostatic, diffusion, and adsorption/surface reaction are the main mechanisms of adhesion [2]. For rubber-cord adhesion, formation of the covalent bond or chemical adhesion is the most important mechanism. Therefore, the type of rubbers, additives, cords

and coating materials for cords are effective parameters in adhesion [3]. It is necessary to bond rubber compounds to synthetic fabric materials when they are used as reinforcing components for rubber products [4].

Rubber adhesion and mechanical properties of polyester cord could be decreased due to the hydrolysis and amination of esteric chains on the cord surface in percentage of the moisture and also amines inside the rubber compound [1].

To improve the adhesion between the rubber compound and the textile reinforcing material, the dry-bonding technology and the RFL (resorcinol formaldehyde latex) dip system are considered important and sometimes used simultaneously. The dry-bonding technology requires the direct addition of a methylene donor and a methylene acceptor into the rubber compound during the compounding process [5, 6]. Through the vulcanization process, the methylene donor cross links with the resorcinol to improve adhesion between the cords treated with the RFL and the rubber compound. Generally, the resorcinol resin (as methylene acceptor) and HMMM (hexamethoxy methyl melamine, as methylene donor) are the most common adhesion improving systems

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s13726-019-00753-z>) contains supplementary material, which is available to authorized users.

✉ Hossein Roshanaei
h.roshanaei@iranyasa.ir; roshanaei.hossein@gmail.com

¹ Department of Research and Development, Iran Yasa Tire and Rubber Company, P.O. Box: 3353154488, Tehran, Iran

in the carcass of radial tires. Aside from the adhesion improvement, the HMMM/resorcinol system also enhances the dynamic and mechanical properties of the rubber compound [7, 8]. In addition to methylene donor and methylene acceptor, sometimes an active white filler such as silica is added to the rubber compound. However, the filler role has been quite unknown yet, it can enhance the miscibility of the formaldehyde donor and resorcinol, or it can act as a catalyst [9]. Also, silica probably retards the vulcanization process, allowing time for the formaldehyde donor to react with resorcinol and the resin to migrate to the interface for bond formation [9–11].

The main ingredients, which can have marked effects on adhesion, are the curatives. Of the straight sulfur/accelerator systems, MBTS gives the highest adhesion levels. If the thiazole is activated, either internally as in the sulfenamides or with a secondary amine-based accelerator, such as DPG the level of adhesion is reduced. This amine-based activation has much less effect on adhesion than the faster thiuram or dithiocarbamate activation. Reducing the sulfur concentration reduces the adhesion levels. The activators, zinc oxide and stearic acid, do not significantly affect adhesion, other than by their effects on the overall curing efficiency of the conventional curing systems. The processed oils and plasticizers can adversely affect adhesion. Migration of the oils to the surface before laying up can have a deleterious effect. In service, migration of the oil or plasticizer into the dip film may occur, thereby reducing the cohesive strength of the dip film and resulting in premature breakdown of the adhesive [12].

As the number of compounding formulations is too high to optimize all effective parameters, the optimization method seems an efficient way that could be used. There are different optimization methods which are reported to optimize the properties of the rubber compound by Serafinska et al. [13], Li et al. [14], Dey et al. [15], Balachandran et al. [16–18], Shiva et al. [19], etc. Among them, RSM (response surface methodology) is a well-known method to optimize the intended properties [20].

RSM is a collective approach to experimental data based on the standard experiment plans; this would lead to a more uniform distribution of data within the range being studied. RSM for rubber compounding was extensively studied by Derringer [21]. RSM includes several methods: Box–Behnken design (BBD) and central composite design (CCD) are two main methods. The design results could be fitted to a second-order polynomial by a least-squares technique with a reduced number of experiments (three levels in BBD and three main levels with two more levels in CCD) [22]. The CCD method in rubber compounding was successively used by Sridhar et al. [23, 24], Kukreja et al. [25], Rajan et al. [26], Salvatori et al. [27], and other researchers. Among the RSM methods, Box–Behnken

design is less applied. In 2010, Balachandran et al. [18] used Box–Behnken design to optimize the formulations for optimal performance of the nanocomposites. At the same time, Ghasemi et al. illustrated the relationship between mixing parameters in internal mixer and properties of the styrene–butadiene rubber/organoclay composites using a Box–Behnken design [28].

The aim of this paper is optimization of the adhesion of NR/SBR compounds to polyester cords, while physico-mechanical properties were retained in desirable values. To gain this purpose, the effect of adding silica, resorcinol, HMMM and the loading value of the CBS accelerator on tensile strength, elongation-at-break, tear strength, hardness and abrasion behavior as well as the adhesion value of polyester cord to the compound (through *H*-pull test) was modeled by Box–Behnken method.

Experimental

Materials and preparation of compounds

Dipped PET tire cords (1500D2) were supplied by Saba Tire Cords, Iran. The natural rubber used was SMR20 grade supplied from Teh Ah Yau Rubber Factory. The styrene–butadiene rubber was SBR 1502 obtained from Bandar Imam Petrochemical Company, Iran. Zinc oxide (Pars Neko, Iran), stearic acid (PT. DuaKuda, Indonesia), high abrasion furnace (HAF) black (Iran Carbon, Iran), aromatic oil (Iranol Co., Iran), petroleum resin (Taizhou Huangyan Donghai Chemical Co., China) and kaolin (Narmkooban, Iran) were obtained. The curing system containing *N*-cyclohexyl-2-benzothiazole sulfenamide (CBS), 2,2-dithiobis (benzothiazole) (MBTS) and sulfur was supplied from Taizhou Huangyan Donghai Chemical Co., China. HMMM GLR 806 (Jiangsu Guoli Chemical Technology Co., China), resorcinol (Amino-Chem, China) and silica Vulcasil-S (Evonik Industries AG, Germany) were used as received.

Silica, resorcinol, HMMM, CBS and MBTS varied according to Table 1, while other ingredients were constant: SMR20 (54 phr), SBR1502 (46 phr), N660 (53 phr), kaolin (9 phr), stearic acid (1.6 phr), trimethyl-1,2-dihydroquinoline (TMQ) (1.1 phr), petroleum resin (2.7phr), aromatic oil (26 phr) and sulfur (2.6 phr).

HMMM, used in this formulation, included 30 wt% inert filler as a carrier. A resorcinol with 99.7% purity was used. The changes of MBTS and CBS accelerators were also considered as a variable in experiment design due to substitute in phr value used in the formulation.

The compounds were prepared in two stages. The non-productive stage was done in the internal mixer (Farrel) (at 70 rpm for 7 min) and the final step was performed on a two-roll mixing mill. Cure characteristics (i.e., minimum torque,

Table 1 Physico-mechanical properties of rubber compounds

No.	Variables				Physico-mechanical properties						Cure characterization				
	A		B		Tensile strength kgf/cm ²	Elongation- at-break %	Tear strength kgf/cm	Hardness Shore A	Abrasion mm ³	<i>H</i> test kgf	ML dNm	MH dNm	<i>t</i> ₂ min	<i>t</i> ₉₀ min	
	Silica	Resorcinol	HM/MM	CBS											MBTS
1	0.0	0.0	1.0	0.8	0.2	163.8	630	25.6	56	210	12.6	6	50	0.7	1.4
2	10.0	0.0	1.0	0.8	0.2	171.5	630	22.1	56	213	13.2	5	42	0.7	1.6
3	5.0	1.0	0.0	0.6	0.4	170.0	620	29.1	57	212	13.7	4	44	0.5	1.2
4	5.0	1.0	0.0	1.0	0.0	171.0	630	26.3	57	208	13.5	4.5	46	0.5	1.2
5	5.0	1.0	2.0	0.6	0.4	172.0	610	29.1	58	201	15.1	5	45	0.5	1.3
6	0.0	2.0	1.0	0.8	0.2	171.0	600	24.2	57	207	13.7	5	47.5	0.8	1.7
7	5.0	1.0	1.0	0.8	0.2	166.0	630	24.6	57	204	16.6	4	49	0.7	1.5
8	10.0	2.0	1.0	0.8	0.2	169.0	610	23.9	61	198	14.6	6	50	0.8	1.7
9	5.0	1.0	2.0	1.0	0.0	168.0	610	26.6	58	200	14.7	5	49	0.7	1.7
10	5.0	0.0	2.0	0.8	0.2	161.4	600	29.0	57	221	14.6	4	45	0.8	1.7
11	5.0	2.0	0.0	0.8	0.2	161.7	620	27.0	59	204	14.2	4	52	0.6	1.4
12	5.0	2.0	2.0	0.8	0.2	161.9	610	26.9	60	199	15.3	4	48.5	0.6	1.5
13	10.0	1.0	1.0	0.6	0.4	166.8	620	27.0	60	200	15.8	6.5	52.5	0.6	1.5
14	5.0	1.0	1.0	0.8	0.2	165.5	600	24.1	58	209	15.2	5	50	0.7	1.6
15	0.0	1.0	1.0	1.0	0.0	173.0	580	26.0	57	216	14.9	4	50	0.7	1.6
16	5.0	0.0	0.0	0.8	0.2	167.9	600	29.0	56	220	13.6	6	50.5	0.8	1.6
17	0.0	1.0	1.0	0.6	0.4	165.4	570	25.2	57	208	12.6	4	48	0.7	1.5
18	10.0	1.0	1.0	1.0	0.0	167.2	580	26.8	61	196	15.5	5.5	49	0.7	1.5
19	5.0	2.0	1.0	1.0	0.0	166.8	580	27.7	60	202	15.2	5.5	50.5	0.8	1.7
20	5.0	0.0	1.0	1.0	0.0	162.2	600	24.9	59	212	13.6	5.5	50.5	0.8	1.7
21	5.0	0.0	1.0	0.6	0.4	175.0	610	29.7	56	230	14.2	5	47.5	0.8	1.6
22	10.0	1.0	0.0	0.8	0.2	171.0	640	27.6	57	208	13.7	5	49	0.7	1.4
23	5.0	2.0	1.0	0.6	0.4	173.0	620	27.8	57	212	14.6	5	48	0.6	1.5
24	5.0	1.0	1.0	0.8	0.2	167.0	620	25.2	57	215	16.9	3	48.5	0.7	1.5
25	10.0	1.0	2.0	0.8	0.2	164.0	610	30.0	58	204	14.8	4.5	46.5	0.7	1.5
26	0.0	1.0	2.0	0.8	0.2	175.0	630	28.9	56	220	14.1	3.5	46	0.7	1.6
27	0.0	1.0	0.0	0.8	0.2	167.0	600	30.3	55	225	13.9	4.5	51	0.6	1.5

ML, maximum torque, MH, scorch time, t_{s2} , optimum cure time, t_{90}) were measured using a Monsanto ODR2000 rheometer (ASTM-D2084) at 195 °C. The compounds were vulcanized in a hot press at 160 °C for their respective optimum cure time.

Methods for characterization

To evaluate the tensile strength and tear strength, dumb-bell (ASTM D412) and die C (ASTM D624) specimens were punched out from the molded sheets, respectively. These mechanical properties were obtained by a Universal Testing Machine (Instron 1026, High Wycombe, UK). The mean of three values was recorded.

The hardness of the samples was measured by Durometer (Bareiss, Germany) according to ASTM D2240.

H-pull (*H*-adhesion) test method is a static and relatively simple method to evaluate the adhesion. The *H* tests were designed to measure the force required to pull a cord in the direction of its axis from a strip of rubber (shaped like an “H”) in which the ends of the cords are embedded as per the ASTM D2138 [29].

Experimental design

Box–Behnken is an incomplete three-level factorial design which was introduced to reduce the sample numbers as the parameters increase. A Box–Behnken design was selected for considering its efficiency in the number of required runs.

The Box–Behnken design is characterized by a set of points lying at the midpoint of each edge of a multi-dimensional cube and center point replicates. This design does not contain any points at the vertices. Hence, it is very useful in situations where physical constraints on the upper and lower limits of the variables make it impossible to test these

points. It requires three levels of each factor and hence permits the response to be modeled by second-order behavior. The main, quadratic and interactive effects of the factors on the properties under study can be estimated from the quadratic equation.

The Box–Behnken design with three levels and four factors was applied in this research with silica content (x_1), resorcinol (x_2), HMMM (x_3) and CBS accelerator (x_4) as the independent variables. The compound was optimized for cord adhesion property.

The experimental data obtained from the above procedures were analyzed by the response surface regression method using the following second-order polynomial equation [16]:

$$y = \beta_0 + \sum_{i=1}^4 \beta_i x_i + \sum_{i=1}^4 \beta_{ii} x_i^2 + \sum_{i < j=1}^4 \sum_{j=1}^4 \beta_{ij} x_i x_j \quad (1)$$

where, y is the response, x_i and x_j are the coded independent variables, and β_0 , β_i , β_{ii} , and β_{ij} are the mean values of responses, linear, quadratic and interaction constant coefficients, respectively. MINITAB 17 software was used for the regression analysis. According to this design, 27 formulations were obtained in total.

Results and discussion

All formulations according to experimental design, along with their properties (by ASTM methods) are shown in Table 1.

The effectiveness of each parameter could be characterized by *F* value or *P* value. The high *F* value or low *P* value displays the highest effect. As observed in Table 2,

Table 2 Results of *F* value and *P* value for each factor

	Elongation-at-break		Tear strength		Hardness		Abrasion		<i>H</i> test	
	<i>F</i> value	<i>P</i> value	<i>F</i> value	<i>P</i> value	<i>F</i> value	<i>P</i> value	<i>F</i> value	<i>P</i> value	<i>F</i> value	<i>P</i> value
Silica	3.84	0.066	–	–	28.45	0	14.91	0.001	6.98	0.017
Resorcinol	–	–	0.48	0.496	24.79	0	23.43	0	6.98	0.017
HMMM	0.96	0.34	0.09	0.769	4.55	0.047	3.4	0.08	7.47	0.014
CBS	2.94	0.104	5.67	0.029	6.2	0.023	2.79	0.11	0.41	0.532
Silica*silica	–	–	–	–	–	–	–	–	18.46	0
Resorcinol*resorcinol	–	–	–	–	–	–	–	–	18.46	0
HMMM*HMMM	–	–	39.83	0	–	–	–	–	12.09	0.003
CBS*CBS	7.9	0.012	6.85	0.017	5.69	0.028	–	–	6.12	0.024
Silica*resorcinol	–	–	–	–	6.07	0.024	–	–	–	–
Silica*HMMM	6.47	0.02	–	–	–	–	–	–	–	–
Silica*CBS	4.5	0.048	–	–	–	–	–	–	4.21	0.056
Resorcinol*CBS	–	–	4.07	0.059	–	–	–	–	–	–

for hardness and abrasion, silica and resorcinol have more effect, while HMMM and CBS have less effect. Additionally, silica and CBS have more effect on elongation and tear strength, respectively. Since the adhesion is evaluated as the main factor, HMMM has the most impact and CBS has the lowest one.

Main effect plots

Main effect plots show the effect of the factors (silica, resorcinol, HMMM and CBS loading values) on the responses and the slope of their line indicates the intensity of the effects. A horizontal line with a slope of 0 displays the absence of the main effect. The more slope of the main effect plot exhibits a stronger effect. Supplementary 1 presented only the main effect plots which modeled in Table 3. It can be seen that by increasing each factor, hardness increases, while abrasion decreases. Reportedly, the abrasion resistance and hardness could be improved by adding reinforcement resins to rubber compound due to their reaction with methylene donor during the vulcanization of the rubber compound [30].

By adding silica, adhesion increased. As a coupling agent was not used in this work, silica probably adsorbed basic accelerators and retarded the vulcanization process. Hence, it allowed more time for the formaldehyde donor reaction with resorcinol and migration of the resin to the interface for bond formation. Also, it reported that the silanol on silica surface could react with soluble zinc and influence on the low cure states [31]. Therefore, the elongation-at-break increased with silica addition. Furthermore, the

elongation-at-break was reduced with HMMM content and showed an optimum value with CBS amount. In the same way, tear strength displayed reduction by increasing resorcinol and down-up behavior for HMMM and CBS contents.

Interaction plots

Interaction plots exhibit the interaction effect of factors on the responses. If the change in the mean of the response from one level of a factor to another level depends on the level of another factor, the two factors are said to have interaction effects. Parallel lines display the absent interaction between the factors. The more deviation from parallel shows the more interaction between the factors [16, 18]. Interaction plots modeled in Table 4 are illustrated in Fig. 1. As observed, the interaction between silica and HMMM for elongation-at-break; resorcinol and CBS for tear strength; silica and resorcinol for hardness; and silica and CBS for *H*-pull test are significant. The last one has again proved that the silica could retard the vulcanization process by adsorbing the activators and accelerators (ZnO and CBS) due to its polarity; therefore, the curing process can prolong and the time for reaction of formaldehyde with resorcinol and migration of the resin to the interface takes place for bond formation. Therefore, the adhesion could be improved using silica.

Contour plots

The two-dimensional contour plot is a series of curves of constant response for different combinations of factor levels. Such plots display the change in properties when two or

Table 3 Modeling of physico-mechanical properties of tread compounds

Physico-mechanical properties	Uncoded regression equation (full second degree)	P value	R2-adj	R2-sq
Elongation-at-break (%)	$Elongation = 365.0 + 14.33 A + 11.67 C + 547 - 321 D^*D - 3.00 A^*C - 12.50 A^*D$	0.002	56.44	69.84
Tear strength (kgf/cm)	$Tear = 55.22 - 4.93 B - 5.708 C - 58.0 D + 2.904 C^*C + 30.1 D^*D + 5.88 B^*D$	0	70.28	79.42
Hardness (shore A)	$Hardness = 65.08 + 0.0500 A + 0.167 B + 0.500 C - 27.1 D + 18.75 D^*D + 0.200 A^*B$	0	74.8	82.56
Abrasion (mm ³)	$Abrasion = 234.32 - 1.117 A - 7.00 B - 2.67 C - 12.08 D$	0	66.62	74.32
H test (kgf)	$H \text{ test} = -2.48 + 1.088 A + 2.842 B + 2.408 C + 31.0 D - 0.0472 A^*A - 1.179 B^*B - 0.954 C^*C - 16.98 D^*D - 0.650 A^*D$	0.001	64.31	76.66

Table 4 Comparison of empirical and predicted values

Variables (phr)					Compound	Physico-mechanical properties				
A	B	C	D			Elongation-at-break	Tear strength	Hardness	Abrasion	H test
Silica	Resorcinol	HMMM	CBS	MBTS		%	kgf/cm	Shore A	mm ³	kgf
5.76	1.17	1.45	0.81	0.19	Predicted	614	25.6	58	206	16.4
					EMPIRICAL	590	27.4	57	201	15.7
					Percent change	-3.90	7.03	-1.72	-2.43	-4.27

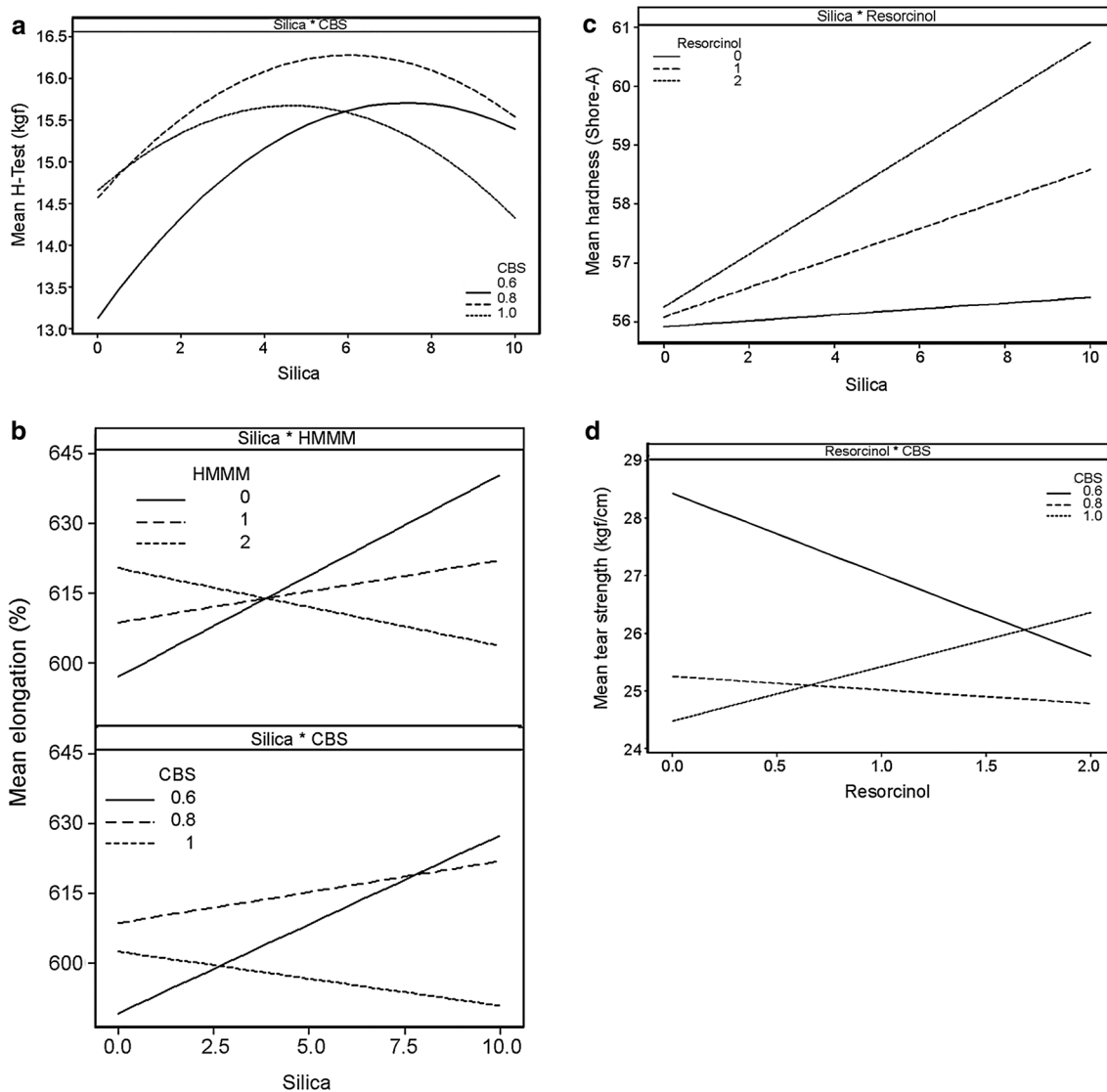


Fig. 1 Interaction plots for: **a** *H* test, **b** elongation, **c** hardness, and **d** tear strength

more variables vary together. They also allow predictions to be taken for combinations not actually being done in the experiment. Analysis of the contour plot for elongation-at-break, abrasion, hardness, tear strength and *H*-pull test is shown in Figs. 2, 3, 4, 5, 6, respectively. Elongation-at-break (Fig. 2) increases with more silica as well as less HMMM and CBS. Indeed, silica without a coupling agent shows a low curing state and reduced cross-linking; therefore, elongation-at-break is increased with silica addition.

As illustrated in Fig. 3, abrasion is reduced by adding more additives, while it is clear that the hardness shows the inverse trend (Fig. 4). As mentioned, resorcinol and HMMM react with each other during the vulcanization process and improve hardness and abrasion resistance. When resorcinol reacts with HMMM, the cross-linked resin is formed. This network increases the hardness of the compound and

reduces the movement of the polymer chains. So by reduced chain motion and inter-chain friction, eventually the abrasion is reduced or abrasion resistance is improved. Low tear strength and high *H*-pull test values were exhibited in the center of contour plots, as shown in Figs. 5 and 6, respectively.

Optimization of formulation

The physico-mechanical properties of compounds are considered as criteria for the optimization process. If in the variance analysis table, the *P* value is less than 0.05 (95% confidence level), then it is significant. Therefore, due to the high *P* value ($P > 0.05$) for tensile strength, this feature is eliminated from the modeling and optimization process. The equations presented in Table 3, provided by the Minitab

Fig. 2 Contour plots of elongation-at-break

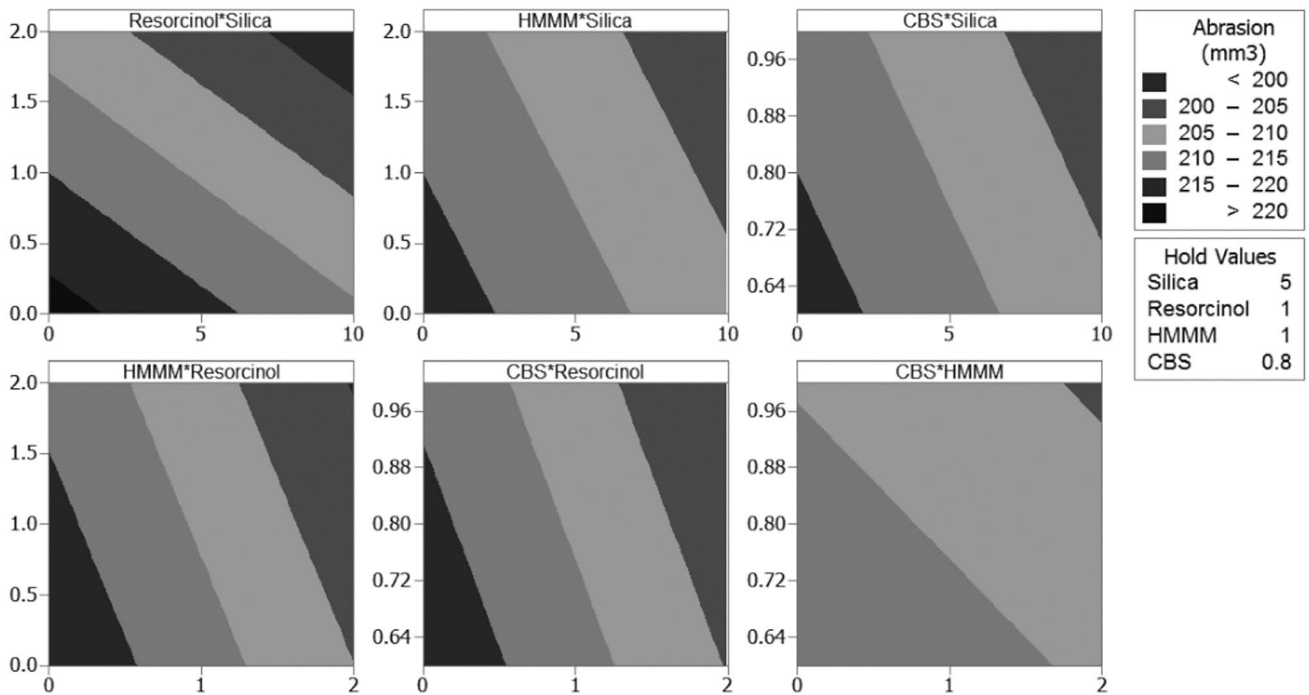
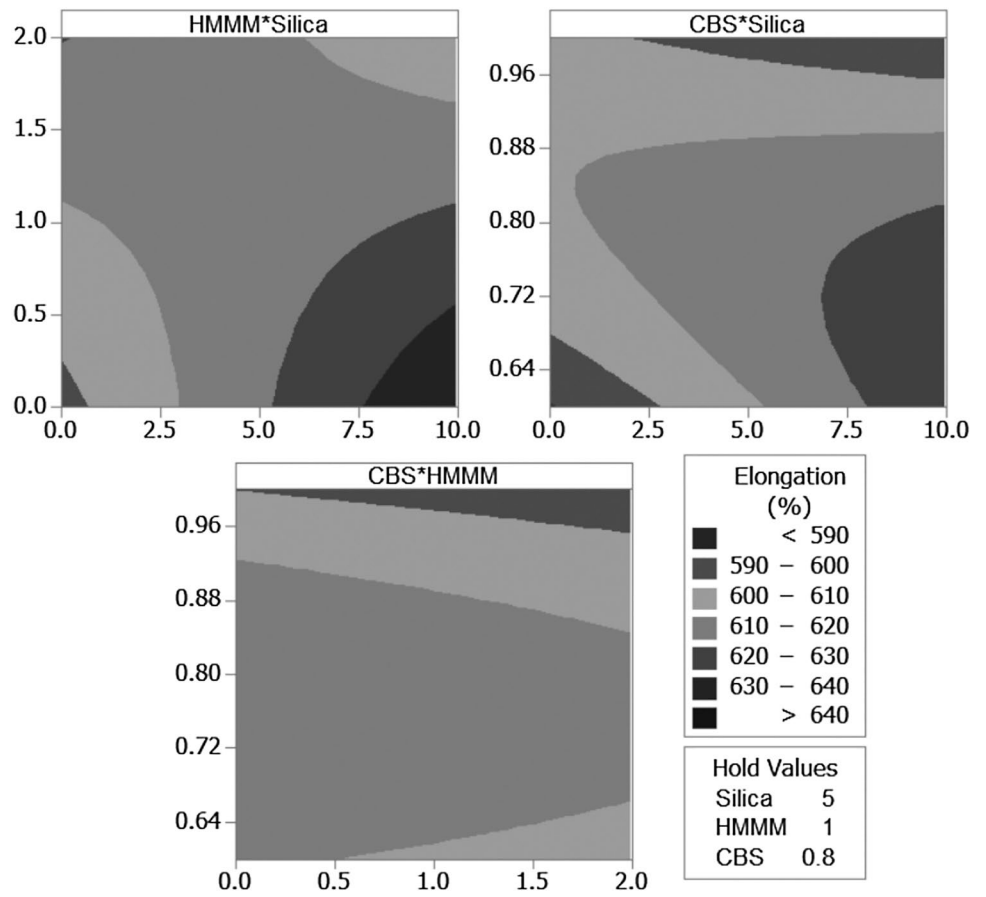


Fig. 3 Contour plots of abrasion

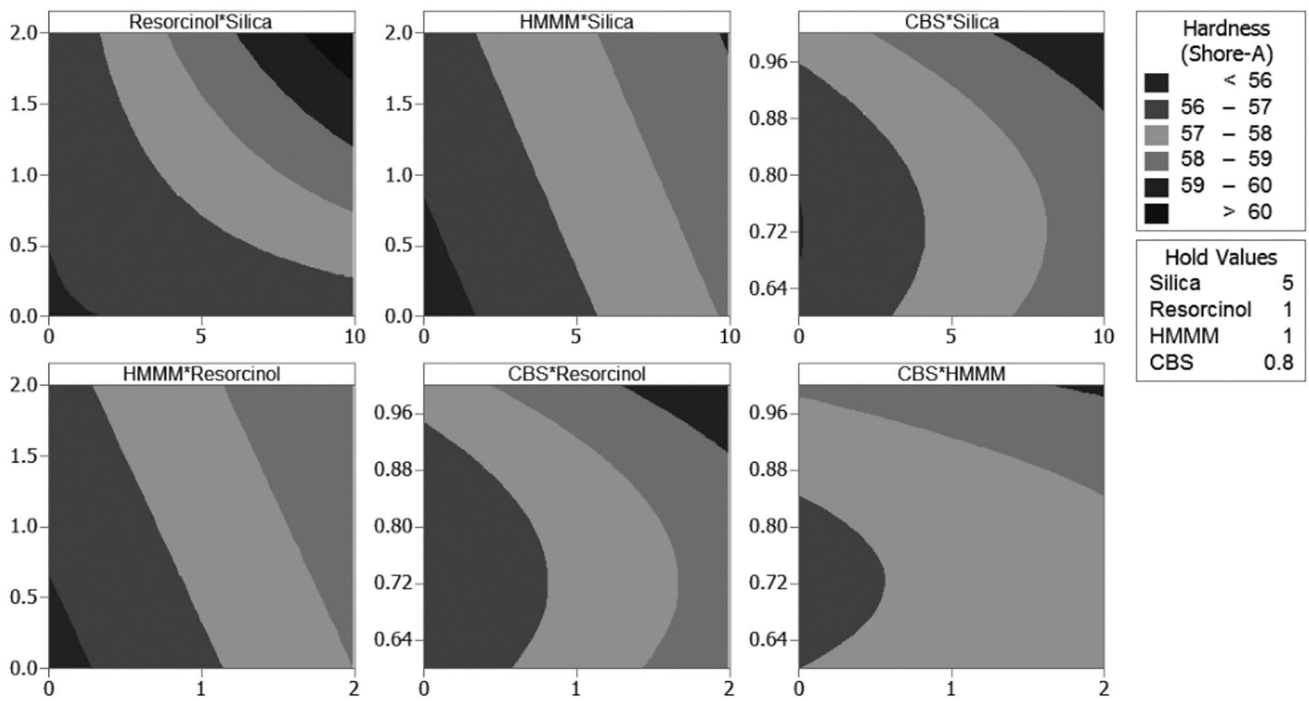
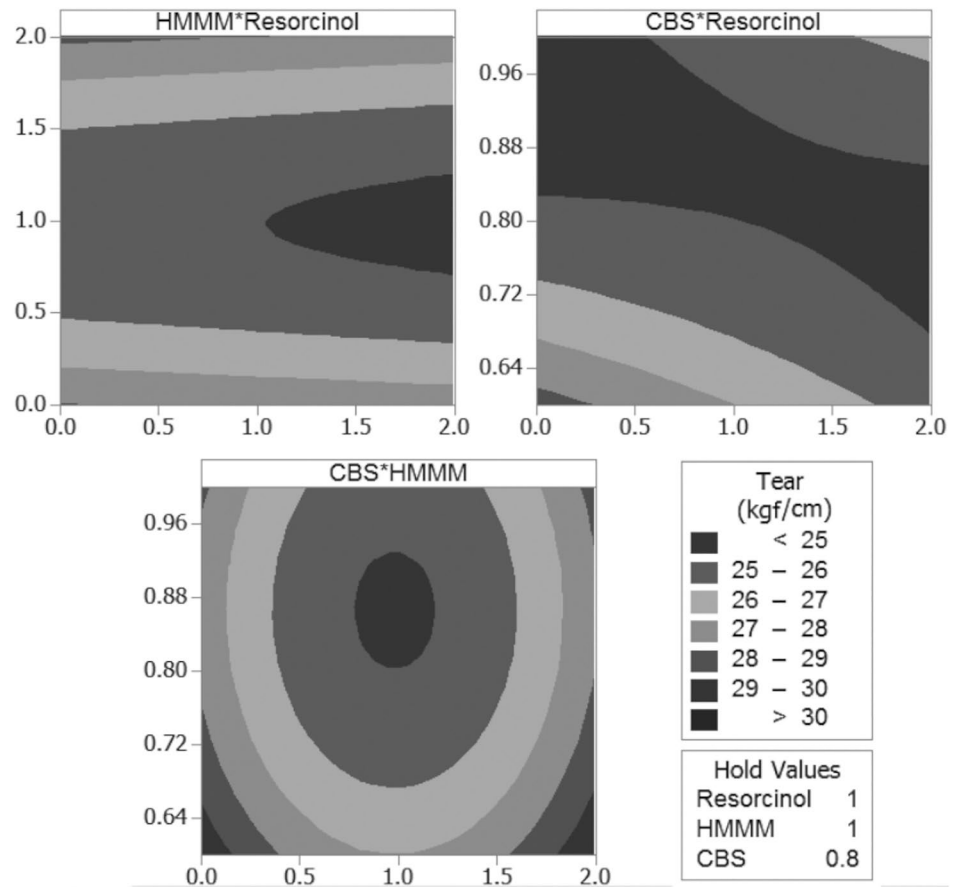


Fig. 4 Contour plots of hardness

Fig. 5 Contour plots of tear strength



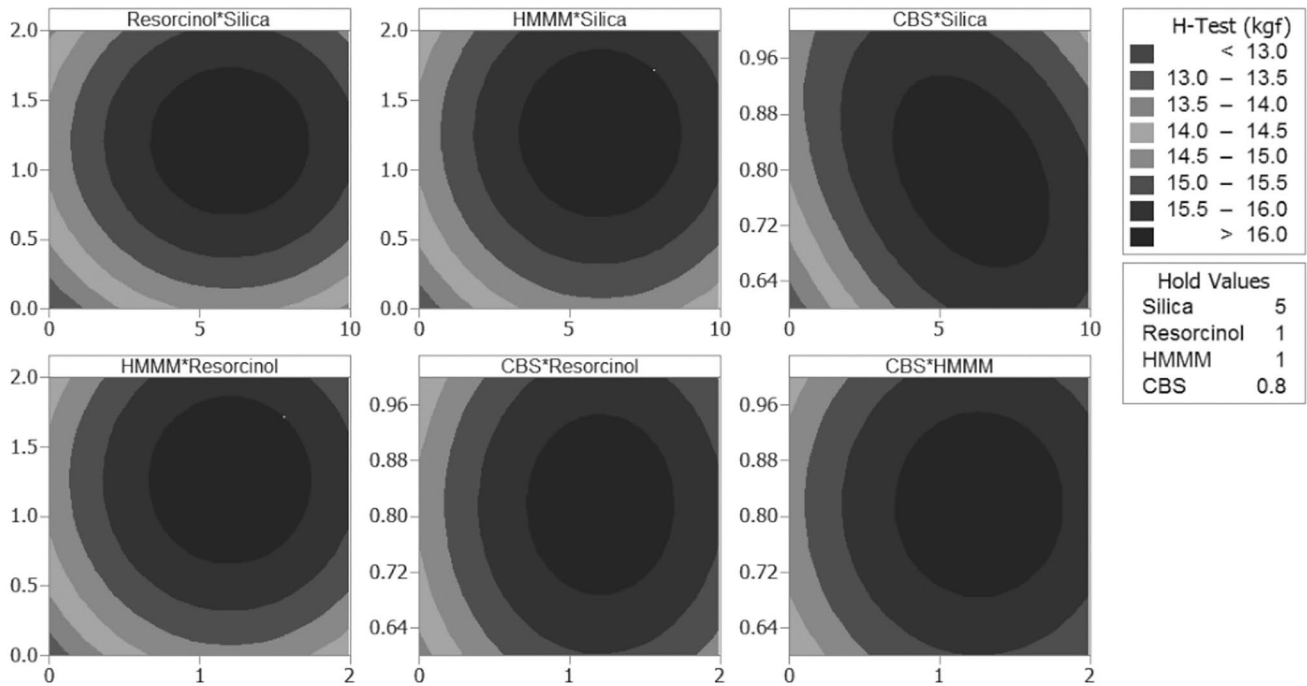


Fig. 6 Contour plots of *H* test

Minitab software shows how independent variables influence the dependent variable. These equations are based on the actual values of variables (without encoding). With regard to the determined and balanced coefficients for the measured properties, the models exhibit a good accuracy. Therefore, the value of each property can be predicted by specifying the real values of phr for each material (A, B, C, and D) in the obtained equations. Regarding the efficient coefficients obtained by fitting, the greatest effect of variables on elongation-at-break, tear strength, hardness and abrasion is respectively, related to the interactions of silica–HMMM, HMMM–HMMM, silica–resorcinol, and also in the case of *H*-pull test, the maximum effect is related to the interactions of silica–silica and resorcinol–resorcinol. Regarding the *H*-pull test, as shown in the contour diagram of Fig. 6, the highest adhesion values are almost related to the mean level of the variables.

To achieve the optimized formulation, higher tear strength, adhesion as well as low abrasion were aimed. Also, the elongation-at-break and hardness were adjusted at 600% and 57 shore A, respectively. Minitab, using the achieved models and expected desirability, offers the best-optimized values to meet the desired properties. In the optimization process, the importance degree of adhesion property (*H*-pull test) is considered 10 and other properties are considered 1. As in this research, the adhesion property between the polyester cord and rubber compound is the target. The optimized values are shown in Fig. 7. As it is observed, the optimized value for each variable includes silica: 5.76 phr, resorcinol:

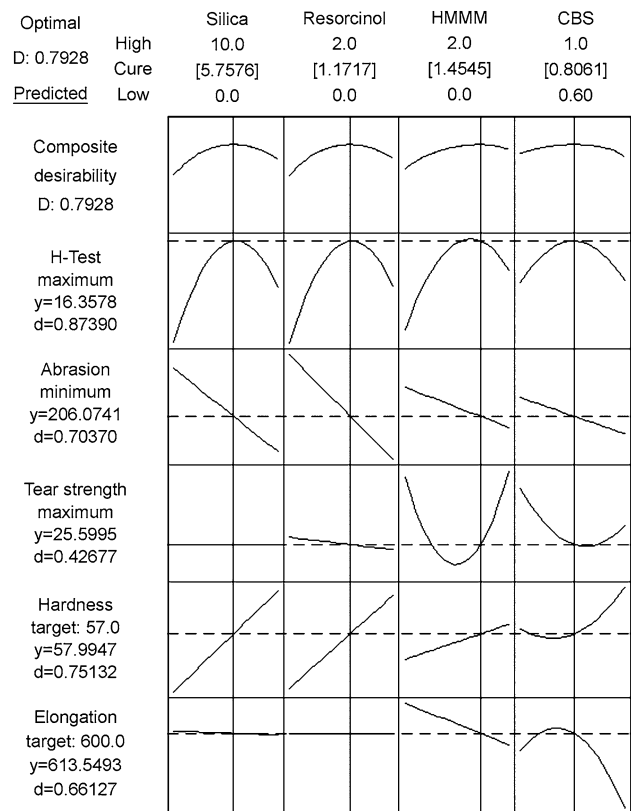


Fig. 7 Optimized values for each variable

1.17 phr, HMMM: 1.45 phr and CBS/MBTS: 0.81/0.19 phr. The results show that desirability for a composite is 0.7928, which indicates that an approximation of 80% of the properties obtained from optimization corresponds to the desirable values.

Verification experiments

According to the optimized values for each variable, the formulation was again prepared and investigated by physical and mechanical tests. Comparison of the results obtained from modeling data and experimental results is presented in Table 4. This shows the proper fitting of the modeling data with the experimental results. All the obtained values for each variable confirmed that most of them displayed more favorable properties in the middle levels, and it was not necessarily possible to optimize the properties by maximum or minimum of these variables.

Conclusion

The goals in this paper were to improve adhesion and provide a mathematical model for adhesion property as well as maintaining some of the mechanical properties. So, the Box–Behnken design was applied to optimize adhesion and physico-mechanical properties of NR/SBR compounds to coat polyester cords for the first time. The investigated factors are silica, resorcinol, HMMM, CBS and MBTS content. According to the importance of adhesion as compared with other properties in the polyester fiber coating, in the final optimization, the degree of adhesion importance (*H* test) and other properties were considered 10 and 1, respectively. The outstanding results of Box–Behnken method suggested that the proposed compound with 5.76, 1.17, 1.45 and 0.81/0.19 phr for silica, resorcinol, HMMM and CBS/MBTS, respectively, can increase the adhesion of the polyester cord to the compound in an acceptable amount, while giving the desired physico-mechanical properties like 27.4 kgf/cm tear strength.

References

- Li Z, Wan J, Zhang L, Cui J, Zhao S (2017) Effects of heat and moisture on characteristics, tensile properties of RFL-coated rayon cords, and their adhesion with NR/SBR matrix. *J Appl Polym Sci* 134:45559
- Ebnesajjad S (2009) *Adhesives technology handbook*, 2nd edn. William Andrew Publishing, Norwich
- Crowther B (2001) *Handbook of rubber bonding*. Rapra Technology Limited, UK
- Dierkes W, Louis A, Noordermeer J, Blume A (2019) A novel approach of promoting adhesion of reinforcing cord to elastomers by plasma polymerization. *Polymers* 11:577
- Walkup M (2013) Resorcinol in tire applications. *Tire Technol Int*
- Lee HK, Kim DS, Won JS, Jin DY, Lee HJ, Lee SG (2016) Effects of thermal and humidity aging on the interfacial adhesion of polyketone fiber reinforced natural rubber composites. *Adv Mater Sci Eng* 2016:1–8
- Shi X, Ma M, Lian C, Zhu D (2014) Investigation of the effects of adhesion promoters on the adhesion properties of rubber/steel cord by a new testing technique. *J Appl Polym Sci* 131:39460
- Tonatto MLP, Forte MMC, Amico SC (2017) Compressive-tensile fatigue behavior of cords/rubber composites. *Polym Test* 61:185–190
- Wennekes WB, Datta RN, Noordermeer JWM, Elkind F (2008) Fiber adhesion to rubber compounds. *Rubb Chem Technol* 81:523–540
- Li P, Wu Y, Zhou Y, Zuo Y (2019) Preparation and characterization of resorcinol-dialdehyde starch-formaldehyde copolycondensation resin adhesive. *Int J Biol Macromol* 127:12–17
- Sen AK (2001) *Coated textiles: principles and applications*. Technomic Publishing Company, USA
- Wootton DB (2001) *The application of textiles in rubber*. Rapra Technology Limited, United Kingdom
- Serafinska A, Kaliske M, Zopf C, Graf W (2013) A multi-objective optimization approach with consideration of fuzzy variables applied to structural tire design. *Comput Struct* 116:7–19
- Li L, Choi Y-J, Boonkerd K, Zhang J, Gao LJ, Zhang ZX, Kim JK (2013) Prediction of the chlorobutyl rubber/natural rubber blend properties using a genetic algorithm and artificial neural network. *Rubber Chem Technol* 86:190–204
- Dey P, Naskar K, Dash B, Nair S, Unnikrishnan G, Nando GB (2014) Thermally cross-linked and sulphur-cured soft TPVs based on S-EB-S and S-SBR blends. *RSC Adv* 4:35879–35895
- Balachandran M, Bhagawan SS, Muraleekrishnan R (2011) Modeling and optimizing properties of nanoclay–nitrile rubber composites using Box–Behnken design. *Rubber Chem Technol* 84:455–473
- Balachandran M, Devanathan S, Muraleekrishnan R, Bhagawan SS (2012) Optimizing properties of nanoclay–nitrile rubber (NBR) composites using face centred central composite design. *Mater Des* 35:854–862
- Balachandran M, Stanly P, Mulaleekrishnan R, Bhagawan SS (2010) Modeling NBR-layered silicate nanocomposites: a DOE approach. *J Appl Polym Sci* 118:3300–3310
- Shiva M, Atashi H, Hassanpourfard M (2012) Studying the abrasion behavior of rubbery materials with combined design of experiment-artificial neural network. *Chin J Polym Sci* 30:520–529
- Ahsan Q, Mohamad N, Tiak Chuan S (2015) Mechanical properties of rubber mat compound via two factors modelling using response surface methodology. *Appl Mech Mater* 761:358–363
- Derringer GC (1988) Statistical methods in rubber research and development. *Rubber Chem Technol* 61:377–421
- Tian M, Park H, Row Kyung H (2013) Optimization of synthesis amounts of polymers with two monomers by different methods based on response surface methodology. *Adv Polym Technol* 33:21405
- Sridhar V, Prasad K, Choe S, Kundu PP (2001) Optimization of physical and mechanical properties of rubber compounds by a response surface methodological approach. *J Appl Polym Sci* 82:997–1005
- Sridhar V, Shanmugaraj AM, Kim JK, Tripathy DK (2009) Optimization of carbon black and nanoclay filler loading in chlorobutyl vulcanizates using response surface methodology. *Polym Compos* 30:691–701
- Kukreja TR, Kumar D, Prasad K, Chauhan RC, Choe S, Kundu PP (2002) Optimisation of physical and mechanical properties of rubber compounds by response surface methodology—two

- component modelling using vegetable oil and carbon black. *Eur Polym J* 38:1417–1422
26. Rajan R, Varghese S, Balachandran M, George KE (2016) Response surface methodology: a tool for assessing the role of compounding ingredients in peroxide vulcanization of natural rubber. *Rubber Chem Technol* 89:211–226
 27. Salvatori Pablo E, Sánchez G, Lombardi A, Nicocia E, Bortolato Santiago A, Boschetti Carlos E (2018) Optimization of properties in a rubber compound containing a ternary polymer blend using response surface methodology. *J Appl Polym Sci* 135:46548
 28. Ghasemi I (2010) Evaluating the effect of processing conditions and organoclay content on the properties of styrene–butadiene rubber/organoclay nanocomposites by response surface methodology. *Exp Polym Lett* 4:62–70
 29. Durairaj RB (2005) Resorcinol chemistry, technology and applications. Springer, Berlin
 30. Dick JS (2014) How to improve rubber compounds. Hanser Publishers, Germany
 31. Hewitt N (2007) Compounding precipitated silica in elastomers. William Andrew, USA