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Research Article

Property Optimisation of EPDM Rubber Composites Using Mathematical and Statistical Strategies

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This paper describes a study in which EPDM-based rubber composites were investigated aiming at developing formulations subjected to restrictions on cost and the properties of the material. The contents of components other than calcium carbonate, paraffinic oil, and CBS vulcanising accelerator, as well as additives and processing conditions, were kept constant. Fractional factorial design coupled with computational numerical optimisation was used to minimise the number of mixtures. The results demonstrate that statistical design of experiments and particle swarm optimisation (PSO) algorithms are promising methods to design composition variables. Mixture costs as low as 1.92 US$/kg can be achieved in compositions containing, for example, 107 phr of calcium carbonate, 95 phr of paraffinic oil, and 1.13 phr of CBS accelerator. The corresponding composite property-predicted values were 66.8 Shore A for hardness, tensile strength of 7.8 MPa, 570.8% elongation at break, and 23.0% rebound resilience. This demonstrates that, in this way, the desired product with specified characteristics can be comfortably manufactured at minimum cost.

1. Introduction

The modern rubber industry offers a very wide variety of technological products derived from synthetic elastomers like ethylene–propylene–diene M-class rubber (EPDM rubber). These products find applications in different fields, namely, in automotive, naval, and mechanical industries. Such rubber compounds (composites) are manufactured from complex mixtures of different raw materials (different kinds of EPDM elastomers, fillers, process oil, vulcanising and protecting agents, etc.), and the production steps involve a variety of processes (e.g., mixing, extruding, cutting, moulding, and vulcanising) [1–6].

The performance and manufacture of rubber products has been receiving more and more attention, and the industry has successfully introduced and applied the usage of a series of quality certification standards. Also, stringent market and price competition demand shorter product development cycles and reduced costs, which include raw materials and processing, as well as research and development costs. All of that makes it difficult to define an adequate new formulation by simple adjustment of older ones, based on rule of thumb or virtue of experience.

The application of statistical design of experiments (DoE) to the industrial formulation of rubber composites can be a convenient and accurate means of obtaining reliable quantitative estimates of properties as the result of any change in contents of raw materials [7, 8]. The modelling of a given property using the design of mixture experiments is becoming common practice [9–17] and was proven, in all cases reported, to lead to greater efficiency and confidence in the results obtained, and to be less demanding in time and both material and human resources.

Many studies on the effects of raw material changes on the physical properties of rubber composites coupled with DoE can be found in the literature, but few data are available on research carried out using the cost characteristics of rubber compounds. However, although standard requirements for physical and mechanical properties of a rubber compound are mandatory, high costs might preclude the product's competitiveness in the market.

Thus, the rubber compound engineer most often needs to produce an optimised formulation, which fits the requirements of physical and mechanical properties, while subjected to processing and cost constraints [18]. In industrially oriented applications of materials like rubbers or ceramics, the technique generally used to optimise equality and inequality property constraints is the graphical overlay of contour plots generated by the regression models of the properties [10, 15, 19–21]. However, as the number of functions and constraints increases, so do the difficulties to handle and design the different contour plots, and these strategies begin to have a rather limited performance.

Computational optimisation of nonlinear programming problems, which include numerical analysis of continuous and discrete variables, has been an active and important engineering research issue. The optimisation problem consists in finding out a solution for the objective function and related constraints. The use of particle swarm optimisation (PSO) algorithms for solving nonlinear, multimodal, and nondifferentiable optimisation problems, which are not well fitted for conventional optimisation algorithms, has gained increasing attention in recent years [22, 23]. PSO algorithms fundamentals result from the observation, interpretation, and modelling of the movements of individuals in bird flocks or fish schools, as well as their group behaviour as a swarm. It is a simple algorithm, so only a few lines of the computer program based on simple mathematical operations are needed to deploy the basic tool of PSO. The computer-aided optimisation method provides an efficient way to predict the optimum formulation without using those awkward contour plot graphical overlays.

In this work, a fractional factorial design of experiments was used to study the effect of filler, process oil, and vulcanising accelerator contents on the mechanical properties (hardness, tensile strength, elongation at break, and rebound resilience) of EPDM rubber composites. Regression models were calculated from the results of the measured properties, under constant processing conditions and contents of other raw materials and additives. The regression models were then used in a PSO algorithm to obtain optimised EPDM rubber formulations subjected to property constraints and cost requirements.

2. Experimental Procedure

2.1. Compound Ingredients and Base Composition. The elastomer used in this work was a commercial M-class ethylene–propylene–diene (EPDM) monomer (Keltan, supplied by Branco Indústria e Comércio Ltda). Other ingredients included carbon black (Spheron 5000, Cabot Brasil Indústria e Comércio), calcium carbonate (Mineração São Judas Ltda), and paraffinic oil (Ipiranga Indústria Quimica Ltda). Besides these ingredients, the mixtures include special additives, namely, vulcanising agent (sulphur, Basile Química), vulcanising accelerator (CBS, N-cyclohexylbenzothiazol-2-sulphenamide, Basile Química), vulcanising activator (ZnO, Brazinco Indústria e Comércio), aging inhibitor (Naugard 495, 4-5-methyl-2-mercaptobenzimidazole, Chemtura Indústria Química do Brasil Ltda), and stearic acid (Basile Química).

Table 1: Base composition of industrial EPDM rubber composites.

Ingredients	Content [phr][a]
EPDM	100.00
Carbon black	115.00
Calcium carbonate (CC)	Variable
Paraffinic oil (PO)	Variable
Sulphur, vulcanising agent	0.40
CBS, vulcanising accelerator (VA)	Variable
ZnO, vulcanising activator	5.00
Naugard 495, aging inhibitor	1.00
Stearic acid	1.00

[a]Per hundred of rubber, by weight.

The rubber formulations were based on that of heat- and air-resistant products, such as the automotive hoses manufactured by NSO Borrachas, Joinville, SC, Brazil. As shown in Table 1, the contents (phr, per hundred of rubber, by weight) of calcium carbonate filler (CC), process (paraffinic) oil (PO), and vulcanising accelerator (VA) were varied in the compositions, but the contents of the other raw materials and additives, as well as processing conditions, were kept constant. The chosen processing conditions closely followed the conventional laboratory rubber compound procedure used in industrial practice [5, 6].

2.2. Experiment Design. A 3^{3-1} fractional factorial design was chosen to model the effect of varying contents of the three factors CC, PO, and VA on the composite properties because it required the minimum number of experiments (nine mixture compositions) for which nonlinear effects and interactions of all the factors could be investigated [7, 8]. Given that the contents of all raw materials and additives other than the three factors were kept constant, a new calculation basis was defined to translate contents of the factors from their usual phr base values, m_i, into fractions, X_i, as needed in the factorial design, and vice versa. Among the factors limiting phr values, the CC content is the highest of them all (125 phr in the reference product). Hence, this was chosen as the base reference value, M, and contents of all the factors were then expressed as weight fraction relative to that of the CC content, that is, $X_i = m_i/M$. To design the matrix of mixture experiments, three weight fraction content levels were chosen within the usual ranges for the manufacture of general-purpose products, as shown in Table 2.

STATISTICA statistical software (StatSoft Inc., 2010) was used to determine the geometric and coded notations as well as randomise the treatment combinations, resulting in a standard experiment order. Table 3 shows the mixing ratios of chosen factors for the nine compounds, obtained from the 3^{3-1} fractional factorial design.

2.3. Mixture Preparation, Moulding, and Property Evaluation. For each of the nine different formulations, in two replications, the selected amounts of raw materials were mixed in a two-roll laboratory mill (Equipabor, Brazil) at 70°C and 1 : 1.20 speed ratio, as recommended by the ASTM

Table 2: Factors and levels (weight fraction) adopted for the 3^{3-1} fractional factorial design used to define EPDM rubber composites.

Factor	Level (weight fraction)		
	Low	Medium	High
Calcium carbonate (CC)	0.000	0.500	1.000
Paraffinic oil (PO)	0.400	0.600	0.800
Vulcanising accelerator (VA)	0.008	0.014	0.020

Table 3: Mixture compositions (weight fractions of chosen factors only) in the 3^{3-1} fractional factorial design.

Mixture	Weight fraction		
	CC (X_1)	PO (X_2)	VA (X_3)
1	0.000	0.400	0.008
2	0.000	0.600	0.020
3	0.000	0.800	0.014
4	0.500	0.400	0.020
5	0.500	0.600	0.014
6	0.500	0.800	0.008
7	1.000	0.400	0.014
8	1.000	0.600	0.008
9	1.000	0.800	0.020

D 15 Standard [24]. The sheets obtained were conditioned at $25 \pm 2^\circ$C for 24 h in a sealed container before the estimation of optimum curing time. Batches were then compression-moulded to a 90% cure using an electrical resistance heated hydraulic press (model EMIC) at ~10 MPa and 160°C during 10 minutes.

The hardness (HD) test was carried out according to ASTM D 676 Standard [25] using a Zwick durometer. The tensile strength (TS) and elongation at break (EB) tests were carried out according to ASTM D 412 Standard [26] using an EMIC DL 2000 testing machine. The rebound resiliency (RS) was determined in accordance with ASTM D2632 using a rebound tester [27]. For each mixture, in each replication, the property final value was taken as the average of the test results obtained for five different test pieces.

2.4. Optimisation Strategy. The experimental results obtained for each property were used to iteratively calculate, with STATISTICA, the coefficients of a regression equation, until a statistically relevant model and response surface was obtained, relating that property value with the weight fractions of calcium carbonate (CC), paraffinic oil (PO), and vulcanising accelerator (VA) present in the corresponding mixture of raw materials.

A PSO algorithm was developed using the property model equations obtained with STATISTICA and common limiting property values (those of the reference automotive hoses manufactured by NSO Borrachas, Joinville, SC, Brazil), aiming at finding the best composition range (weight fractions) that meets the property limits while minimising costs of the composites.

3. Results and Discussion

3.1. Measured Properties and Statistical Analysis. Table 4 presents the values of hardness (HD), tensile strength (TS), elongation at break (EB), and rebound resiliency (RS) obtained for the nine mixtures in two replications. Material costs for the nine mixtures in replication 1 are also shown in Table 4.

Table 5 shows the results of the variance analysis of the regression equations obtained for HD, TS, EB, and RS, using the nomenclature commonly found in the literature (major statistical properties: p values and coefficient of multiple determination R^2) [7, 8]. It can be seen that, in all cases, the nonlinear models are statistically significant at the required level (p value ≤ significance level) and present small variability (high coefficients of multiple determination). Although only effects with p value lower than 0.10 were considered significant, p values higher than 0.10 were kept in Table 5 because those effects should appear in the models. In all cases, the errors could be considered randomly distributed around a zero mean value (i.e., they are uncorrelated), which suggests a common constant variance. On the basis of this analysis, the regression models obtained were accepted to describe the effect of contents of raw materials (CC, PO, and VA) on HD, TS, EB, and RS, and the final results are (1). These equations are all referred to the weight fractions of the components calcium carbonate (X_1), paraffinic oil (X_2), and vulcanising accelerator (X_3), so that mixing of raw materials can easily be carried out.

$$\begin{aligned} \mathrm{HD} &= 91.45 - 42.13X_2 + 252.07X_3 + 10.64X_1X_2^2, \\ \mathrm{TS} &= 12.39 - 0.06X_1 + 1.72X_1^2 - 1.76X_2^2 - 890.02X_3^2 \\ &\quad - 19.51X_1X_2 + 16.22X_1X_2^2, \\ \mathrm{EB} &= 475.55 - 56.58X_1 - 103.51X_2 + 471.24X_2^2 \\ &\quad - 1040.76X_3 - 77.94X_1X_2^2, \\ \mathrm{RS} &= 29.49 - 4.94X_1 - 132.57X_3 - 2.44X_1X_2^2. \end{aligned} \tag{1}$$

3.2. Experimental Validation of the Models. Three extra mixtures, F_1, F_2, and F_3 (check-point mixtures), were used to validate the calculated statistical models (the mixtures and their test pieces were prepared following the same procedure as before). Table 6 presents the compositions of those three mixtures and the corresponding measured and predicted values for HD, TS, EB, and RS. It can be seen that the estimates calculated using (1) can be higher or lower than, but are always very close to, the corresponding experimental value (low error), which validates the calculated models.

3.3. Cost and Property Optimisation Using PSO Algorithm. Following the same procedure and reasoning described above for the compounds mechanical properties, a valid and significant regression model was also obtained for the cost of mixtures (CT), which can be described by the following equation:

$$\mathrm{CT} = 2.68 - 0.66X_1 - 0.26X_2 + 0.28X_3. \tag{2}$$

TABLE 4: Measured values of hardness (HD), tensile strength (TS), elongation at break (EB), rebound resilience (RS), and material costs (CT) for the nine designed mixtures in two replications.

Mixture		HD (Shore A)	TS (MPa)	EB (%)	RS (%)	Cost (US$/kg)
Replication 1	1	77.0 ± 0.7	12.5 ± 0.1	496.7 ± 08.2	27.8 ± 1.1	2.63
	2	70.8 ± 0.5	10.7 ± 0.2	588.0 ± 10.9	26.6 ± 0.6	2.55
	3	59.4 ± 0.6	11.2 ± 0.6	670.0 ± 11.6	26.8 ± 0.5	2.47
	4	79.0 ± 0.0	9.5 ± 0.2	453.3 ± 10.3	25.0 ± 0.7	2.22
	5	70.0 ± 0.7	9.0 ± 0.2	550.0 ± 10.0	24.4 ± 0.6	2.17
	6	61.4 ± 0.6	8.7 ± 0.2	636.0 ± 08.9	26.4 ± 0.6	2.11
	7	80.8 ± 0.5	8.1 ± 0.2	403.3 ± 08.2	22.6 ± 0.9	1.91
	8	72.4 ± 0.9	7.2 ± 0.2	468.0 ± 10.9	23.8 ± 0.5	1.89
	9	65.2 ± 1.3	7.4 ± 0.2	565.0 ± 10.5	24.6 ± 0.6	1.87
Replication 2	1	79.8 ± 0.5	11.6 ± 1.8	496.6 ± 08.2	29.6 ± 0.9	—
	2	70.8 ± 0.8	11.8 ± 0.4	552.0 ± 10.6	26.0 ± 0.0	—
	3	63.4 ± 0.9	11.1 ± 0.4	670.0 ± 11.6	29.0 ± 1.2	—
	4	82.4 ± 1.1	9.3 ± 0.2	436.0 ± 08.9	25.2 ± 1.1	—
	5	76.4 ± 0.9	9.6 ± 0.2	512.5 ± 09.6	26.2 ± 0.5	—
	6	61.2 ± 0.8	9.0 ± 0.2	647.5 ± 09.6	26.6 ± 0.6	—
	7	73.0 ± 0.7	8.8 ± 0.2	470.0 ± 11.6	*	—
	8	73.0 ± 1.2	7.5 ± 0.2	486.6 ± 10.3	25.2 ± 0.5	—
	9	75.6 ± 0.6	7.4 ± 0.2	566.7 ± 10.3	22.6 ± 0.6	—

*Value could not be measured.

TABLE 5: Major statistical properties [7, 8] relevant for variance analysis (R^2, p value for the significance and lack-of-fit tests) of the calculated regression models (X_1 calcium carbonate, X_2 paraffinic oil, and X_3 vulcanising accelerator).

Effect	p value*			
	HD	TS	EB	RS
X_1	—	0.00	0.00	0.00
X_1^2	—	0.07	—	0.07
X_2	0.00	—	0.00	—
X_2^2	—	0.00	0.13	0.00
X_3	0.06	0.10	0.12	0.10
X_3^2	—	0.20	—	0.20
X_1X_2	—	0.27	—	0.27
$X_1X_2^2$	0.03	0.26	0.09	0.25
R^2	0.85	0.82	0.96	0.96
Lack-of-fit	0.99	0.89	0.77	0.55

*p values of the effects used in the regression models. Other insignificant p values (>0.10) are not presented.

In mathematical language, the optimisation problem consists in minimising this objective function (CT) with respect to the design variables X_1, X_2, and X_3, subjected to the nonlinear inequality constraints posed on HD, TS, EB, and RS, as presented in Table 7. These property ranges (optimisation goals) were chosen having in mind the standard property specification range commonly required for the heat- and air-resistant products used as reference. From an industrial competitiveness point of view, property values outside (above) that range are not so interesting, as they certainly imply extra cost.

The result of the PSO algorithm procedure (minimum costs) is returned as a composition range (weight fraction) for each of the raw materials CC, PO, and VA. These ranges are also presented in Table 7.

An endless number of mixture composition points can be found which meet the requirements of the properties at low cost values. Alternatively a 2D graphical visualisation can be used, by keeping constant one of the factors (feasibility curve). In the present case, the CC content was chosen as the base reference value, and contents of all the factors were expressed as weight fraction relative to that CC content. Thus, feasibility curves can be obtained as VA versus PO for various constant CC contents, as shown in Figure 1.

Figure 1 clearly shows that the function describing the infinite number of mixtures with properties within the specified ranges is complex and nonlinear. For a constant CC weight fraction equal to 1.00, optimised costs vary from 1.82 to 1.87 US$/kg, and the PO weight fraction can vary between 0.60 and 0.78, whereas the VA weight fraction has a much narrower range, varying from 0.008 to 0.009. Similarly, when the CC level is kept constant at 0.50, the composite costs vary between 2.17 and 2.23 US$/kg. The optimum VA weight fraction varies nearly in the same range, but the optimum PO weight fractions have a different range, in this case from 0.47 to 0.68. Calcium carbonate at lower levels has a more complex effect on the optimisation and results in higher costs, ranging from 2.53 to 2.58 US$/kg. Although the PO weight fraction range is narrower (0.40 to 0.60), the VA weight fraction has a broader range, from 0.008 to 0.017.

Table 8 presents the raw materials' weight fractions and property-predicted values for three illustrative mixtures within the optimum range and shows the corresponding costs, ranging from 1.92 to 2.37 US$/kg. From those weight fractions' compositions, the formulations of the corresponding optimised EPDM rubber composites can now be calculated back from the reference CC content (125 phr). The full formulations corresponding to the mixtures in Table 8, meeting the

Table 6: Composition (weight fraction) of the check-point mixtures F_1, F_2, and F_3 and corresponding measured (M) and predicted (P) values of hardness (HD), tensile strength (TS), elongation at break (EB), and rebound resilience (RS).

Wt. fraction	F_1		F_2		F_3	
X_1 (CC)	0.303		0.677		0.860	
X_2 (PO)	0.438		0.616		0.755	
X_3 (DV)	0.008		0.015		0.009	
Property	P	M	P	M	P	M
HD (Shore A)	68.5	69.8 ± 0.7	76.2	78.4 ± 0.7	69.5	70.6 ± 0.7
RS (%)	26.5	27.0 ± 0.0	25.0	25.0 ± 0.0	24.3	24.0 ± 0.0
EB (%)	585.3	668.3 ± 9.8	470.5	522.0 ± 19.2	551.0	526.7 ± 12.1
TR (MPa)	10.47	10.10 ± 0.3	8.92	9.32 ± 0.3	7.65	7.43 ± 0.2

Table 7: Constraint requirements posed on mechanical properties (HD, TS, EB, and RS) and raw materials' (CC, PO, and VA) weight fraction ranges that minimise costs of the composites, as returned by the PSO algorithm.

Name	Goal	Lower limit	Upper limit
HD (Shore A)	In the range	65	75
TS (MPa)	In the range	7.5	12.5
EB (%)	In the range	490	560
RS (%)	In the range	23	28
CT (US$/kg)	Minimise	—	—
X_1 (CC)	In the range	0.00	1.00
X_2 (PO)	In the range	0.40	0.80
X_3 (VA)	In the range	0.008	0.020

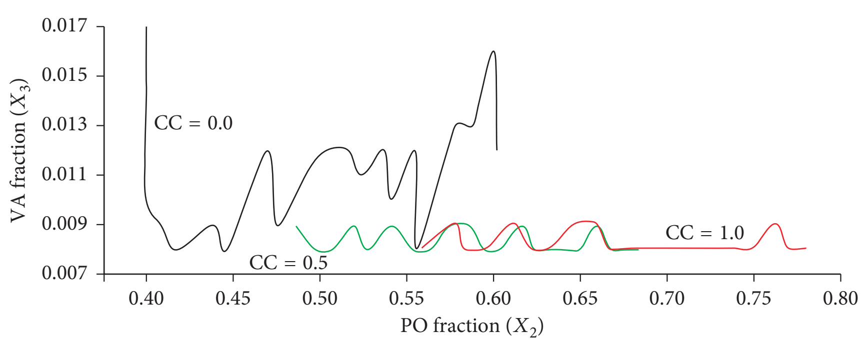

Figure 1: Feasibility curves relating VA and PO weight fractions for optimised cost of composites with properties within the specified ranges, when the CC weight fraction is kept constant at 1.0, 0.5, and 0.0.

Table 8: Examples of optimised mixture compositions and predicted values of the corresponding properties.

Mixture point	Weight fractions			HD	RS	EB	TS	Cost
	X_1 (CC)	X_2 (PO)	X_3 (VA)	(Shore A)	(%)	(%)	(MPa)	(US$/kg)
1	0.860	0.755	0.009	66.8	23.0	570.8	7.8	1.92
2	0.677	0.616	0.015	72.0	23.5	516.7	8.3	2.08
3	0.303	0.438	0.008	75.6	26.8	490.6	10.5	2.37

requirements of all the mechanical properties, are presented in Table 9.

4. Conclusions

This study showed that fractional factorial design of experiments in two replicates based on three mixture ingredients generally used on EPDM rubber composites and a particle swarm optimisation (PSO) algorithm are promising methods to design composition variables.

For the chosen key raw materials (calcium carbonate, paraffinic oil, and CBS vulcanising accelerator) and the processing conditions under consideration, the optimisation results readily show that there is an infinite number

Table 9: Examples of optimised EPDM rubber composites with costs between 1.92 and 2.37 US$/kg.

Ingredients (phr)[a]	Formulation 1	Formulation 2	Formulation 3
EPDM	100.00	100.00	100.00
Carbon black	115.00	115.00	115.00
Calcium carbonate (CC)	107.00	85.00	38.00
Paraffinic oil (PO)	95.00	77.00	55.00
Sulphur, vulcanising agent	0.40	0.40	0.40
CBS, vulcanising accelerator (VA)	1.13	1.88	1.00
ZnO, vulcanising activator	5.00	5.00	5.00
Naugard 495, aging inhibitor	1.00	1.00	1.00
Stearic acid	1.00	1.00	1.00

[a]Per hundred of rubber, by weight.

of compositions that meet specified property values with minimum costs. For example, within the base composition range investigated, mixture costs as low as 1.92 US$/kg can be achieved with the use of 107 phr of calcium carbonate, 95 phr of paraffinic oil, and 1.13 phr of CBS vulcanising accelerator. The predicted values for the corresponding compound properties are 66.8 Shore A for hardness, tensile strength of 7.8 MPa, 570.8% elongation at break, and 23.0% rebound resilience.

In this way, this investigation showed that the specified characteristics of the desired product can be subjected to restrictions typical of the manufacture process, and a broad range of compositions can still be selected so that the final product has minimum cost and can be comfortably manufactured.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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