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Effect of extruder parameters on mechanical properties of revulcanized ground rubber tire powder

Summary — Response Surface Methodology (RSM) was used to study the effects of processing parameters *viz.* barrel temperature and screw speed on the mechanical properties of revulcanized waste ground rubber tire powder from a counter rotating twin screw extruder. Central composite rotatable design (CCRD) for two variables at five levels was chosen as the experimental design. The following parameters were analyzed: rheometric maximum torque, tensile strength, elongation at break, stress at 100 % elongation, hardness and tear strength. The results obtained after measuring the responses were fitted as two variable second order equations which were confirmed experimentally. Barrel temperature is found to be the more influencing factor than the screw speed. Optimum properties were predicted at temperature 197 °C and screw speed 25 rpm.
Key words: twin screw extruder, design of experiments, response surface methodology, revulcanization, waste ground rubber tire, mechanical properties.

WPŁYW PARAMETRÓW PRACY WYTŁACZARKI NA WŁAŚCIWOŚCI MECHANICZNE REWULKANIZOWANEGO PROSZKU GUMOWEGO OTRZYMYWANEGO Z OPON

Streszczenie — Do badania wpływu parametrów procesu rewulkanizacji takich jak temperatura bębna i szybkość obrotów ślimaka na właściwości mechaniczne rewulknizowanego proszku z odpadów gumowych zastosowano metodologię analizy powierzchni odpowiedzi (RSM z ang. *response surface methodology*). Do sporządzenia planu eksperymentu wybrano metodę CCRD (z ang. *central composite rotatable design*) z dwiema zmiennymi na pięciu poziomach (tabela 1). Analizowano następujące właściwości rewulkanizatów: maksymalny moment obrotowy, wytrzymałość na rozciąganie (rys. 1—4), wydłużenie przy zerwaniu, naprężenie przy 100 % wydłużeniu, twardość i wytrzymałość na rozerwanie. Wyniki pomiarów otrzymane jako „odpowiedź" badanego układu opisywano równaniami dwu zmiennych drugiego rzędu. Stwierdzono, że temperatura bębna jest czynnikiem bardziej wpływającym na właściwości produktu niż szybkość obrotów ślimaka. W wyniku analizy otrzymano prognozę optymalnych parametrów pracy wytłaczarki tj. temperaturę bębna równą 197 °C, a szybkość obrotów ślimaka 25 rpm.
Słowa kluczowe: wytłaczarka dwuślimakowa, plan eksperymentu, metodologia analizy powierzchni odpowiedzi, rewulkanizacja, odpady z opon gumowych, właściwości mechaniczne.

Waste ground rubber tire is an abundantly available alternate elastomer source which has not been fully utilized and explored in detail. Since vulcanized elastomers have three dimensional crosslinked structure, it is difficult to get satisfactory mechanical properties even after revulcanization. A number of ways have been suggested [1] for increasing the properties like grinding the waste tires into powder, reclaiming by thermo-mechanical methods and devulcanization of sulphur crosslinking points using ultrasonic waves, microwaves, bacteria or by chemicals like disulphides, including disulphides derived from natural products like garlic. Adhikari *et al.* [2] have given a comprehensive summary of the latest methods of waste rubber utilization. New methods [3, 4] involving extruders have also been developed to break the crosslinking in the vulcanizates and render them recyclable to useful products. Recently, Maridass and Gupta [1] have developed a counter rotating twin screw extruder with L/D ratio equal 10 for devulcanization of ground rubber powder vulcanizates. But, most of the reported studies on extruder optimizations were done by the one variable at a time (OVAT) experimental method, which means that only one experimental parameter like screw speed or input feed or die diameter *etc.* was changed while the remaining parameters were constant for a given set of extrusions. In our earlier studies, it has been found [1] by OVAT method that extruder temperature and screw speed are the two main factors that influence the extent of devulcanization in a twin screw extruder (TSE) while other parameters like feed input or die diameter did not have significant impact. Thus, the extruder performance can be optimized

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by establishing a functional relationship between the two important variables, namely, barrel temperature and screw speed with the responses found using response surface methodology (RSM) as discussed in detail by Box [5] and Montgomery [6]. If Y is the response of a process dependent on the levels of k factors (x_1, x_2, x_3,..., x_k) which can be precisely measured and controlled, then the quadratic response surface model for combination of these factor levels is given by:

$$Y_u = \varphi(x_{1u}, x_{2u}, x_{3u},..., x_{ku}) + \varepsilon_u \quad (1)$$

where: $u = 1, 2, 3, ..., N$; N — *number of experiments;* φ — *functional relationship between all the factors and response,* ε — *error involved.*

The fitted model is called a response because this model can be plotted as two-dimensional contour plots or three dimensional response surface diagrams.

EXPERIMENTAL

Materials

Ground rubber tire (GRT) powder of 40-mesh size was obtained from local market, Kolkata, India.

Natural rubber (ISNR-5) was obtained from the Rubber Board, Kottayam, India.

Zinc oxide, stearic acid, *N*-cyclohexyl-2-benzothiazole sulphenamide (CBS), 2,2,4-trimethyl-1,2-dihydroquinoline polymer (TMQ antioxidant) and sulphur were obtained from the standard rubber chemicals suppliers from India.

Methods

A laboratory scale intermeshing counter-rotating twin screw extruder was used of the same design in our previous studies except that this had a modified L/D ratio of 8 while other parameters remained the same [1]. The extrudate coming out as product was designated as extrusion processed ground rubber tire (EPGRT). It was then mixed with natural rubber and other ingredients as shown in the recipe in Table 1, vulcanized and the rheometric and mechanical properties were tested. These properties were treated as responses.

A Monsanto Oscillating Disc Rheometer ODR-100S was used to measure the curing behavior of the compounds at 150 °C. Maximum rheometric torque was noted for each compound. Molding was carried out at the optimum cure time t_{90}, derived from rheometer testing. The tensile and tear samples were punched out from the compression molded sheets and tests were done using a Zwick universal testing machine (UTM) at 500 mm/min rate. All mechanical tests were done following ASTM standards. Stress at 100 % elongation was recorded during the tensile testing. Hardness was determined using Shore A durometer.

T a b l e 1. Coded levels for two variables: barrel temperature and screw speed

Mix number	Coded level for barrel temperature, °C	Barrel temperature, °C	Coded level for screw speeds, rpm	Screw speed rpm
1	-1	184.5	-1	14.5
2	0	195.5	-1.414	10
3	+1	206.5	-1	14.5
4	-1	184.5	+1	35.5
5	-1.414	180	0	25
6	0	195.5	0	25
7	+1	206.5	+1	35.5
8	0	195.5	0	25
9	+1.414	210	0	25
10	0	195.5	+1.414	40
11	0	195.5	0	25
12	0	195.5	0	25

RESULTS AND DISCUSSION

A central composite rotatable design (CCRD) was chosen as the experimental design to find exact quantitative relationship between the response functions and process variables. CCRD is an efficient and proven design especially for two factors. The design is rotatable which means that all the points in the design area are at equal distance from the centre. This leads to the equal distribution of errors among all points. The number of design points in CCRD is based upon a complete 2^k factorial. The total numbers of experiments are:

$$N = 2^k + 2k + m \quad (2)$$

where: N — *total number of experiments,* k — *number of factors,* m — *number of replicates of the centre point.*

Multiple linear regression analysis was used and the data was fitted as a second order equation. The response equation in a two variable design is given by:

$$Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_3X_1^2 + \beta_4X_2^2 + \beta_5X_1X_2 \quad (3)$$

Computer analysis using the commercially available package Statgraphics plus™ gives the value of β_0, coefficients of regression for first order equation *i.e.* β_1 and β_2, and coefficients of regression for second order equation *i.e.* β_3 and β_4.

Twelve combinations were chosen for experimental design based on CCRD. The experimental region was extended from -1.414 to +1.414 in terms of coded levels of independent variables, barrel temperature and screw speed, as shown in Table 2. The lower and higher values of the variables were based on the smoothness of the extrudate with minimum surface roughness and its ease of processing in the two-roll mill by easy formation of bands, both based on visual observation. The temperature was set between 180 °C and 210 °C and screw speed between 10 rpm to 40 rpm. Twelve sets of experiments were conducted by extrusion of GRT at different temperature and screw speed as shown in Table 1.

T a b l e 2. Basic formulation of components in vulcanization

Ingredients	Contents, phr
EPGRT	70
ISNR-5	30
Zinc oxide	5.0
Stearic acid	3.0
TMQ	0.8
CBS	0.8
Sulphur	1.5

The data obtained from the twelve experiments like rheometric maximum torque, tensile strength, elongation at break (in %), stress at 100 % elongation, hardness and tear strength are given in Table 3. They were fitted into a mathematical model by multiple regression analysis. The coefficients of regression for the model fitting the eq. (3) are shown in Table 4. The model equations were used to draw two-dimensional contour plots, three dimensional response surfaces, Pareto chart and main effect plots using the software package Statgraphics Plus™ for all responses. Although all the responses are modeled into an equation, we have chosen only the tensile strength as the benchmark property and optimization was attempted with respect to this single property. We would like to clearly indicate that we have not attempted to optimize simultaneously all the responses taken together to give a single set of optimized temperature and screw speed. The plots for tensile strength are given in Figures 1—4 respectively, which are discussed as follows:

Contour plots

A contour plot can be defined as a series of curves that identify the values of factors, for which the response is constant. In general, contour plots in RSM fall under four categories *viz.* peak type, hillside type, rising ridge type and saddle type [6]. Figure 1 represents the contour

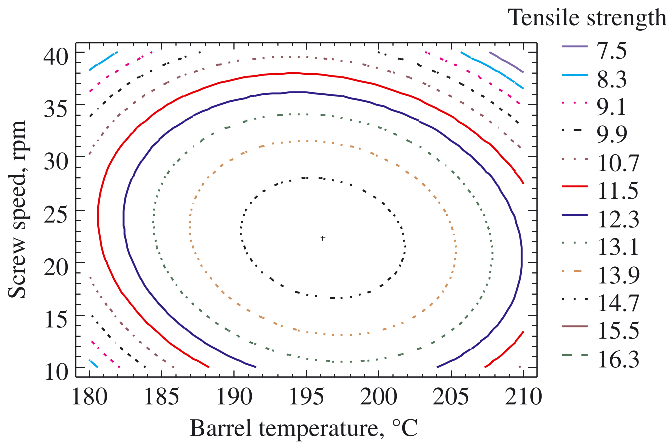

Fig. 1. Contour plot of tensile strength (in MPa) dependent on barrel temperature and screw speed

T a b l e 3. Mechanical properties of vulcanizates as responses of the experiments for different values of barrel temperature and screw speed

Coded level for extruder barrel temperature	Coded level for extruder screw speed	Rheometric max. torque, dNm	Tensile strength MPa	Elongation at break, %	Stress at 100 % elongation, MPa	Hardness Shore A	Tear strength N/mm
-1	-1	65	11.2	420	2.5	54	43.2
0	-1.414	67	12.5	370	3.4	58	41.5
+1	-1	72	13.2	260	3.5	58	42.2
-1	+1	66	10.2	220	2.7	55	43.8
-1.414	0	66	11.5	250	2.6	52	44.1
0	0	71	15.1	400	3.1	58	43.3
+1	+1	74	15.5	280	3.2	59	42.7
0	0	72	15.1	410	3.1	57	43.1
+1.414	0	72	11.5	320	3.2	59	43.8
0	+1.414	68	10.5	360	3.3	60	42.4
0	0	71	14.8	425	3.1	57	43.2
0	0	72	15.3	418	2.8	58	43.1

T a b l e 4. Coefficient of the corresponding regression equations

Property	β_0	β_1	β_2	β_3	β_4	β_5
Rheometric maximum torque, dNm	71.5	5.8714	1.1035	-2	-3.4999	0.5
Tensile strength, MPa	15.175	0.6293	-1.5071	-3.8001	-3.6999	-0.6
Elongation at break, %	413.25	-0.218	-48.5357	-143.255	-63.2487	110
Stress at 100 % elongation, MPa	3.025	0.5871	-0.0604	-0.2	0.25	-0.25
Hardness Shore A	57.5	4.4749	1.2071	-2.3751	1.1250	0.6183
Tear strength, N/mm	43.175	-0.6311	0.5932	0.7875	-1.2125	-0.05

plot for tensile strength, which is peak type. This means that the optimum response can be found in the centre point corresponding to barrel temperature 197 °C and screw speed of 25 rpm. Although, similar analysis can be done for other responses to optimize them correspondingly, we have not attempted to do this. This is because we have taken the tensile strength as the benchmark of maximum mechanical properties. Also, it is not the aim of this paper to optimize simultaneously all the responses for a given set of responses.

Response surface plots

A response surface plot is a three dimensional geometric representation of response variable (like tensile strength) plotted as a function of dependent variables (temperature and screw speed). Figure 2 represents the response surface plot of tensile strength. It can be noted

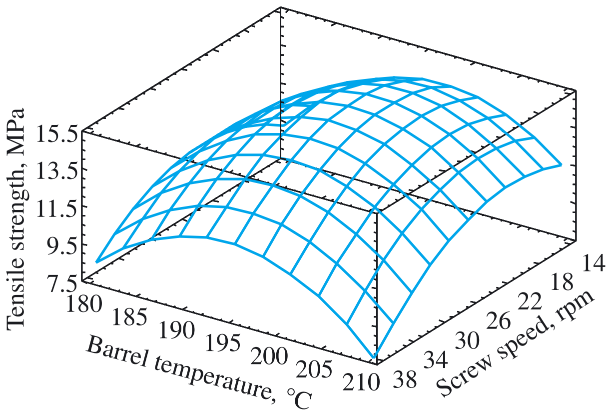

Fig. 2. Response surface plot of tensile strength

that with increase in both the temperature and screw speed, the tensile strength increases gradually to a maximum and then starts to decrease. This is because with increase in the temperature beyond 200 °C, degradation of the natural rubber starts thereby resulting in poor properties. Moreover, the SBR present in the GRT starts hardening due to cyclization reactions thereby reduce the tensile strength. With increase in the screw speed above 25 rpm, the shearing action causes more main chains break down in GRT resulting in a low quality extrudate.

Pareto chart

A Pareto chart takes a list of data that is assumed to consist of discrete categories. It determines the frequency of each category in the list, converts the frequencies to percentages, and creates the plot. Thus, it represents graphically the relative importance of different factors. Figure 3 represents the Pareto chart of tensile strength. The positive sign stands for significant factor

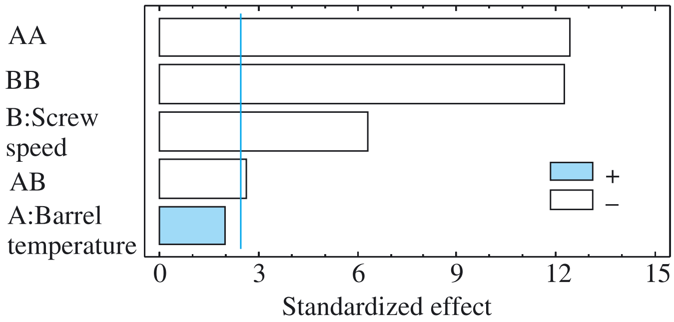

Fig. 3. Pareto chart of tensile strength

while the negative for insignificant. We can clearly see that of all the factors, barrel temperature is relatively more important than screw speed.

Main effect plot

A main effect plot describes the extent of influence of the two factors within their respective limits on the response. This gives an idea about the behavior of the response with respect to the two different factors. Figure 4 represents the main effect plot for tensile strength. This confirms the results of the response plot discussed earlier that by increasing both the temperature and screw

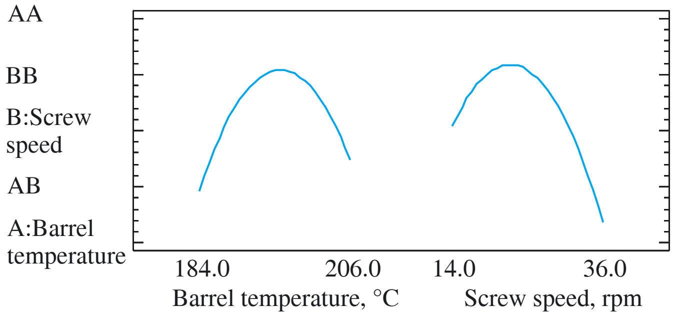

Fig. 4. Main effect plot of tensile strength

speed, the tensile strength increases, goes through a maximum and then decreases. Although there is a smooth rise and decrease in tensile strength in the studied temperature range, this is not so in screw speed. It starts deteriorating rapidly as early as 25 rpm. In order to check the validity of the proposed equation, a confirmational experiment was done by choosing random temperature and screw speed. Since 197 °C and 25 rpm was found to be optimum, we choose temperature 200 °C and screw speed 25 rpm. The responses predicted using eq. (3) after substituting the coefficients from Table 4 were compared with the experimentally determined values as shown in Table 5 and were found to agree well thereby proving the accuracy of the model. Variance analysis (ANOVA) calculation resulted in the coefficient of determination values greater than 0.9 for all the responses indicating a perfect model fit.

T a b l e 5. Comparison of experimental and predicted by regression equation properties of vulcanizate obtained for barrel temperature 200 °C and screw speed 25 rpm

Property	Predicted value	Experimental value
Rheometric maximum torque, dNm	73.97	72
Tensile strength, MPa	14.5	15.3
Elongation at break, %	377	385
Hardness Shore A	59	61
Tear strength, N/mm	43	44
Stress at 100 % elongation, MPa	3.27	3.1

CONCLUSIONS

Response surface methodology was used to model the properties of revulcanized ground rubber tire powder devulcanized in a counter rotating twin screw extruder, into a second order equation. Optimum properties, which is maximum tensile strength, was found to be at a barrel temperature of 197 °C and 25 rpm screw speed and was confirmed experimentally.

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