# Use of response surface methodology in characterization of properties of recycled high density polyethylene/ground tire rubber compositions

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**Abstract**: Searching for new ways to utilize rubber and polymer waste has been the focus of many studies conducted by research centers and companies worldwide. In this study, the influence of extrusion conditions on the properties of thermoplastic compositions containing post-production recycled high density polyethylene (PE-HD) and ground tire rubber (GTR) at 50:50 mass ratio was examined. Response Surface Methodology (RSM) was used to determine the effects of the barrel temperature (160—240 °C), screw speed (250—650 rpm) and feed rate of extrusion (1—2.5 kg/h) on the quality of the obtained products. Empirical models for determining the relationship between extrusion conditions, mechanical properties and the mass flow rate of obtained thermoplastic compositions were developed. It has been determined that tensile strength, elongation at break, hardness and the mass flow rate of thermoplastic compositions depend mainly on the barrel temperature. Properties of PE-HD/GTR blends deteriorated with increased barrel temperature, which was due to degradation of polyethylene and reclaiming of ground tire rubber. In studied extrusion conditions the influence of screw speed and feed rate on the properties of recycled high density polyethylene/ground tire rubber compositions was negligible.

**Keywords**: recycling, rubber waste, twin screw extrusion, response surface methodology.

## Zastosowanie metody powierzchni odpowiedzi w charakterystyce właściwości kompozycji recyklat polietylenu dużej gęstości/miał gumowy (PE-HD/GTR)

**Streszczenie**: Przeprowadzono badania wpływu warunków wytłaczania na właściwości kompozycji termoplastycznych poprodukcyjnego polietylenu dużej gęstości (PE-HD) z miałem gumowym (GTR), w stosunku masowym 50:50. Wpływ temperatury cylindra (160—240 °C), prędkości obrotowej ślimaków (250—650 rpm) oraz wydajności wytłaczania (1—2.5 kg/h) na jakość otrzymanych produktów określano metodą powierzchni odpowiedzi — RSM (ang. *Response Surface Methodology*). Opracowano modele matematyczne umożliwiające określenie zależności pomiędzy zmiennymi procesu wytłaczania a masowym wskaźnikiem szybkości płynięcia (*MFR*) oraz właściwościami mechanicznymi kompozycji termoplastycznych recyklat PE-HD/GTR. Stwierdzono, że wytrzymałość na rozciąganie (*TS*), wydłużenie przy zerwaniu ($E_b$), twardość (*H*) oraz masowy wskaźnik szybkości płynięcia (*MFR*) zależą głównie od temperatury cylindra ($T_b$). Właściwości otrzymanych kompozycji ulegały pogorszeniu wraz ze wzrostem temperatury cylindra, w wyniku degradacji plastomeru oraz regeneracji miału gumowego. W badanych warunkach wytłaczania wpływ prędkości obrotowej oraz wydajności na jakość otrzymanych produktów był nieznaczny.

**Słowa kluczowe**: recykling, odpady gumowe, wytłaczanie dwuślimakowe, metoda powierzchni odpowiedzi.

Waste rubber, mainly derived from used tires, is a significant source of recyclable materials. It has been estimated that about 200 thousand tons of end-of-life tires [1] per year are disposed of in Poland. This alarming number and the existing legislation [2] on waste management stimulate the scientific community and manufacturers to conduct research on the potential industrial-scale production of novel and environment-friendly products, which would be based on recycled materials. At present, the development of tire recycling technology proceeds in three main directions, i.e. a size-reduction of waste rubber [3, 4], pyrolysis of ground tire rubber and whole tires

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[5—7] and reclaiming/devulcanization of ground tire rubber [8—10].

Ground tire rubber can be used, as a filler in rubber compounds [11—13], thermoplastic compositions and thermoplastic elastomers [14—16], and as a modifier of cement [17, 18] and asphalt [19, 20]. However, a high level of ground tire rubber in the aforementioned compositions generally has negative effects on mechanical properties of the final products, because of weak interfacial interactions between the matrix and the particles of ground tire rubber. In order to increase these interactions, the processing parameters such as temperature, shear forces during the mixing process, processing efficiency, pressure, etc. should be optimized.

Kowalska [21] investigated the possible use of silane compatibilizers, and the influence of the processing conditions on the properties of polyolefin composites filled with waste rubber or phosphogypsum. Polyolefin-rubber compositions were obtained using a single screw extruder and co-rotating twin screw extruder. The final product with the best mechanical properties was obtained when it was modified with silanes in a single screw extruder.

Parasiewicz et al. [22] described a technology for the production of thermoplastic compositions via reactive mixing of EPDM/PP blends in a co-rotating twin screw extruder. The obtained composites can be used in the production of automotive parts, sport pavements, anti-vibration elements and in many other applications.

Scaffaro et al. [23] studied the influence of temperature, rotational screw speed, the dosage sequence, ratio of used components and molding methods (injection molding/compression molding) on mechanical properties of the compositions based on recycled polyethylene and ground tire rubber produced using a co-rotating twin screw extruder. Increasing barrel temperature resulted in the devulcanization of ground tire rubber, which subsequently increased interfacial compatibility. Too high temperature of the extruder barrel, i.e. at the level of 300 °C, caused carbonization of the ground tire rubber particles which then resulted in higher viscosity of the plastic phase and low values of elongation at break. Under the described conditions the method of component mixing and residence time had only a slight effect on the quality of the obtained compositions.

At present, the use of statistical methods for studying the co-rotating twin screw extrusion of polymeric compositions combined with computer simulations are gaining importance [24]. The biggest advantage of such approach is the reduction of the number of time-consuming and expensive experiments.

Costa et al. [25] used the response surface methodology to optimize the composition of PP/EPDM/ground tire rubber blends prepared with a co-rotating twin screw extruder. The obtained results confirmed that ethylene-propylene-diene rubber can be used as a compatibilizer in the compositions studied.

Zhang et al. [26, 27] conducted research on the properties of PP/ground tire rubber blends compatibilized with asphalt and SEBS-*g*-MA [polystyrene-*b*-poly(ethylene-co-butylene)-*b*-styrene grafted with maleic anhydride]. In order to optimize blend composition obtained with a co-rotating twin screw extruder, statistical methods based on neural networks and genetic algorithm were applied. This approach allowed to reduce the number of experiments to 7. Based on the measurements of mechanical properties, the optimization of the composition of different blends was performed. It was then followed by a validation procedure using experimental data. The final results confirmed that optimization of a composition of polymer blends is fully justifiable.

In the present work, thermoplastic compositions based on ground tire rubber and post-consumer high density polyethylene were obtained using a co-rotating twin screw extruder. The influence of the barrel temperature, screw speed and extrusion feed rate on the mass flow rate and mechanical properties of obtained products was determined by means of the Response Surface Methodology (RSM). In this work we continue our previous research on compatibilization and processing condition of thermoplastic compositions filled with ground tire rubber [28—31].

## EXPERIMENTAL PART

### Materials

Ground tire rubber (GTR) (size fraction up to 1.5 mm) was prepared from worn tires (passenger and truck tires in ratio ca. 50:50) by grinding at the ambient temperature. GTR was obtained from ABC Recykling, Krosno Odrzańskie (Poland). The characteristics of ground tire rubber were presented in Table 1.

**T a b l e 1. Characteristics of ground tire rubber**

| Components | Mass contents, % | Methods of testing |
|---|---|---|
| Acetone extract | 8.7 | PN- 92/C04219 |
| Rubber addities | 15.3 | TGA |
| Rubber (SBR, NR) | 48.7 | TGA |
| Carbon black | 32.7 | TGA |

Microgranules of high density polyethylene (PE-HD) were obtained in the process of size-reduction ($MFR_{190\ ^{\circ}C/2.16\ kg}$: 23.1 g/10 min) of post-production scrap purchased from Kalman-Plastics, Warsaw (Poland).

### Sample preparation

Post-production scrap of high density polyethylene was mixed with ground rubber at the 50:50 mass ratio using a co-rotating twin screw extruder (Bühler BTSK

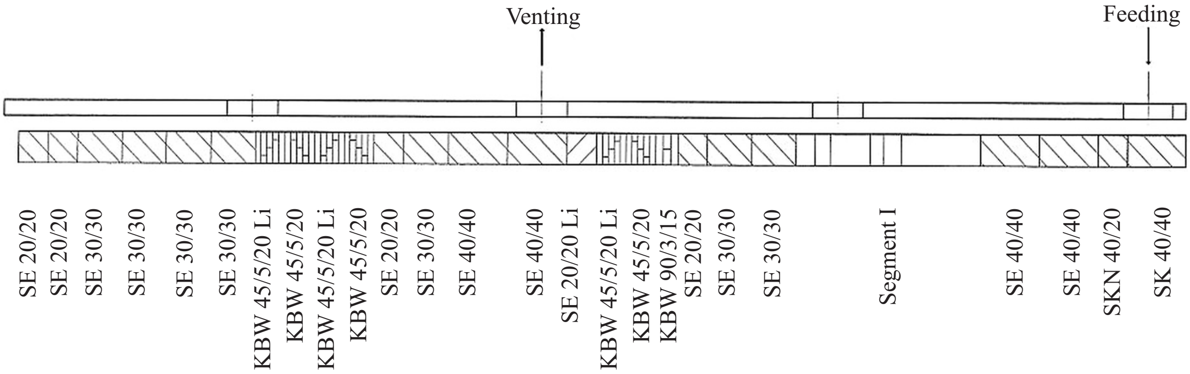


**Fig. 1. Screw configuration**

20/40, Germany). The screw configuration was presented in Fig. 1. Used kneading and mixing elements generated the high shear stress on the processed materials, which allowed to perform effective mixing. Ground tire rubber and PE-HD microgranulate were added via a gravimetric dosing device (Inno-Plast GmbH, Germany) with variable feed rate ranging from 1 to 2.5 kg/h. The other variables were: the barrel temperature ranging from 160 to 240 °C, and rotational speed of the screws between 250 and 650 rpm. The prepared thermoplastic compositions were compression molded into 2 mm thick samples at 180 °C and under the pressure of 4.9 MPa.

## Measurements

In this study the experimental methodology described by Myers and Montgomery [32] was used to design mathematical models for determining the variability of mass flow rate and mechanical properties of compositions based on recycled PE-HD and ground tire rubber. The considered set of independent variables (adjustable settings) included: the barrel temperature ($T$ in °C), rotational speed of the screws ($n$ in rpm) and feed rate ($Q$ in kg/h). The dependent variables (resultant quantities) included the following mechanical and processing properties of the studied compositions: tensile strength at maximum stress ($TS$ in MPa); elongation at break ($E_b$ in %); hardness ($H$ in °Sh D); and the mass flow rate ($MFR$ in g/10 min). The two-level fractional factorial design with addition of center points replication was used in the experiments [32]. The design of the experiment is presented in Table 2. Based on the assumptions of the design chosen, measurements of the mass flow rate and mechanical properties, the empirical models were constructed.

— Mass flow rate ($MFR$) was determined with a Dynisco LMI 4003 capillary rheometer (Germany) according to PN-ISO 1133.

— Tensile strength at maximum stress ($TS$) and elongation at break ($E_b$) were measured using a ZWICK Z020 Materials Testing Machine (Germany) at the speed of 50 mm/min, in accordance with PN-ISO 527. Shore D hardness ($H$) was determined with a ZWICK model 3131 durometer (Germany) according to PN-ISO 868.

— Dynamic mechanical properties of the samples were tested with a TA Instruments model DMA Q800 analyzer (USA). Samples cut to the dimensions of 40 × 10 × 2 mm were loaded with a variable sinusoidal deformation force in the single cantilever bending mode at the frequency of

**T a b l e 2. Effect of extrusion conditions on processing parameters, mass flow rate and mechanical properties of thermoplastic compositions**

| Run | $n$, rpm | $T_b$, °C | $Q$, kg/h | $M$, Nm | $E_c$, kJ/kg | $T_m$, °C | $p_m$, bar | $MFR$ g/10 min | $TS$, MPa | $E_b$, % | $H$, °Sh D |
|---|---|---|---|---|---|---|---|---|---|---|---|
| PR1 | 250 | 160 | 1.00 | 17.4±0.4 | 3240±72 | 167±3 | 15.3±0.7 | 7.0±0.1 | 10.0±0.6 | 8±1 | 46±1 |
| PR2 | 650 | 160 | 1.00 | 14.6±0.2 | 7020±144 | 176±2 | 12.9±0.8 | 7.4±0.3 | 10.08±0.04 | 9±1 | 43±2 |
| PR3 | 250 | 240 | 1.00 | 12.8±0.6 | 2484±108 | 252±1 | 8.2±0.4 | 9.5±0.2 | 10.0±0.3 | 7.6±0.3 | 44±2 |
| PR4 | 650 | 240 | 1.00 | 11.6±0.3 | 5472±144 | 254±1 | 6.4±0.5 | 13.3±0.2 | 8.7±0.2 | 6.3±0.6 | 40±2 |
| PR5 | 250 | 160 | 2.50 | 27.9±0.3 | 2002±14 | 172±1 | 24.0±0.9 | 5.1±0.1 | 8.8±0.1 | 8.9±0.6 | 40±2 |
| PR6 | 650 | 160 | 2.50 | 20.3±0.3 | 3902±58 | 177±2 | 18.7±0.5 | 6.5±0.1 | 9.3±0.3 | 8±1 | 44±2 |
| PR7 | 250 | 240 | 2.50 | 18.5±0.6 | 1354±43 | 252±1 | 13.2±0.4 | 6.8±0.3 | 8.8±0.2 | 7.8±0.5 | 42±1 |
| PR8 | 650 | 240 | 2.50 | 14.2±0.3 | 2707±72 | 255±2 | 9.8±0.8 | 9.8±0.3 | 9.2±0.5 | 7±1 | 42±1 |
| C | 450 | 200 | 1.75 | 19.0±0.1 | 3621±29 | 214±1 | 13.6±0.4 | 6.0±0.4 | 8.7±0.4 | 7.8±0.5 | 42±1 |

$n$ — screw speed, $T_b$ — barrel temperature, $Q$ — feed rate, $M$ — torque, $E_c$ — unit energy consumption, $T_m$ — temperature of composition in extrusion die, $p_m$ — pressure of composition in extrusion die, $MFR$ — mass flow rate, $TS$ — tensile strength, $E_b$ — elongation at break, $H$ — hardness.

1 Hz under the temperature rising rate of 4 °C/min within the range between -100 °C and 100 °C.

## RESULTS AND DISCUSSION

The observations of the dependent variables $TS$, $E_b$, $H$ and $MFR$ are presented in Table 2. It was postulated and then correctly verified that the relationship between process variables $n$, $T$, $Q$ and responses η (η stands for $TS$, $E_b$, $H$ or $MFR$) could be approximated by a polynomial equation:

$$\eta(n,Q,T) = a_0 + a_1 n + a_2 Q + a_3 T + a_{12} nQ + \\ + a_{23} QT + a_{13} nT + a_{123} nQT$$

It consists of the main effects ($a_n$ — constant and linear terms, n = 1, 2, 3 etc.) and interactions terms. The general idea pertaining to a polynomial approximation is the Taylor series expansion. When the models revealed excessive nonlinear effect, an additional term accounting for a pure curvature was included.

The collected measurements were used to estimate the coefficients of regression models. Further tests such as: residual analysis, a test for significance of regression as well as tests concerning individual regression coefficients were conducted to verify model building assumptions and to assess the adequacy of postulated effects. The results of the analysis are presented in Tables 3—6. The Pareto analysis of the standardized effects was used to present clearly the results of statistical analysis (see Fig. 4), namely to compare particularly effects with each other.

**T a b l e 3. Model of mass flow rate — ANOVA table. $R^2 \approx R_{adj}^2$ = 0.98**

| Source of variation | SS | df | MS | $F_o$ | P |
|---|---|---|---|---|---|
| Curvature | 59.40 | 1 | 59.39 | 815.43 | 0.000 |
| n | 47.42 | 1 | 47.42 | 651.05 | 0.000 |
| T | 110.84 | 1 | 110.84 | 1521.69 | 0.000 |
| Q | 49.61 | 1 | 49.60 | 681.05 | 0.000 |
| nt | 15.66 | 1 | 15.66 | 214.95 | 0.000 |
| nQ | 0.04 | 1 | 0.04 | 0.61 | 0.438 |
| TQ | 6.96 | 1 | 6.96 | 95.60 | 0.000 |
| nTQ | 2.03 | 1 | 2.03 | 27.91 | 0.000 |
| Error | 3.71 | 51 | 0.07 | | |
| Total SS | 295.68 | 59 | | | |

SS — sum of squares, df — degrees of freedom, MS — mean squares, $F_o$ — Snedecor statistics and P — the smallest level of significance. The arbitrarily chosen value of the significance level for a statistical test, i.e. α = 0.05.

### Model of mass flow rate

The modelling of mass flow rate ($MFR$) showed that barrel temperature ($T$) was the most important factor (Fig. 2 and Table 3). The increased barrel temperature caused partial thermomechanical degradation of polyethylene and ground tire rubber, which has influence on increasing values of this parameter. The other two factors, i.e. rotational speed ($n$) and extrusion feed rate ($Q$) showed similar numerical effect on the shape of response surface. The increase in rotational speed was associated with increasing fluidity of the composition. Higher feed rate during extrusion causes decrease in mass flow rate ($MFR$) of obtained products. The modelling results also demonstrated the presence of two statistically significant interactions, namely the interaction between rotational speed and temperature ($n$ and $T$ — positive effect) and the interaction between temperature and extrusion feed rate ($T$ and $Q$ — negative effect). In addition, the $MFR$ model was characterized by the significant negative effect of curvature. The change in mass flow rate ($MFR$) is correlated with processing parameters (screw torque, melt temperature and pressure) as it was presented in Table 2. Decrease of screws torque and increase of energy consumption were caused by a change in mass flow rate ($MFR$) of the obtained products. This phenomenon may be explained by a tendency of the degraded material (high shear force and temperature) to stick to the screws, which causes problems with extrusion of dosed material and increase of the energy consumption per 1 kg. Higher feed rate of extrusion (more filled space between screws

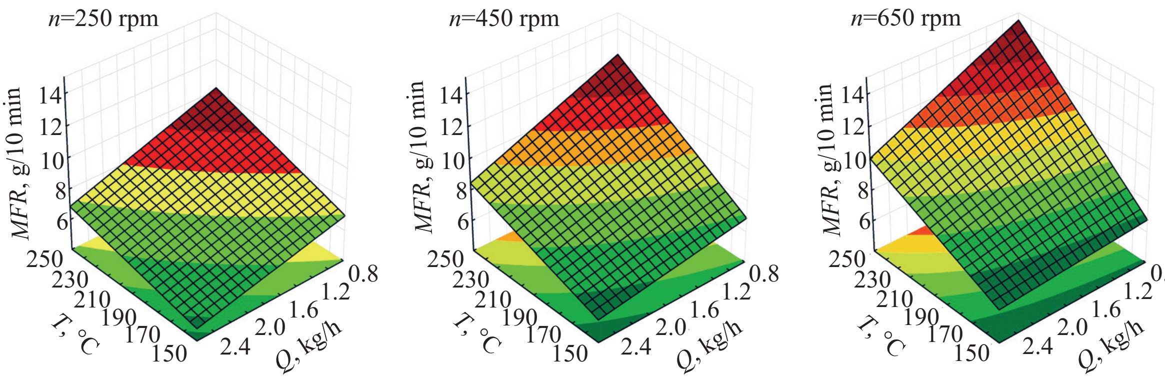


**Fig. 2. Response surface plots of mass flow rate as a function of extrusion conditions**

and barrel) caused better mixing and transport of material during extrusion.

## Tensile strength model

Based on the conducted research, it can be concluded that barrel temperature (*T*) had the strongest and decisive effect on tensile strength (Fig. 3 and Table 4). A similar variability pattern was observed for the relationship between tensile strength and extrusion feed rate (*Q*). The varying rotational screw speed (*n*) was not associated with statistically significant changes in the tensile strength, though the variable rotational speed appeared in two significant interactions, i.e. between *n* and *Q*, and *n* and *T*. Both interactions statistically significant as depicted by the characteristic shape of response surface (Fig. 3) and Pareto analysis (Fig. 4). Moreover, the statistical analysis showed that the model of tensile strength has a negative effect of the curvature (Fig. 3, Table 4). Because of the experimental design type chosen, only the detection of such an effect was possible. The exact estimation of coefficients of the pure quadratic terms could not be performed.

**T a b l e 4. Model of tensile strength — ANOVA table. $R^2 \approx R_{adj}^2 = 0.72$**

| Source of variation | SS | df | MS | $F_o$ | P |
|---|---|---|---|---|---|
| Curvature | 3.66 | 1 | 3.66 | 26.30 | 0.000 |
| n | 0.04 | 1 | 0.04 | 0.31 | 0.580 |
| T | 0.67 | 1 | 0.67 | 4.83 | 0.037 |
| Q | 2.91 | 1 | 2.91 | 20.88 | 0.000 |
| nT | 0.79 | 1 | 0.79 | 5.68 | 0.024 |
| nQ | 1.63 | 1 | 1.63 | 11.71 | 0.002 |
| TQ | 0.43 | 1 | 0.43 | 3.099 | 0.089 |
| Error | 3.76 | 27 | 0.14 | | |
| Total SS | 13.91 | 34 | | | |

## Model of elongation at break

The analysis of measurements presented in Table 5 shows that barrel temperature (*T*) is the only statistically significant factor affecting elongation at break. The values of elongation at break decrease with increasing barrel temperature. The other factors, i.e. rotational screw speed (*n*) and extrusion feed rate (*Q*) did not affect the shape of response surface and hence they were not illustrated on any plots.

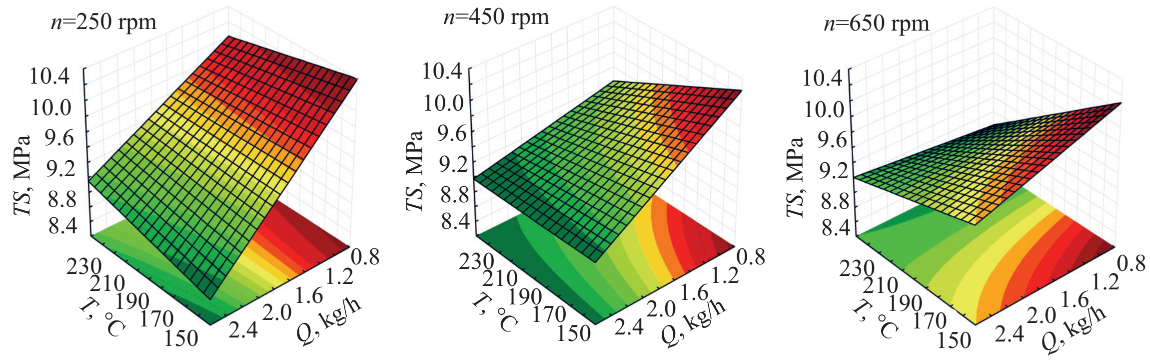


**Fig. 3. Response surface plots of tensile strength as a function of extrusion conditions**

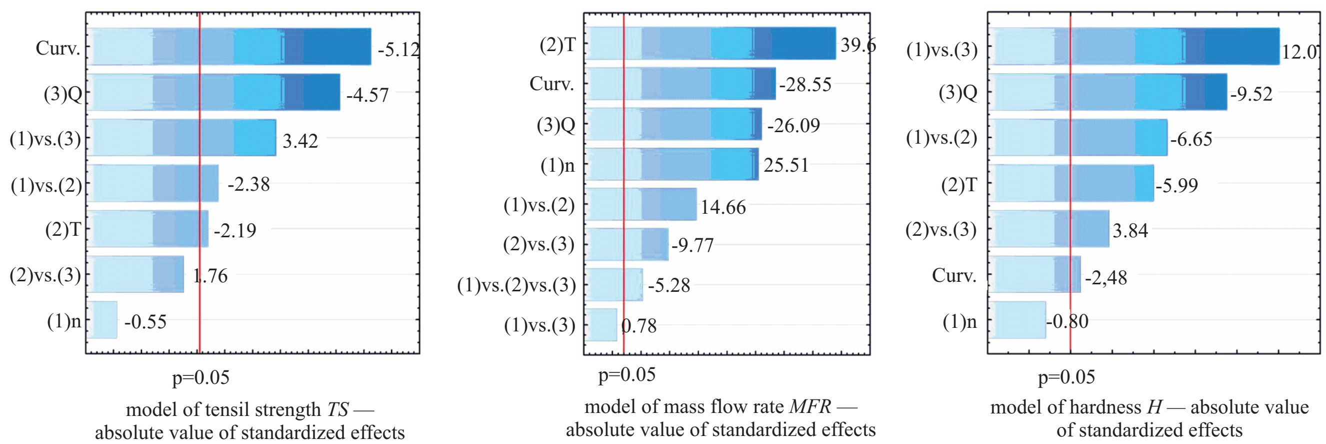


**Fig. 4. Pareto analysis of the standardized effects of empirical models *TS*, *MFR*, *H*; the vertical line in each Pareto plot corresponds to the arbitrarily chosen level of significance (p = 0.05)**

**T a b l e 5. Model of elongation at break — ANOVA table. $R^2$ = 0.45; $R_{adj}^2$ = 0.31**

| Source of variation | SS | df | MS | $F_o$ | P |
|---|---|---|---|---|---|
| Curvature | 0.032 | 1 | 0.03 | 0.06 | 0.812 |
| n | 1.42 | 1 | 1.42 | 2.54 | 0.122 |
| T | 8.86 | 1 | 8.86 | 15.82 | 0.000 |
| Q | 0.30 | 1 | 0.30 | 0.53 | 0.471 |
| nt | 0,87 | 1 | 0.87 | 1.55 | 0.224 |
| nQ | 0.42 | 1 | 0.42 | 0.75 | 0.393 |
| TQ | 0.88 | 1 | 0.88 | 1.57 | 0.220 |
| Error | 15.19 | 27 | 0.56 | | |
| Total SS | 27.89 | 34 | | | |

## Hardness model

The results of hardness modelling were presented in Fig. 5 and Table 6. The hardness of investigated thermoplastic compositions showed that the strongest effect on the response surface was associated with the interaction between two factors, i.e. rotational speed and extrusion feed rate (*n* and *Q*). This effect introduced a significant curvature in response surface H (*n*, *Q*, *T*). Another factor affecting hardness was extrusion feed rate (*Q*). Hardness of the investigated compositions decreased with increasing extrusion feed rate. The remaining factors, i.e. barrel temperature (*T*) and the interaction between rotational screw speed and temperature (*n* and *T*) had a negative effect of similar magnitude on hardness of PE-HD ground tire rubber compositions.

**T a b l e 6. Model of hardness — ANOVA table. $R^2$ = 0.79; $R_{adj}^2$ = 0.78**

| Source of variation | SS | df | MS | $F_o$ | P |
|---|---|---|---|---|---|
| Curvature | 5.82 | 1 | 5.82 | 6.02 | 0.016 |
| n | 0.61 | 1 | 0.61 | 0.63 | 0.43 |
| T | 33.84 | 1 | 33.84 | 34.96 | 0.000 |
| Q | 85.50 | 1 | 85.50 | 88.33 | 0.000 |
| nt | 41.72 | 1 | 41.72 | 43.09 | 0.000 |
| nQ | 136.73 | 1 | 136.73 | 141.25 | 0.000 |
| TQ | 13.95 | 1 | 13.95 | 14.0 | 0.000 |
| Error | 78.41 | 81 | 0.97 | | |
| Total SS | 387.89 | 88 | | | |

## Dynamic mechanical analysis

Figure 6 shows the results of the dynamic mechanical analysis (DMA) of thermoplastic compositions (samples: PR1, PR3, PR4 and C). The maximum values of the loss tan δ have shifted towards the higher temperature values in response to increasing barrel temperature and shear forces (rotational screw speed) during extrusion. This phenomenon can be caused by different reclaiming degree of ground tire rubber, cyclization reaction of styrene-butadiene rubber (main component of ground tire rubber) [33] and higher volatilization of low molecular weight ingredients content in GTR [34] at elevated temperature. Increase of barrel temperature has negligible influence on shift of alpha transition temperature of polyethylene, which corresponded with its degree of crystallinity [35].

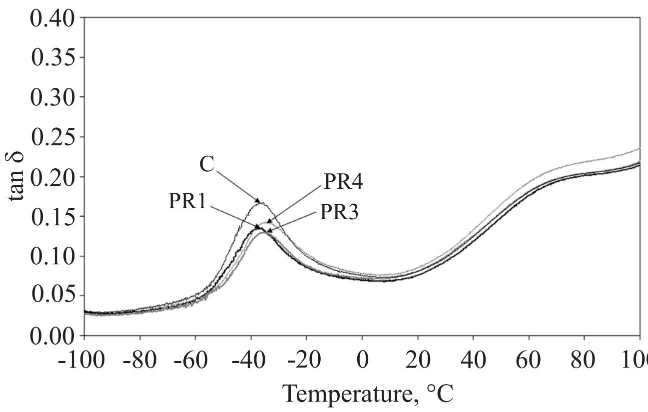


**Fig. 6. Loss tangent as a function of temperature plotted for samples PR1, PR3, PR4 and C**

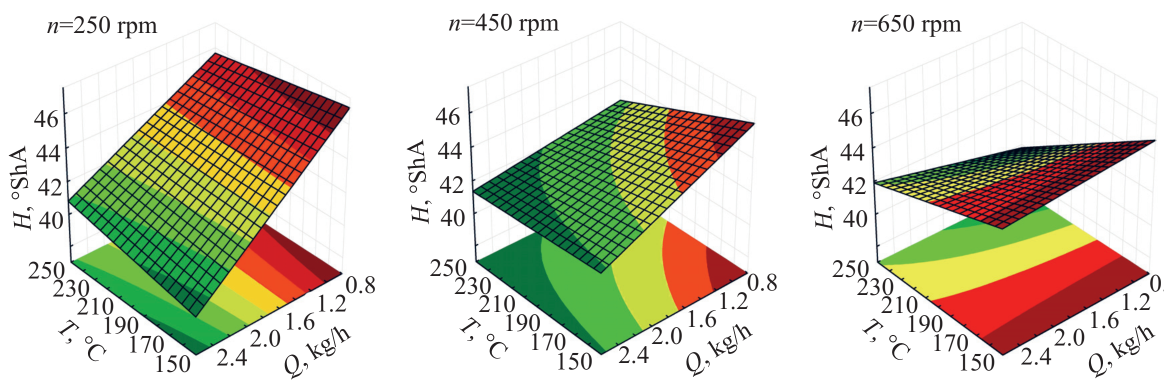


**Fig. 5. Response surface plots of hardness as a function of extrusion conditions**

## CONCLUSIONS

In this study thermoplastic compositions based on ground tire rubber and post-production polyethylene was produced using a co-rotating twin screw extruder. Based on the results of this study it can be stated that barrel temperature (*T*) had the strongest effect on the quality of the produced thermoplastic compositions. Thermal degradation of polyethylene as well as partial scission of cross-link bonds and degradation of elastomers present in ground tire rubber were observed with increasing barrel temperature. This subsequently resulted in increased mass flow rate and decreased mechanical properties, i.e. tensile strength, elongation at break and hardness of the prepared thermoplastic compositions. Dynamic mechanical analysis shows that barrel temperature has influence on cross-link density of obtained compositions, which corresponds with change in structure of ground tire rubber.

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