The influence of screw configuration and screw speed of co-rotating twin screw extruder on the properties of products obtained by thermomechanical reclaiming of ground tire rubber

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Abstract: The results of our investigations on the process of continuous thermomechanical reclaiming of ground tire rubber (GTR) carried out using a twin screw extruder are presented. We used a co-rotating twin screw extruder with a special configuration of plasticizing unit, enabling generation of considerable shear forces. The influence of screw configuration and screw speed on breaking of chemical crosslink bonds contained in ground tire rubber are characterized by extrusion parameters, the acetone extract content, the content of the soluble fraction, the degree of reclaiming and the crosslink density of the obtained products. The effect of a secondary curing on the mechanical properties (tensile strength, elongation at break, hardness, resilience and abrasion resistance) of the obtained materials was also determined. It has been established that the thermomechanical reclaiming process depends on shear forces acting on the rubber particles, while the degree of reclaiming increase with the increasing screw speed. A change in screw configuration significantly affects the processing of ground tire rubber during reclaiming process. For example, preliminary pulverization of the fine rubber particles in the first zone of barrel facilitates further processing in the twin screw extruder, which is beneficial in order to decrease the screw torque and the energy consumption in the production of reclaimed rubber. Thermogravimetry analysis (TG/DTG) showed that thermal stability of obtained reclaimed GTR depends strongly on the screw configuration.

Keywords: rubber waste, reclaiming, twin screw extruder, screw configuration, thermogravimetric analysis.

Wpływ konfiguracji oraz prędkości obrotowej ślimaków na właściwości produktów termomechanicznej regeneracji odpadów gumowych prowadzonej przy użyciu współbieżnej wytłaczarki dwuślimakowej

Streszczenie: Zbadano proces termomechanicznej regeneracji ciągłej rozdrobnionych odpadów gumowych, prowadzony przy użyciu wytłaczarki dwuślimakowej współbieżnej o specjalnej konstrukcji układu uplastyczniającego generującego bardzo duże siły ścinające miał gumowy. Scharakteryzowano wpływ konfiguracji ślimaków (przesunięcia segmentu rozcierającego) oraz prędkości obrotowej ślimaków na przebieg procesu regeneracji, zawartość ekstraktu acetonowego i frakcji rozpuszczalnej, stopień regeneracji oraz wyniki analizy termograwimetrycznej otrzymanych produktów. Określono wpływ wtórnej wulkanizacji uzyskanych regeneratów gumowych na właściwości mechaniczne wytworzonych materiałów.

Słowa kluczowe: odpady gumowe, regeneracja, wytłaczarka dwuślimakowa, konfiguracja ślimaków, analiza termograwimetryczna.

A dynamic development of the automotive industry has significantly contributed to the huge amount of waste rubber produced worldwide of which 80 % is discarded as automobile tires. Annually, more than 17 million used tires are generated around the world of which almost 3.3 million belong to the EU [1]. According to estimates made by the Department of Elastomers and Rubber Technology of the Institute of Polymer Materials Engineering and Dyes (Poland), about 200 000 tons of waste tires are generated each year in Poland which puts the country in sixth place within the EU [2]. The legislation obliging manufacturers to manage end-of-life tires, while simultaneously banning tires from landfills aroused the industry interest in technologies that make use of the recycled rubber products. Among the main trends in the waste rubber

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recycling is grinding of waste rubber [3—5] and its further application in particulate form, *e.g.* as filler in thermoplastic compositions [6, 7], rubber compounds [8, 9] or thermoplastic elastomers [10, 11]. Other solutions are pyrolysis [12, 13] and reclaiming [14, 15], both consisting of further treatment of the ground rubber waste.

A relatively new form of the waste rubber recycling is continuous reclaiming in a extruders which requires determination of the relationships between the processing parameters and properties of the resulting reclaimed rubber.

Sutanto *et al.* [16, 17] conducted extensive studies to determine the kinetics of the reclaiming of EPDM vulcanizates. Jalilvand *et al.* [18] characterized the effect of barrel temperature, screw speed and reclaiming agent (disulfide) on the reclaiming of EPDM compound. Maridass and Gupta [19, 20] characterized the effect of the working parameters of a counter-rotating twin screw extruder during the process of reclaiming of ground tire rubber. Tzoganakis [21] patented the thermomechanical reclaiming of waste rubber in the presence of a supercritical fluid conducted in co-rotating twin screw extruder.

Isayev *et al.* published many research works [22—24] about continuous reclaiming of waste rubber conducted in single screw extruder with ultrasounds as source of heat. Reclaiming with using of ultrasounds is quick, simple, efficient method which does not require any solvent. Limited use of this method is due by the high cost of industrial apparatus.

The first information on continuous reclaiming of waste rubber conducted in a twin screw extruder was presented in the late 1990s by a research team working at Toyota [25]; it was a description of an innovative method for the continuous reclaiming of sulfur vulcanized EPDM. Based on the study results, Mouri *et al.* [26] demonstrated that the configuration and geometry of the plasticizing unit are important factors of reclaiming process which together with the processing parameters allow obtaining reclaimed materials with the desired properties.

Fukumori *et al.* [27] investigated the application of the products resulting from continuous reclaiming of waste rubber from used tires. Truck tires produced from rubber compound containing 10 wt. % of reclaimed natural rubber were tested after 200 000 km of use; they possessed properties comparable with those of commercial tires.

Parasiewicz *et al.* [28] presented the results of studies on the effect of barrel temperature distribution on the rubber reclaiming process. Pilot-scale tests (feed rate = 40 kg/h) were carried out in a co-rotating twin screw extruder prototype, produced by the Institute of Polymer Materials Engineering and Dyes (Poland), with a unique screw configuration that provides different shear stress in barrel. The best properties of the reclaimed rubber were obtained with the following barrel temperature profile (from hopper to extrusion die): 170/180/185/190/130/110/90 °C.

Yazdani *et al.* [29] studied the effect of temperature and screw speed on the continuous reclaiming process which was carried out at the laboratory in a twin screw extruder (Brabender TSE 20/40, feed rate = 1 kg/h) with a special screw design which combines mixing and kneading segments that generate a considerable pressure during the reclaiming of ground tire rubber. The study results showed a clear influence of screw speed (shear forces) on the degree of reclaiming carried out at temperatures between 220—280 °C.

The aim of this present study was to characterize the effect of the plasticizing unit design containing a special kneading segment and of the screw speed on the quality of the resulting reclaimed materials. We investigated the extruder parameters, the acetone extract and sol fraction contents, the degree of reclaiming and thermal properties of the reclaimed materials. The mechanical properties of the obtained reclaimed rubber mixed with curing system were also determined.

EXPERIMENTAL PART

Materials

Ground tire rubber (GTR), 1.5 mm fraction, from whole used tires was prepared by ABC Recykling (Poland). The characteristic of ground tire rubber are presented in Table 1.

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

Properties	Mass contents, %	Methods
Acetone extract	8.7	PN-92/C-04219
Rubber additives	15.3	TGA
Rubber (SBR, NR)	48.7	TGA
Carbon black	32.7	TGA

Solvents (acetone, toluene) for chemical analyses and the curing agents for vulcanization (stearic acid, sulfur, zinc oxide, TBBS-*N*-tertbutyl-2-benzothiazyl sulfenamide) were purchased from POCh S.A. and „STANDARD" Sp. z o.o. (Poland).

Sample preparation

Thermomechanical reclaiming of ground tire rubber was performed using a Bühler's BTSK 20/40 twin screw extruder with a special screw design, as shown in Fig. 1. Segment I contains a unique system of kneading segments which allows generation of significant shear forces which has enabled decrease of the barrel temperature during reclamation process. Reclaiming process was conducted at 180 °C, this temperature was constant in whole barrel of extruder. Rubber waste was fed through a volumetric screw feeder at a constant rate. The second investi-

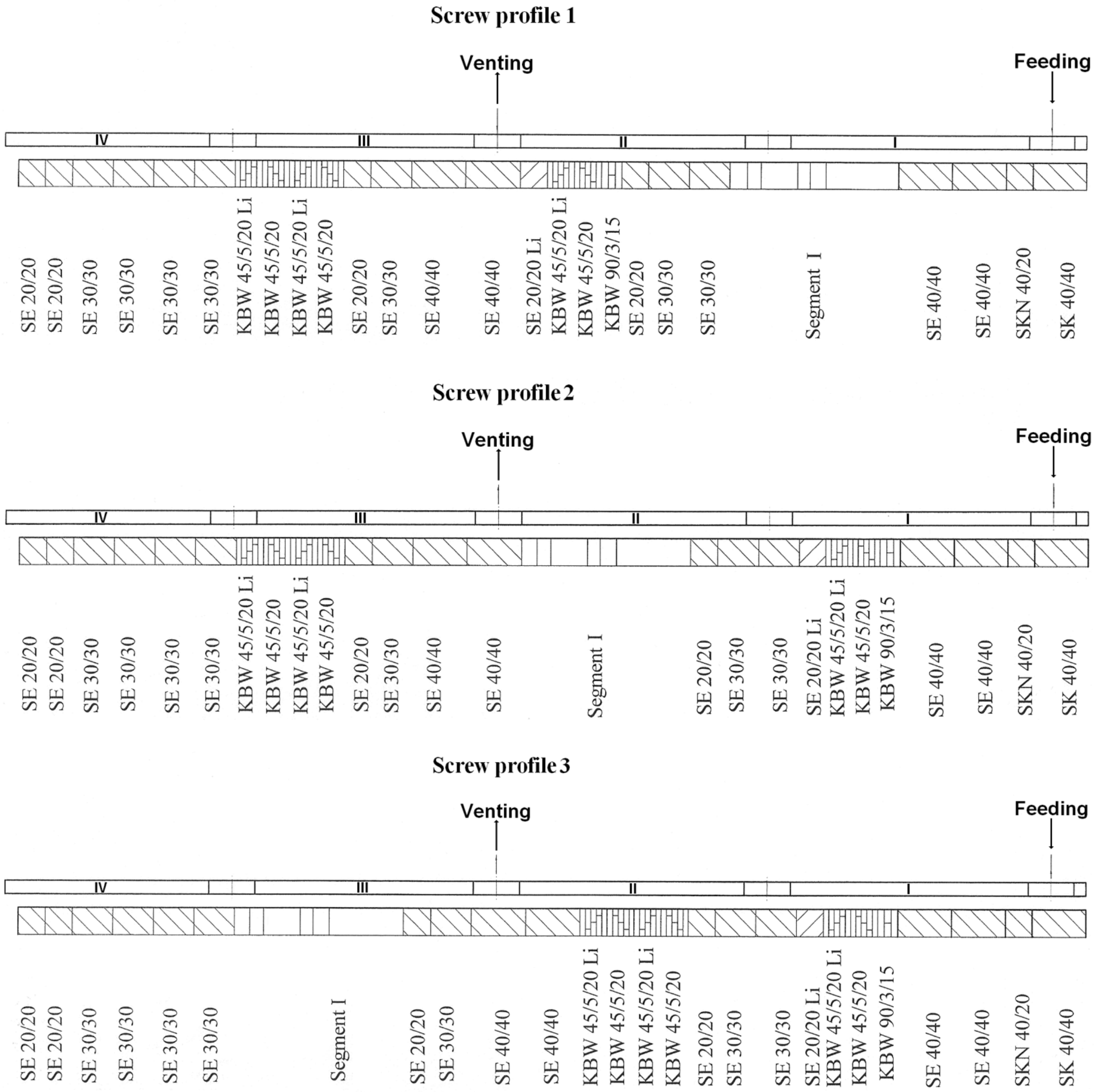

Fig. 1. Schematic view of screw configuration

gated variable was screw speed which had been set to 200, 300, 400, 500 and 600 rpm. Due to overload (very high torque), the machine failed to produce samples at the speed of 200 rpm for screw configurations 2 and 3. In order to evaluate the resulting reclaimed material, a rubber compound with the following composition (parts by mass) was prepared: 100.0 parts reclaimed rubber, 2.5 parts ZnO, 1.0 part stearic acid, 1.5 parts sulfur, and 0.35 part TBBS. The compound was vulcanized at 150 °C under 4.9 MPa pressure in curing press for the optimum vulcanization time determined according to PN-ISO 3417:1994 standard.

Methods of testing

In order to control the conditions of thermomechanical reclaiming process, the effect of screw configuration and screw speed on torque was determined, and energy consumption of the extruder drive shaft was measured. Samples were subjected to a preliminary extraction with acetone in order to remove low molecular weight substances (48 h, at room temperature, using discontinuous cold extraction).

— The crosslinking density of the obtained samples was determined by equilibrium swelling in toluene (72 h, at room temperature, using discontinuous cold extraction), according to the Flory-Rehner equation [30] without Kraus correction (based on ASTM D 6814):

$$\nu_e = \frac{-[\ln(1-V_r)+V_r+\chi V_r^2]}{[V_1(V_r^{1/3}-V_r)/2]} \tag{1}$$

where: v_e — crosslinking density (mol/cm^3), V_r — volume of the rubber gel in the swollen sample, V_1 — molar volume of solvent (cm^3/mol), χ — polymer-solvent interaction parameter (χ = 0.391 was used in calculations) [31].

— The degree of reclaiming was determined according to the crosslinking density changes in the reclaimed material (v_1) in relation to crosslinking density of the original ground tire rubber (without reclaiming) (v_2) by using the formula:

$$\text{degree of reclaiming} = \frac{\nu_1 - \nu_2}{\nu_1} \cdot 100 \quad (2)$$

— The percentage of sol fraction was determined from the mass difference in reclaimed samples before swelling (W_1) and after solvent removal (W_2) according to the formula:

$$\%\ \text{sol fraction} = \frac{W_1 - W_2}{W_1} \cdot 100 \quad (3)$$

— The obtain reclaimed rubber was subjected to thermogravimetric analysis by means of a NETZCH, TG 209 device. The study was conducted in an argon atmosphere at a heating rate of 10 °C/min.

— The vulcanization process was studied at 150 °C according to the standard PN-ISO 3417:1994. The measurements were performed using a Monsanto R100S vulcameter with an oscillating rotor. The rotor oscillation angle was 3°, while torque ranged between 0 and 100 dNm.

— Tensile strength and elongation at break of the obtained vulcanizates were tested according to PN-ISO 37 standard by using a Zwick Z020 testing machine.

— Shore A hardness was determined with a Zwick 3130 durometer in accordance with ISO 7619-1 standard.

— Abrasion resistance was measured according to ISO 4649.

— The rebound resilience determination was performed using the Schob pendulum according to ISO 4662.

RESULTS AND DISCUSSION

Table 2 contains the results of our study on the effect of screw configuration and screw speed during reclaiming on the processing parameters, the acetone extract and sol fraction contents, crosslinking density and the degree of reclaiming of the obtained samples. Moving towards the head of the plasticizing unit of segment I, which generates strong shear forces, there is a significant increase in torque and energy consumption causing the elevated shear of the rubber particles inside the barrel. An increase in screw speed (shear force) results in a noticeable decrease in torque. It also increases the energy consumption per 1 kg of reclaimed material which is related to the ongoing process of degradation of the sulfide crosslinking bonds and the degradation of bonds in main chains. During the thermomechanical reclaiming process the crosslinked particles of ground tire rubber become more plastic which allows partial flow of the blend and the decrease of the screw torque. More reclaimed/plastic material may have tendency to stick to the screws which cause problems with extrusion of dosed material and increase of the energy consumption per 1 kg of reclaimed material.

A slight increase in the content of acetone extract obtained in reclaimed material is comparable with the values for pure ground tire rubber. This suggests no formation of low molecular weight substances and evaporation of low molecular ingredients from GTR during reclaiming. The soluble fraction content and the degree of reclaiming under test conditions increase with the position shift of segment I in the plasticizing unit and with the increasing speed, which is linked to the scission of crosslinking bonds. A decrease in crosslinking density of the resulting reclaimed materials suggests the breakage of crosslinking bonds and partial degradation of main chains during thermomechanical reclaiming of ground tire rubber. The above mentioned results confirm a significant impact of the plasticizing unit configuration on the quality of the obtained reclaimed materials [27]. The Horikx's theory [32] was used to specify difference between main-chain scission and crosslink scission. According to Horikx's

T a b l e 2. The effect of screw configuration and the rotor speed of extruder screws on reclaiming of GTR

Sample	n, rpm	Screw configuration	Screw torque, Nm	Energy consumption kJ/kg	Acetone extract, %	Sol fraction %	Crosslink density mol/cm^3 · 10^{-4}	Degree of reclaiming, %
P1	200	Profile 1	28.5	2378	9.6	11.8	3.98	50
P2	300		22.4	3022	9.2	13.1	4.46	44
P3	400		21.2	3867	8.2	14.4	3.89	51
P4	500		18.9	4311	8.0	15.5	3.80	52
P5	600		18.1	4956	9.0	15.0	3.80	52
P6	300	Profile 2	29.3	3978	8.2	14.1	4.68	41
P7	400		26.2	4778	7.9	13.6	3.94	50
P8	500		23.2	5289	7.4	14.4	3.81	52
P9	600		21.0	5733	7.4	15.6	3.78	52
P10	300	Profile 3	29.6	4022	8.9	12.1	4.39	45
P11	400		26.4	4800	7.4	14.1	3.94	50
P12	500		23.5	5356	7.8	15.2	3.76	53
P13	600		21.8	5956	8.8	15.7	4.16	48
GTR	—		—	—	8.7	2.4	7.94	0

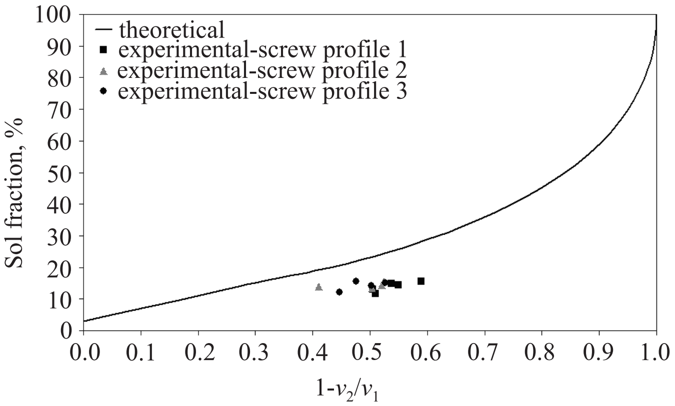

Fig. 2. Sol fraction percentage versus the relative decrease in crosslink density according Horikx theory (for theoretical curve S_1 was 2.3 %)

theory, the data below the chain scission curve mean that the reclaiming process has occurred by selective breakage of crosslink bonds and main chain scission are not present. The results presented on Fig. 2 shows that crosslink bonds of ground tire rubber are broken selectively during thermomechanical reclaiming process conducted in twin screw extruder, but it should be considered that used theory has limitations which do not include the intermediate case between crosslink bonds and main chain scission.

Figure 3 shows samples of reclaimed material taken from screws 1—3 at a screw speed of 300 rpm. As shown in the picture of screw 1/sector I, the presence of fine powder particles confirmed the preliminary pulverization [33] and partial reclaiming of ground tire rubber. In the case of screws 2 and 3, in which the location of kneading segment I was shifted towards the extrusion die, this effect was not observed. A preliminary pulverization of rubber waste facilitates its further processing in the twin screw extruder. It is beneficial for further processing because it decreases the screw torque and energy consumption during the production of reclaimed rubber. Table 3 contains the values of acetone extract and sol fraction contents, crosslinking density and the degree of reclaiming of samples taken from each zone of the screws, as illustrated in Fig. 4.

The slight fluctuations in the content of acetone extract obtained were probably due to the complex and heterogeneous nature of the waste rubber powder originating from used car tires. The acetone extract content values were comparable with those obtained for origin waste powder, which suggests no formation of low molecular weight substances during reclaiming. A significant increase in the sol fraction and the degree of reclaiming as well as a decrease in crosslinking density was observed which confirms the occurrence of reclaiming. For all the screws, an increase in crosslinking density of obtained reclaimed GTR was determined in zone IV. The surface temperature of reclaimed material after exiting from extrusion die was about 10—15 °C higher than the set value (180 °C in whole barrel), which was due by shear stress and heat transfer acting on ground tire rubber feeding into extruder. This may be related to the fact that increasing temperature promotes the cyclization reaction of styrene-butadiene rubber, a material commonly used in the production of automobile tires [34].

T a b l e 4. Mass loss (Δm), temperature range (ΔT) from TG curves and the peak position (T_p) from DTG curves for reclaimed GTR (rotor speed of extruder screws = 300 rpm)

Sample	ΔT, °C	Δm, %	T_p, °C
GTR without treatment	200—350	14.7	—
	350—400	25.6	373
	400—550	21.4	413
GTR profile 1	200—350	12.6	—
	350—400	23.7	369
	400—550	23.4	419
GTR profile 2	200—350	13.4	—
	350—400	25.7	372
	400—550	25.7	422
GTR profile 3	200—350	11.8	—
	350—400	25.4	376
	400—550	28.1	424

Figure 5 and Table 4 present the results of the thermogravimetric analysis and differential thermogravimetric analysis for the reclaimed rubber materials sampled from three different screw profiles. Based on the TGA and DTG curves for the various configurations of the plasticizing unit and the rubber powder before reclaiming, it was possible to determine the effect of the screw configuration on the process of rubber powder reclaiming. Several clear differences between the TGA and DTG curves were noted for screw configuration 1 and screws 2—3 which showed the same trend (Table 4). The

T a b l e 3. The effect of screw configuration and barrel zone (presented in Fig. 2 and 3) on reclaimed GTR

Screw configuration	Acetone extract, %				Sol fraction, %				Crosslink density, mol/cm^3 · 10^{-4}				Degree of reclaiming, %			
	I	II	III	IV	I	II	III	IV	I	II	III	IV	I	II	III	IV
Profile 1	10.3	9.7	9.4	9.2	6.4	12.9	12.6	13.1	4.22	4.74	4.13	4.46	46	40	47	44
Profile 2	8.9	9.9	9.1	8.2	4.9	11.5	11.0	14.1	5.38	4.09	3.55	4.68	32	48	55	41
Profile 3	8.9	9.3	10.7	8.9	4.3	12.3	11.9	12.1	5.24	4.17	3.73	4.39	34	47	53	45

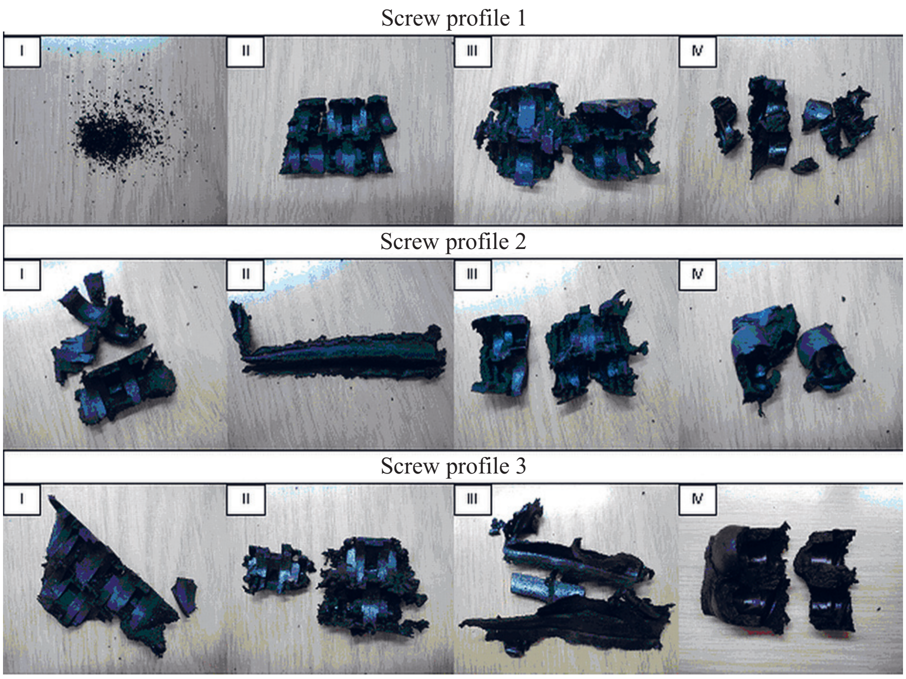

Fig. 3. Reclaimed GTR from zones I—IV of the barrel (rotor speed of extruder screws was 300 rpm)

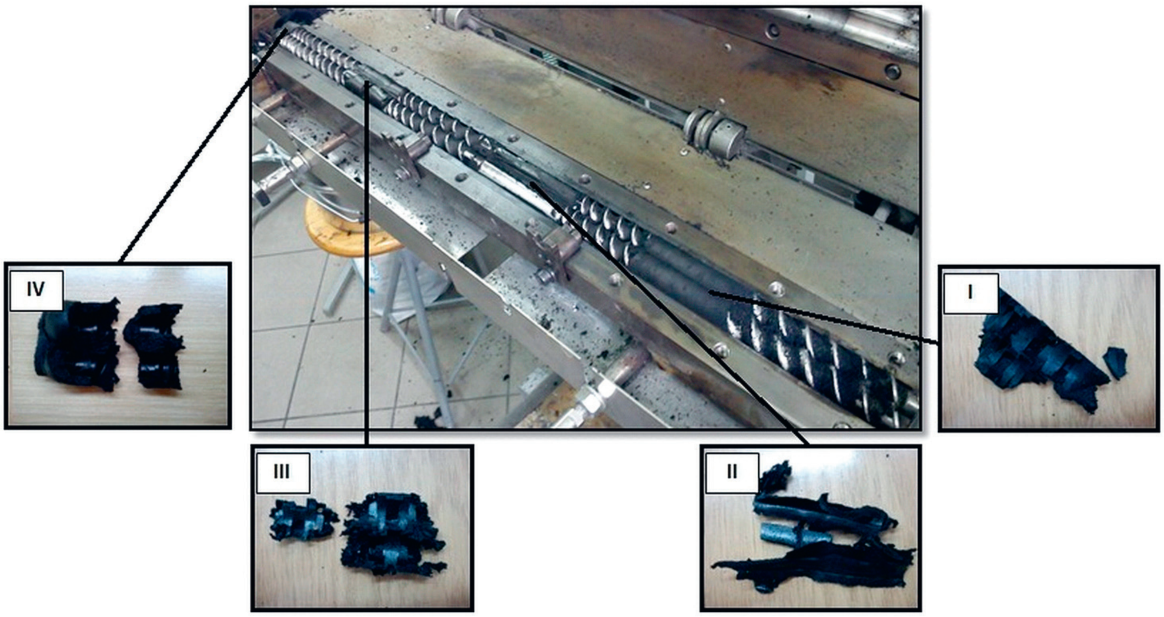

Fig. 4. Sampling of reclaimed GTR from different barrel zones (screw profile 2 was used in this picture)

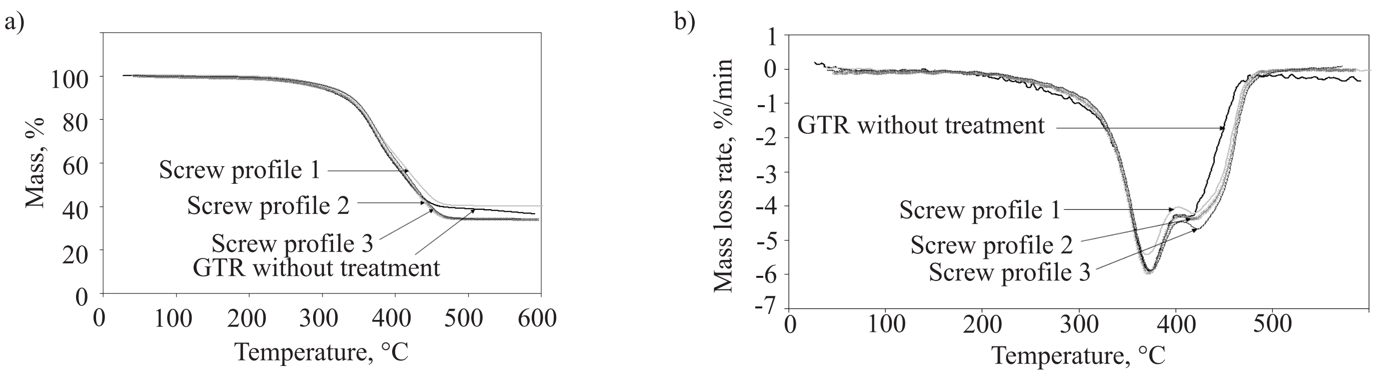

Fig. 5. TG — a) and DTG — b) curves obtained for origin GTR (GTR without treatment) and samples after thermomechanical reclaiming conducted in different screw profiles (rotor speed of extruder screws was 300 rpm)

T a b l e 5. Curing characteristics of reclaimed rubber vulcanized at 150 °C

Sample	n, rpm	Screw configuration	Parameters of vulcanization curves				
			torque rheometer		ΔM	t_2, min	t_{90}, min
			M_{min}, dNm	M_{max}, dNm			
P1	200	Profile 1	7.7	32.2	24.5	2.9	10.4
P2	300		6.3	30.9	24.6	2.9	10.5
P3	400		5.6	30.3	24.7	2.5	10.6
P4	500		6.2	29.2	23.0	2.6	10.3
P5	600		5.9	27.8	21.9	2.8	10.1
P6	300	Profile 2	8.0	36.6	28.6	3.2	11.4
P7	400		7.1	29.7	22,6	3.4	11.0
P8	500		6.5	29.4	22,9	3.3	11.5
P9	600		5.8	28.4	22,6	3.2	12.1
P10	300	Profile 3	6.5	32.2	25.7	3.0	11.2
P11	400		5.5	30.0	24.5	3.0	11.5
P12	500		5.5	29.7	24.2	3.2	11.8
P13	600		5.2	28.7	23.2	3.3	11.8

mass loss for the temperature range 200—350 °C was due to either evaporation or decomposition of plasticizers or other low molecular additives present in rubber waste. Also, two other thermal events occurred between 350 and 600 °C. The observed thermal events were probably due to two-stage decomposition of natural and synthetic rubber because these materials have different decomposition temperatures [35]. Visible differences in crosslinking density of the rubber subjected to thermomechanical reclaiming, performed for screw configuration 1 and screws 2—3, confirmed the significant effect of the plasticizing unit configuration on the degree of reclaiming. Despite the heterogeneous composition of the rubber powder obtained from used tires, which affects the quality of the obtained products, the TGA and DTG curves confirmed the crosslinking density values determined by equilibrium swelling (Table 2) and the mechanical properties of the generated revulcanized materials (Table 5). The results confirmed the significant impact of the degree of reclaiming of ground tire rubber on the shape of thermogravimetric curves. This, in turn, confirmed the possibility of characterizing the reclaiming process using this analytic method [36, 37].

The effect of screw configuration and screw speed on the process of vulcanization of the reclaimed materials is presented in Table 5. It was observed that change of screw configuration and the increase of screw speed during the reclaiming of waste rubber is followed by decrease in the minimum and maximum torque, which has been confirmed by decreased crosslinking density of the obtained

T a b l e 6. Mechanical properties of revulcanized rubber*)

Sample	n, rpm	Screw configuration	TS_b, MPa	E_b, %	H, °Sh A	R, %	A, mm^3
P1	200	Profile 1	7.24 ± 0.37	164 ± 6	61	18	145
P2	300		6.82 ± 0.35	173 ± 7	58	16	155
P3	400		6.54 ± 0.05	167 ± 3	58	16	162
P4	500		6.43 ± 0.18	177 ± 3	56	16	179
P5	600		5.41 ± 0.51	164 ± 8	55	16	196
P6	300	Profile 2	6.78 ± 0.22	165 ± 3	57	21	150
P7	400		6.16 ± 0.32	182 ± 7	55	19	166
P8	500		5.42 ± 0.03	168 ± 6	55	18	175
P9	600		5.13 ± 0.19	177 ± 4	54	18	198
P10	300	Profile 3	6.55 ± 0.17	182 ± 5	57	21	150
P11	400		5.60 ± 0.25	179 ± 7	56	19	153
P12	500		5.90 ± 0.11	181 ± 3	56	19	158
P13	600		5.80 ± 0.06	176 ± 3	56	18	165

*) TS_b — tensile strength, E_b — elongation at break, H — hardness, R — resilience, A — abrasion loss.

revulcanizates. The slightly increase of scorch time and optimal time of vulcanization for samples obtained with using screw profile 2 and 3, can be due higher shear stress/heat acting on ground tire rubber than in screw profile 1. Increasing temperature promotes the evaporation of low molecular additives *e.g.* curing accelerator residues content in ground tire rubber. However, the effect of the studied conditions of reclaiming on the optimal time of vulcanization is negligible which has already been confirmed by previous studies [38].

Table 6 lists mechanical properties of the vulcanized materials obtained from reclaimed rubber. Shifting the kneading segment (segment I) towards the extruder head and increasing speed adversely affects the mechanical properties of the obtained revulcanized materials which become worse with increasing degradation of the rubber hydrocarbon backbone. A high degree of reclaiming achieved in rubber under high shear resulted in a decrease of tensile strength, elongation at break, hardness, resilience and abrasion resistance for the majority of samples, which has already been determined in our previous works [38]. In the case of screw profile 3, its effect on screw speed, abrasion and hardness of the tested vulcanizates was negligible.

CONCLUSIONS

In this paper the influence of the plasticizing unit design and screw speed on the quality of the reclaimed rubber obtained by continuous thermomechanical reclaiming in a twin screw extruder with a special screw configuration was assessed. The analysis of the collected data allows formulation of the following statements:

— The reclaiming process depends on shear force acting on the rubber particles, *i.e.* the degree of reclaiming increases with the increasing screw speed.

— Preliminary pulverization of the fine rubber facilitates its further processing in the twin screw extruder, which is beneficial with regard to decreasing the screw torque and energy consumption in the production of reclaimed rubber waste.

— Changing the configuration of the plasticizing unit significantly affects the quality of the products of reclamation.

— Thermogravimetry as an analytical technique can be successfully used to study the thermomechanical reclaiming process of waste tire rubber, which was confirmed by Horikx theory.

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