Advances in Science and Technology Research Journal

Advances in Science and Technology Research Journal, 18(8), 385–402
https://doi.org/10.12913/22998624/195209
ISSN 2299-8624, License CC-BY 4.0

Received: 2024.10.12
Accepted: 2024.10.26
Published: 2024.11.01

Research on the Influence of the Type and Content of Regranulate on Selected Mechanical Properties of the Three-Layer Polyethylene Film Obtained by Blown Film Extrusion

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ABSTRACT

The article presents the results of experimental studies on selected, mechanical properties of three-layer film, produced by blown film extrusion. Three-layer film was developed for making garbage bags. The research was carried out under industrial conditions. The three-layer film tubes tested were made of low and high density polyethylene and differed in raw material composition, including a different content of recycled polyethylene in individual film layers. Static tensile strength, tear strength and puncture strength tests were carried out using the falling dart method. On the basis of the results obtained, the impact of the content of polyethylene regranulate on the composition of the raw material of the multilayer film in the above-mentioned selected mechanical properties was assessed, in relation to the expected values of these parameters, ensuring the correct use of the three-layer film in the production process and obtaining a garbage bag with appropriate utility features. A classification system was prepared to develop recipes for dosing waste materials by weight and combining them with the primary LLDPE material, using different proportions of its share in individual layers. Increasing the degree of utilization of waste materials, by increasing their share in the unit product to 55%, allowed maintaining the mechanical parameters of the film, meeting the defined industry expectations in terms of static tensile strength and tear strength and puncture resistance.

Keywords: blown film extrusion, multilayer film, polyethylene, polyethylene regranulate.

INTRODUCTION

The blown film extrusion process involves extruding a tubular film and immediately inflating it with air of insignificant pressure followed by pulling it through a take-off device. During the blowing process, transverse stretching occurs primarily, while during the pulling phase, stretching occurs in the longitudinally. To control the transverse and longitudinal stretching processes better, the tubular film is cooled directly behind the die in an air stream from the blowing device [1, 2]. There are three variations of the blown film extrusion method: vertically up, vertically down and horizontally. In each of these cases, the blown film extrusion process line consists of a head with slotted ring nozzle, an inflating device, a tubular film flattening device, a receiving device and a winding device [1]. The most common film extrusion is free blowing extrusion vertically upwards. In this method, transverse and longitudinal stretching are controlled most favourably [3]. Vertical downward and horizontal blown film extrusion methods are less commonly used in industrial practice because of the disadvantages and problems of process control [1].

When extruding tubular film, it is essential that the thickness of the film be kept within the correct tolerance around the perimeter and that the local wrinkles and folds occurring along the tubular film are distributed evenly, as these can overlap during winding on the shaft of the winding device and thus impair the quality of the film tube produced [4]. In order to eliminate this disadvantage, a rotary-reverse drive of the winding device is used.

The mechanical properties of tubular foil depend primarily on the temperature of the foil as it leaves the nozzle of the boring head, the degree of stretch longitudinally, the degree of blowing and the thickness of the foil. The film thickness is mainly determined by the mass flow rate of the plastic, the diameter of the extruded tubular film, the amount and pressure of the blowing air and the film take-up speed [1, 5].

The wide range of applications for film tube has led to the need to manufacture such film as multilayer film. Three- and five-layer film is common and the ever-improving blown film extrusion technology now makes it possible to produce eight-, nine- and ten-layer film, achieving single-layer thicknesses of several micrometres. The occurrence of multiple layers is mainly used to increase mechanical strength, increase barrier properties, improve optical properties and achieve thermal shrinkage [1, 3, 6].

It is well known that the recovery of secondary raw materials for reprocessing is of great importance for environmental protection [7]. Recycling effectively reduces the negative environmental impact of used plastic products and promotes sustainability positively [8, 9, 10]. This is particularly evident for products with a short life cycle such as packaging and other everyday products made from polymer film [11], such as rubbish bags. Film reprocessing reduces waste, saves energy and natural resources and reduces greenhouse gas emissions [12]. For general-purpose plastics, including polyolefins, feedstock recycling, as opposed to energy and chemical recycling, plays an important role [13, 14]. The recycling of raw materials allows used plastics to be reused as a useful raw material, after the waste plastics have been sorted, cleaned, shredded and processed into pellets or flakes that can be used to manufacture new products [15]. The recycling process typically deteriorates the functional and processing properties of waste plastics, due to factors such as mechanical and thermal degradation; however, they are still in demand on the processing market due to their relatively low price [16, 17, 18]. Companies often have their own processing lines for recycling defective products, immediately putting the processed waste back into production. The environmental trend, which manufacturers are effectively exploiting by presenting their products as using recycled plastics, also plays an important role [19, 20]. This is particularly evident in the packaging and everyday products industry.

Multilayer film technology effectively lends itself to the use of waste raw materials and, depending on the intended use of the film and the number of layers, the overall regranulate content of such film can be varied over a wide range [21, 22, 23]. It is particularly easy to introduce regranulate into the appropriate layers in blown film extrusion, when the individual plasticising systems of the extruders feeding the extrusion head allow the delivery of simple regranulate or regranulate mixed with virgin plastic [24, 25, 26]. In this way, the performance properties of the film produced can be influenced, ranging from mechanical to barrier and optical properties [27, 28]. A typical example of this would be multilayer film used for rubbish bags, in which the outer layer is made of virgin plastic containing high-quality dyes and fragrances, the middle layer being based largely on regranulate with various additives and the inner layer possibly being a mixture of regranulate and virgin plastic with added odour neutralising substances.

The aim of industrial experimental research was to obtain a three-layer film of low-density polyethylene containing a total of at least 55% by weight of polyethylene regranulate. The regranulate added could be homogeneous or as a mixture of different PE regranulates. Accordingly, dozens of formulations for three-layer film were developed, taking into account the different types of PE regranulate supplied by the film manufacturer at whose premises the research project was carried out. The three-layer film produced had to have sufficient mechanical strength to ensure satisfactory performance downstream, of the rubbish bag made from them. Static mechanical tensile strength, tear strength and puncture resistance were chosen as performance indicators, sufficiently defining the characteristics of the film product. The minimum expected values of the individual strength parameters were determined by reference to their values obtained for bags made entirely of original polyethylene. These values were lower than for the original polyethylenes, but sufficient to provide the right performance characteristics for a rubbish bag that meets customer expectations.

SUBJECT AND PROGRAMME OF EXPERIMENTAL RESEARCH

The subject of the study was three-layer polyethylene film with a total thickness of 25 micrometres and a layer ratio of 30-40-30% of the total thickness. The diameter of the extruded film tube was 827 mm, which, when flattened, gave the width of the wound film tube equal to 1300 mm (Figure 1). During winding, the beam was cut in half into two 650 mm wide sections, later used to make 650x505 mm reference bags. The average film thickness, determined by analysing data from industrial measurements made with the Octagon film thickness measuring device, ranged from up to 23.1 to 29.7 micrometres. The technological parameters of the blown film extrusion process (zone temperatures in the plasticising system and extrusion head, blowing pressure, film tube extraction speed and others) were individually selected for each recipe, while keeping the blown film extrusion process mass output close to production conditions. These parameters, due to the obligation of company secrecy, cannot be published.

Four virgin plastics, which are linear low-density polyethylene LLDPE and twelve regranulates, eight of which were declared by suppliers as low-density polyethylene LDPE and four as high-density polyethylene HDPE, were used to produce the triple layer film. Only for three of the regranulates did the suppliers provide a range of values for the MFR mass flow rate, so each regranulate was pre-tested to determine its mechanical and processing properties. As a preliminary study of the raw materials used during the project, measurements were carried out on the static tensile strength of the monolayer film made from the different types of regranulate and the mass melt index of the regranulates. Based on the results of the preliminary tests, presented later in this article, the regranulates were divided into three groups (A, B and C), which were related to the composition of the three-layer film formulation.

The recipes for the three-layer film were developed according to the following scheme:

- the outer layer consisted of a mixture of virgin plastic and Group A regranulate and auxiliary substances (dye, lubricant, flavourings and others), the percentage of regranulate in this layer was constant;

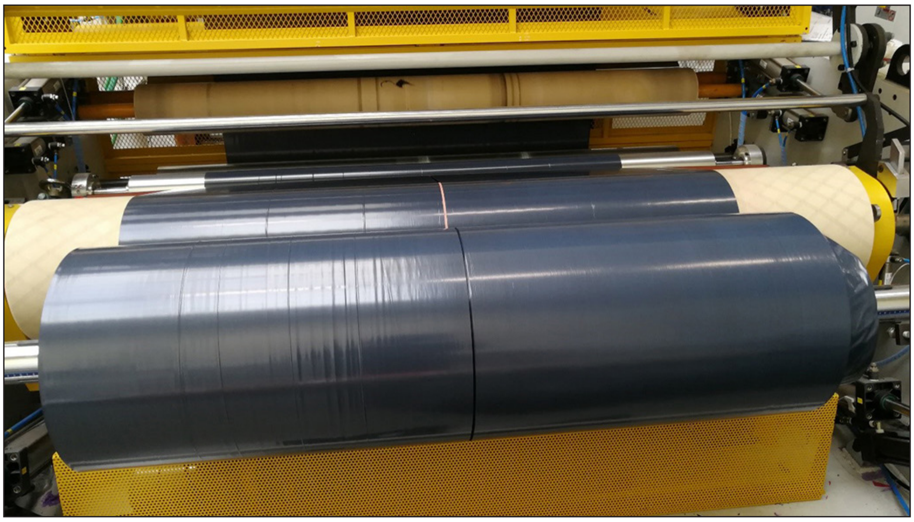

Fig. 1. Film tube made of three-layer film, separated into two separate bales

- the middle layer was available in three versions: as a mixture of virgin plastic with regranulate from group A, as a mixture of regranulates from groups A and B and as a mixture of regranulates from groups A and C, with no additives; mixtures of regranulates A and B and A and C had two types of different percentages, the total content of regranulate in this layer varied, depending on the mechanical properties of the regranulates used in this layer;
- the inner layer consisted of a mixture of virgin plastic and regranulate from group A and auxiliary substances (dye, lubricant and others); the percentage of regranulate in this layer was constant.

The proportions of the ingredients in the formulations allowed a three-layer film with a total mass percentage of regranulate ranging from up to 54% to 65.7%. The minimum total content of regranulate of 54% differed slightly from the 55% planned in the project with this being due to the need to include ancillary substances in the formulations: dyes, desiccants, auxiliaries, fragrances and other additives. The percentages of regranulate content in each layer are covered by company confidentiality. The film made from the individual recipes was subjected to measurements of the following mechanical parameters: static tensile strength, tear strength and puncture resistance. As the film tube is stretched longitudinally (MD - Machine Direction) and cross-directionally (CD - Cross Direction) during the blown film extrusion process, there was a need to test the mechanical strength of the film in these two directions. Often, plastics manufacturers provide values for selected strength parameters in the raw materials data sheets in a similar way, so the tensile strength and tear strength were determined in both the longitudinal and transverse directions.

Industry expectations to increase the competitiveness of the three-layer film product for the manufacture of rubbish bags allowed the definition of research objectives in terms of minimum values for individual mechanical parameters:

- research objective 1: tensile strength MD > 25 [MPa] for LDPE and >35 [MPa] for HDPE;
- research objective 2: tensile strength CD > 20 [MPa] for LDPE and >30 [MPa] for HDPE;
- research objective 3: puncture strength >= 100 [g] for LDPE and >= 50 [g] for HDPE;
- research objective 4: MD tear strength > 50 [N/mm] for LDPE and HDPE;
- research objective 5: CD tear strength > 50 [N/mm] for LDPE and HDPE.

METHODOLOGY OF EXPERIMENTAL TESTS AND MEASURING APPARATUS

Determining the melt flow rate

Measurements of the melt flow rate were made in accordance with PN-EN 1133-1:2011 using an Instron weight plastometer, model CEAST MF20. The general appearance of the measuring device is shown in Figure 2. The plastometer had an interface to connect it to a computer. VisualMELT C-0710-650 software was used to control the measurement of the melt flow rate in order to perform the acquisition and processing of measurement data. The operating principle of the CEAST MF20 plastometer makes it possible to measure the Melt Volume Rate (MVR), expressed in $cm^3/10$ min. The value of this indicator is read directly in VisualMELT C-0710-650 and is determined using the relationship:

Fig. 2. General appearance of the CEAST MF20 Plastometer

$$MVR_{(T,\,m_{nom})} = \frac{A_m \cdot t_{ref} \cdot L}{t} \quad (1)$$

where: MVR – volumetric melt flow rate in cm^3/10 min, T – temperature of measurement in C, m_{nom} – piston load in kg, A_m – average cross-sectional area of piston and cylinder in cm^2, t_{ref} – reference time equal to 600 s, L – cut-off step length in cm, t – cut-off step time in s.

The data recorded during measurement by the plastometer and transferred to the VisualMELT C-0710-650 software can be exported as a *.csv file. Examples of results saved in the *.csv file are shown in Figure 3. As the volumetric melt flow rate MVR is less common and the mass flow rate MFR (Mass Flow Rate) is mostly given in plastics data sheets, there was a need to calculate the latter. The cut-off step time value recorded by the plastometer (converted as an average value for the measurement carried out) was used for this purpose, as well as the total mass of the cut-off extrudate fragments, which was measured using a laboratory balance. The value of the mass flow rate index was determined from the formula:

$$MFR_{(T,\,m_{nom})} = \frac{m_{śr} \cdot t_{ref}}{t} \quad (2)$$

where: MFR – mass flow rate in g/10min, T – temperature of measurement in °C, m_{nom} – piston load in kg, $m_{śr}$ –average mass of measurement samples in g, T_{ref} – reference time equal to 600 s, t – cut-off step time in s.

The measurements were carried out using a 2.16 kg load, at a temperature of 190 °C. Approximately 3 g of granulated plastic was poured into the cylinder of the plastometer.

Determining static tensile strength

Static tensile testing enables the following strength properties to be determined: elastic limit, yield strength or yield point, maximum tensile stress (tensile strength), breaking stress, relative elongation at elastic limit and yield point, relative elongation at maximum tensile stress, relative elongation at breaking point, coefficient of elasticity. From the point of view of knowing the performance characteristics of the rubbish bag made of the tested triple layer film, out of all the static tensile test results, only the tensile strength test results are presented in this paper.

Measurements for determining the tensile strength of three-layer film were carried out in accordance with EN ISO 527-1:2020 and EN ISO 527-3:2019. The test machine Testometric, model X250-2.5, was used for the measurements. The general appearance of the testing machine is shown in Figure 4. The testing machine was connected to a PC. Using WinTest Analysis EC software, the measurement was controlled, and measurement data acquisition and processing was carried out.

The test specimens for determining the mechanical properties of the film in static tension were rectangles measuring 15 by 50 mm. The shape of the rectangle is acceptable, in accordance with PN-EN ISO 527-3:2019. The specimens were cut mechanically with a die-cutter and were prepared in order to be free of surface and material anomalies. Seven film samples were usually prepared for measurement, in order to

Column1	Column2	Column3	Column4	Column5	Column6	Column7	Column8	Column9
[Shared parameters]								
Parameter Name	Compiler	Comment	Date	Melt Type	Melt Type Unit	Standard Name	Temperature	Temperatur
Regranulat 7 04.03.2020	CEAST Lab.	Melt Flow Modular TEMPLATE parameters set	04.03.2020 19:45:52	1		ISO 1133 D	190.00	°C
[Step parameters]								
Load	Load Unit	Pressure	Pressure Unit	MFR-MVR Expected Title TYPE	MFR-MVR Expected	MFR-MVR Expected Unit	Step lenght	Step lenght
2.160	kg	0.000	Pa	MVR	1.000	cm³/10min	30.000	mm
[Shared data]								
Test name	Instrument Name	Reference	Specimen weight	Specimen weight Unit	Density	Density Unit	Test date	Operator na
Regranulat 7 04.03.2020	MF20	0	1.5960	g	1.000	g/cm³	04.03.2020 19:26:42	TN
[Step data]								
Number Of Received Measures	Number Of Valid Measures	Total Time	Total Time Unit	MFR Mean	MFR Mean Unit	MFR Std. Deviation	MFR Std. Deviation Unit	MFR Sigma
15	15	0.00	s	2.290	g/10min	0.189	g/10min	0.189
[Data Step n.: 1]								
N.	Time [s]	Measure [mm]	MFR [g/10min]	MVR [cm³/10min]	ShearRate [1/s]	Viscosity [Pa*s]		
1:	37.07	2.000	2.300	2.300	4.247	4558.276		
2:	28.79	2.004	2.968	2.968	5.480	3533.123		
3:	36.96	1.985	2.290	2.290	4.228	4578.722		
4:	37.94	2.011	2.260	2.260	4.173	4639.731		
5:	37.87	1.990	2.241	2.241	4.138	4679.177		
6:	38.39	2.015	2.238	2.238	4.132	4685.317		
7:	37.85	1.994	2.246	2.246	4.147	4668.187		
8:	38.02	1.995	2.237	2.237	4.131	4686.556		
9:	38.05	1.996	2.237	2.237	4.130	4688.151		
10:	38.38	2.019	2.243	2.243	4.141	4675.060		
11:	38.16	1.996	2.230	2.230	4.118	4701.826		

Fig. 3. Appearance of plastometer data saved as *csv file

have at least five valid samples after possibly discarding two clearly outliers (bottom and top). The samples were conditioned for at least 24 hours at room temperature of 23±2°C and a relative air humidity of 50±5%. The tensile speed of the sample was constant at 100 mm/min for the film tested

WinTest Analysis EC software enabled the desired relationships to be directly visualised on the monitor screen in graphical form (e.g. stress-strain) and the calculation of the sought-after strength properties, as well as the export of both measurement data and calculation results in the form of a data sheet and graphs.

Determining tear strength

Tear strength tests of three-layer film were undertaken in accordance with PN-EN 6383-1:2016. The measurements were taken by the Testometric testing machine, model X250-2.5, which was described before. According to the standard, the tear strength test specimens have a square shape with a side of 50 mm. A distinctive feature of the specimen is a 25 mm long notch, which facilitates two ends that are fixed in the grips of the testing machine. When this happens, the distancing grips of the testing machine cause the specimen to tear

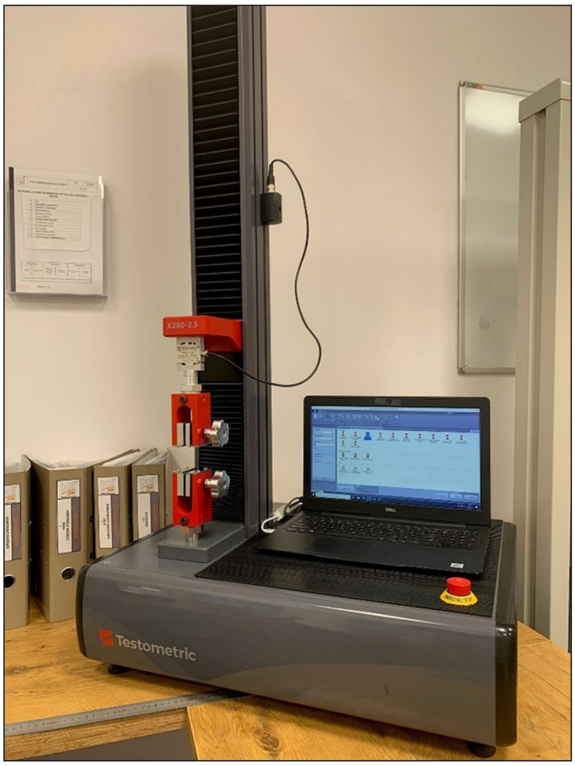

Fig. 4. General appearance of the Testometric X250-2.5 testing machine

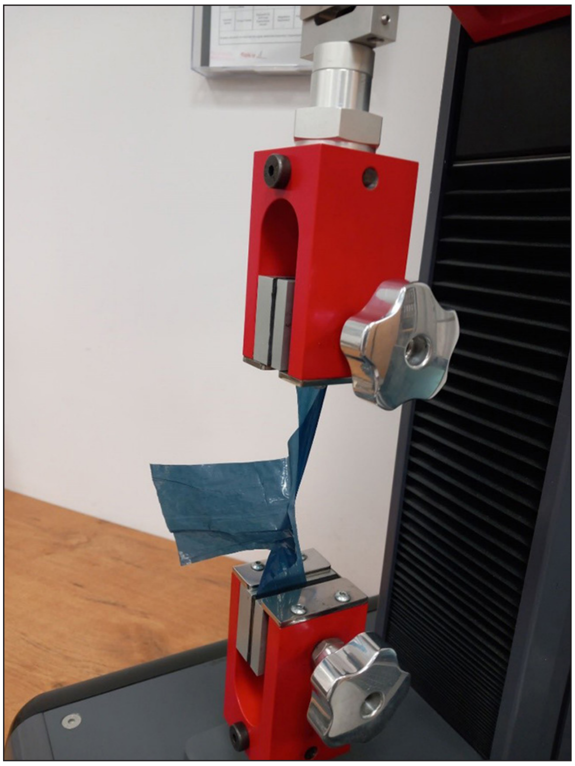

Fig. 5. Tear strength test

at the incision (Figure 5). The tensile speed of the specimen was constant at 200 mm/min.

Determining puncture resistance

Puncture resistance tests for three-layer film were undertaken in accordance with PN-EN ISO 7765-1:2005 and PN-EN ISO 7765-2:2000. The measurements were carried out using a device designed to determine the resistance to impact of film made of plastic using the free-falling arrowhead (Dart-Drop) method by Remi-Plast. The general appearance of the Dart-Drop device is shown in Figure 6.

According to the standard, the puncture resistance test specimens are square with a side of at least 150 mm. Samples can be larger, but it is essential that the sample can be correctly inserted and fixed in the holder of the measuring machine.

During the measurements, Method A was used, whereby a weight was dropped onto the test film from a height of 66 cm and the radius of rounding of the spherical part of the weight was 38 mm. The minimum mass M0 of the weight that caused the specimen to puncture was reported in grams as the test result, which was in line with the way puncture resistance is presented in the plastics data sheets provided by the plastics manufacturers.

RESULTS OF EXPERIMENTAL STUDIES

Test results of raw materials used

As mentioned earlier in the article, prior to the main testing of the three-layer film, tests were carried out on the melt mass index of all the LDPE and HDPE regranulates used in the formulations and the static tensile strength and puncture strength of the monolayer film made from these regranulates were determined.

The results of the melt mass index tests are shown in Table 1. It also shows the values of the volumetric melt flow rate MVR, which allowed the density of the individual regranulates to be determined in the molten state. Knowing the density of the regranulates was useful for verifying their type (low or high density) and for selecting some technological parameters of the blown film extrusion process.

Based on the results obtained from the melt mass index tests, the regranulates were divided into three groups, labelled A, B and C, for use in formulations for three-layer film created according to the scheme described earlier in the experimental programme. Regranulates R1, R2, R3 and R8 were classified into group A. Group B included regranulates R4, R5, R6, R7, R10, R11 and R12. Only regranulate R9 was included in Group C. The results of preliminary tests determining selected strength properties of the monolayer film made from the individual regranulates are shown in Table 2.

Fig. 6. General appearance of the device for measuring puncture resistance using the falling dart method

The preliminary test results obtained made it possible to determine a range of values for the strength properties of the individual raw materials, characteristic of a film-type product. This provided some basis for predicting what to expect from three-layer film made according to subsequent formulations, depending on which raw materials they consisted of and their percentage.

Table 1. Results of melt flow rate measurements for virgin plastics and polyethylene regranulates by volume and weight

Test material	Volumetric melt flow rate MVR (2.16 kg, 190°C) [cm^3/10 min]	Mass flow rate MFR (2.16 kg, 190°C) [g/10 min]
Regranulat 1 (R1) LDPE	1.864	1.428
Regranulat 2 (R2) LDPE	1.770	1.345
Regranulat 3 (R3) LDPE	1.981	1.527
Regranulat 4 (R4) LDPE	0.688	0.541
Regranulat 5 (R5) LDPE	0.644	0.500
Regranulat 6 (R6) LDPE	0.914	0.742
Regranulat 7 (R7) LDPE	0.666	0.524
Regranulat 8 (R8) LDPE	1.898	1.446
Regranulat 9 (R9) HDPE	0.13	0.109
Regranulat 10 (R10) HDPE	0.52	0.407
Regranulat 11 (R11) HDPE	0.710	0.531
Regranulat 12 (R12) HDPE	0.481	0.369

Table 2. Summary of static tensile strength measurement results of test film made from individual raw materials

Test material	Static tensile strength [MPa]		Puncture resistance M0 [g]
	MD	CD	
Regranulat 1 (R1) LDPE	19.79	24.01	100.0
Regranulat 2 (R2) LDPE	18.89	24.62	100.0
Regranulat 3 (R3) LDPE	22.56	24.52	145.0
Regranulat 4 (R4) LDPE	11.55	17.84	100,0
Regranulat 5 (R5) LDPE	18.91	22.69	140.0
Regranulat 6 (R6) LDPE	15.73	19.66	120.0
Regranulat 7 (R7) LDPE	20.58	26.22	145.0
Regranulat 8 (R8) LDPE	19.33	23.07	110.0
Regranulat 9 (R9) HDPE	33.08	31.94	55.0
Regranulat 10 (R10) HDPE	33.94	31.33	40.0
Regranulat 11 (R11) HDPE	48.69	42.63	40.0
Regranulat 12 (R12) HDPE	32.68	28.72	50.0

Test results for three-layer film

The results of the experimental studies carried out are presented in the form of tables and graphs. Each graph representing a separate survey is divided into four sections. Areas delineated from the crossed lines (horizontal and vertical) represent the minimum expected measurement values identified the formulations that achieved the research objectives. The way in which a given research formulation, placed on the chart is represented, includes the following vector of information:

- information on the total % regranulate content in the film (vertical axis);
- information on the values of the mechanical quantities tested (horizontal axis);
- a geometric colour/marker that indicates the leading regranulate;
- a label which shows, in sequence, the recipe number, name and % share of the additional ingredient (R - supplementary regranulate or V - original) in the middle layer of the three-layer film.

The graphical presentation of the test results adopted makes it possible to calculate the actual proportion of leading regranulate in each recipe. The manner in which it is determined is as follows:

- case I – green label F025, R7, 9.7% (Fig. 7). Establishing the amount of the leading regranulate is as follows: from the reading on the vertical axis (total percentage of regranulate), i.e. 66%, the value recorded in the label 9.7% (content of the supplementary regranulate), is subtracted resulting in a total share of the leading regranulate R2 (green) of 56.3%, with the MD strength parameter equal to 13.26 MPa (bold value in Table 3);

Table 3. Summary of the results of the static tensile strength measurements of the three-layer film, in which the leading raw material was LDPE regranulate

Formulation symbol	Static tensile strength [MPa]	
	longitudinally (MD)	transversely (CD)
F004	24.79	25.16
F005	21.78	19.73
F006	21.86	20.53
F009	15.14	19.56
F010	17.30	18.32
F011	24.01	23.03
F012	24.22	22.63
F013	20.85	22.64
F014	23.84	25.58
F007	16.07	15.36
F015	16.80	26.91
F016	23.03	25.54
F017	19,14	21.92
F024	15.52	18.36
F025	13.26	14.95
F026	13.23	16.94
F030	20.64	18.60
F031	21.87	24.22
F032	25.09	22.17
F033	18.57	21.92
F034	22.60	18.76
F035	22.73	21.88
F036	14.34	23.95
F037	17.11	21.90
F038	19.08	22.25
F039	20.01	19.99
F041	26.59	19.72
F044	19.22	20.15
F045	23.24	24.14
F050	23.53	24.71
F051	25.67	22.22

- case II – red label F038, V4, 11.6% (Fig. 7). The amount of the leading regranulate R1 (red) is read directly from the vertical axis (total percentage of regranulate) and is 54%, due to the use of original raw material (V4), with an MD strength parameter equal to 19.08 MPa (bold value in Table 3).

This form of notation allows diversification of the share of various types of raw materials in the production of three-layer film and its correlation with the mechanical parameters achieved, to be represented. The tables and graphs presented later in the article contain results related to the formulations that enabled the production of three-layer film using the blown film extrusion method in an uninterrupted technological process while maintaining production efficiency typical for the workstation. Three-layer film, produced according to formulations with numbers not listed in the above tables did not qualify for further industrial research due to failure to meet technological, economic, or usability conditions, which were not directly related to the industrial research programme adopted (e.g., in some cases, it was not possible to obtain a film tube from the film with mechanical strength allowing its stable diameter or planned thickness to be maintained).

Results of static tensile strength tests

The results of static tensile strength tests are presented in Tables 3 and 4, respectively for formulations in which the dominant raw material was either LDPE or HDPE regranulate. The graphical interpretation of the results is presented in Figures 7 to 10.

Table 4. Summary of the results of the static tensile strength measurements of the three-layer film, in which the leading raw material was HDPE regranulate

Formulation symbol	Static tensile strength [MPa].	
	longitudinally (MD)	transversely (CD)
F046	26.01	25.78
F047	30.12	26.89
F048	33.28	27.22
F052	33.98	25.02
F054	42.52	25.67
F055	32.83	23.81
F056	20.10	21.52
F057	19.98	23.01
F058	28.68	22.29
F059	28.68	28.68

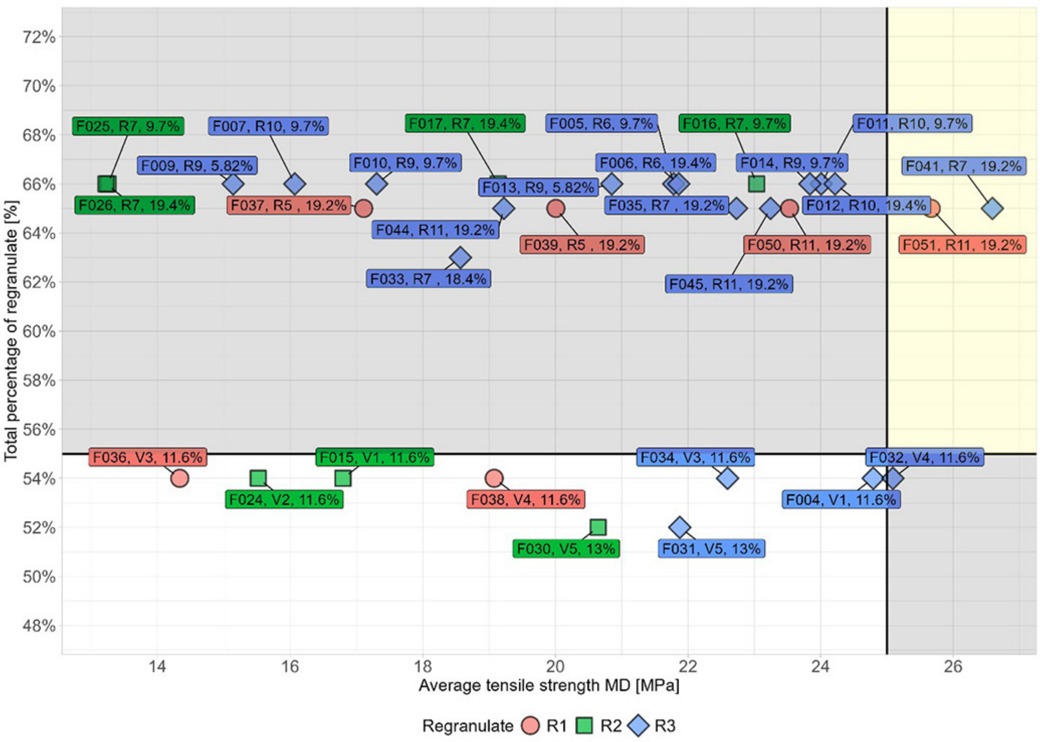

Fig. 7. Values of static tensile strength in the longitudinal direction of three-layer film, in which the dominant raw materials were LDPE regranulates

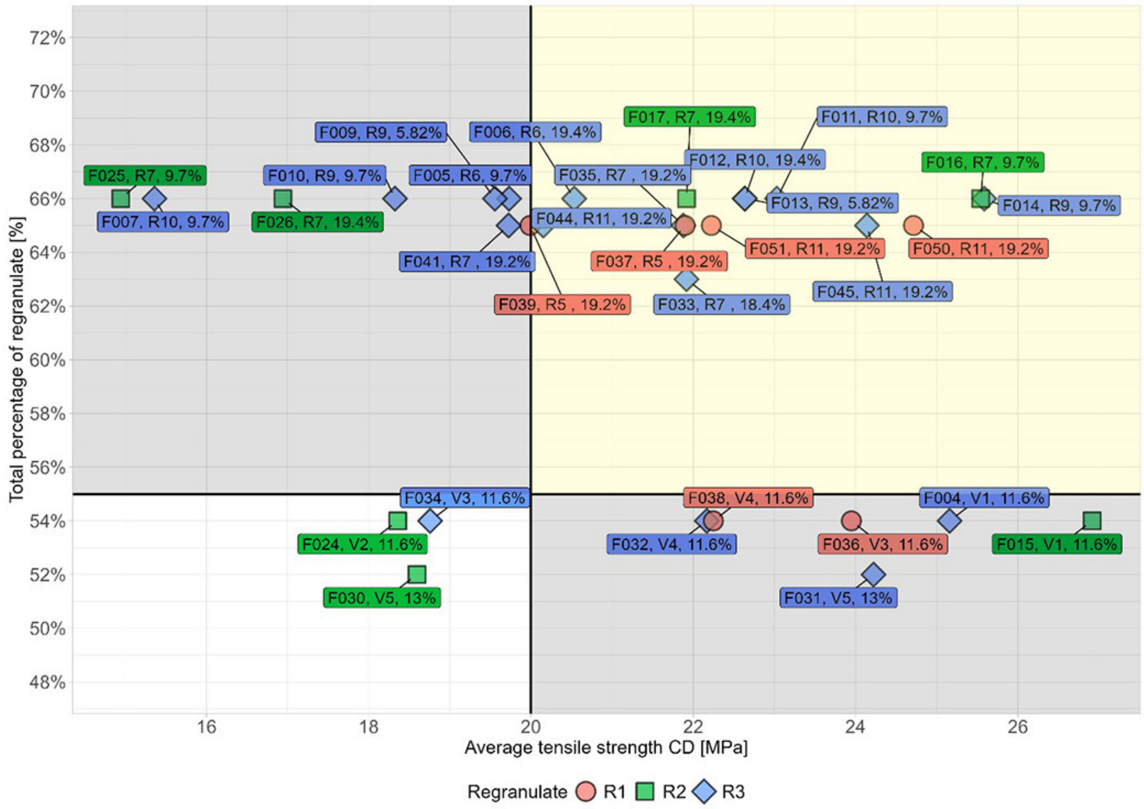

Fig. 8. Values of static tensile strength in the transverse direction of three-layer film, in which the dominant raw materials were LDPE regranulates

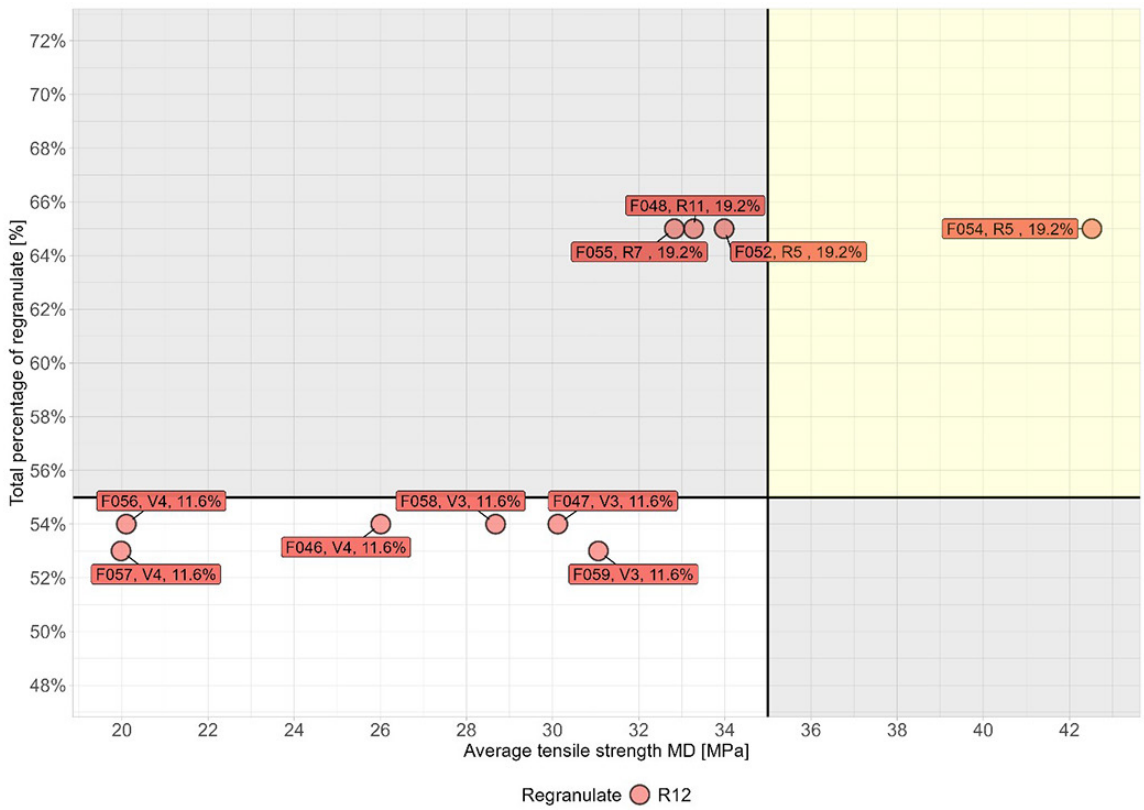

Fig. 9. Values of static tensile strength in the longitudinal direction of three-layer film, in which the dominant raw materials were HDPE regranulates

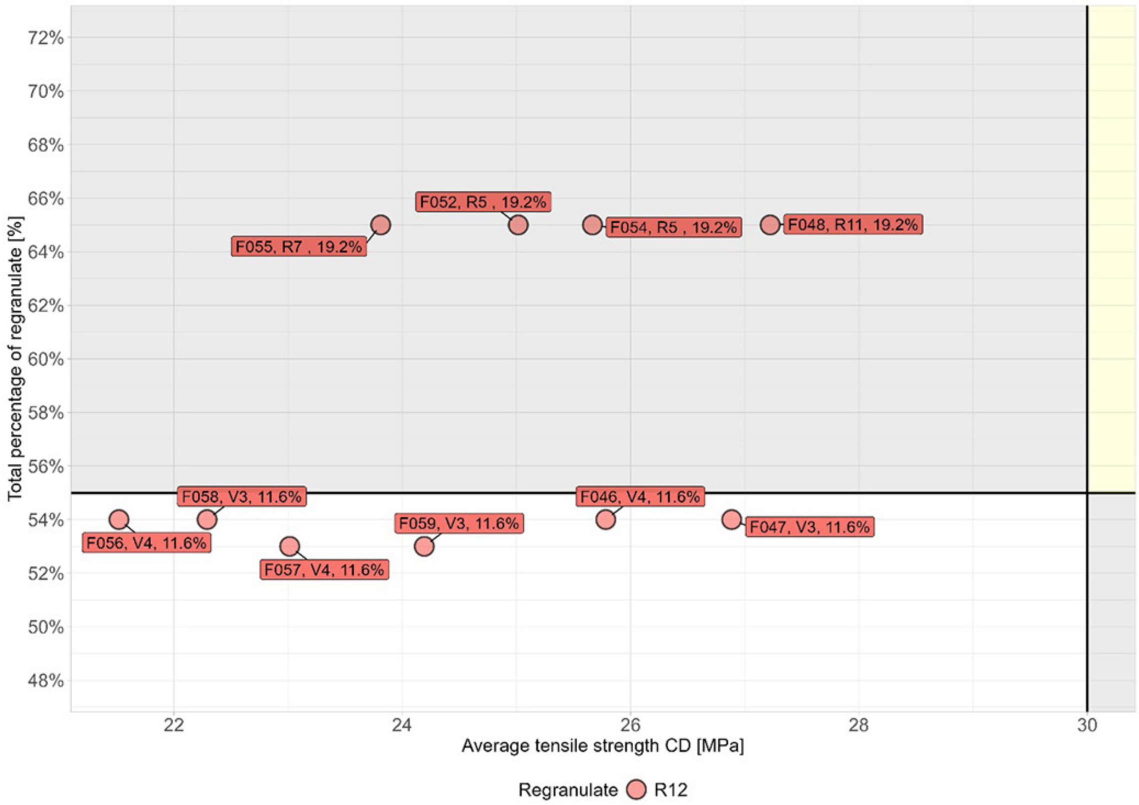

Fig. 10. Values of static tensile strength in the transverse direction of three-layer film, in which the dominant raw materials were HDPE regranulates

Results of tear strength tests

The results of the tear strength tests are presented in Tables 5 and 6, respectively, for formulations in which the dominant raw material was either LDPE or HDPE regranulate. The graphical interpretation of the results is presented in Figures 11 to 14.

Results of puncture strength tests

The results of the puncture resistance tests are presented in Tables 7 and 8, respectively for formulations in which the dominant raw material was either LDPE or HDPE regranulate. The interpretation of the results in graphic form is presented in Figures 15 and 16.

Tests results analysis

The three-layer film produced with significant percentages of various LDPE (R1, R2, R3) and HDPE (R12) leading regranulates and various complementary materials (other regranulates, original LLDPE and complementary additives) was subjected to longitudinal MD and transverse CD strength tests. The MD strength values obtained for the LDPE film group show that the target value (defined at 25 MPa - research objective 1) was achieved by three formulations, of which, in two formulations, i.e. F041 and F032, the leading regranulate R3 was at the level of 46% and 54%, respectively and in one formulation (F051) the leading regranulate R1 was at the level of 46%. Thus, the total regranulate content in the film was at the level of 65% for formulations F041 and F051, and 54% for formulation F032. In turn, the film produced from formulations containing the leading regranulate R2 did not achieve the expected value of longitudinal strength MD. Figure 7, shaded in yellow, shows two formulations (F041 and F 051) that meet the test objectives adopted, i.e. an MD strength of not less than 25MPa and a total regranulate content of not less than 55%. In the CD transverse strength test, significantly more formulations (with the leading LDPE regranulates R1, R2 and R3) achieved the target strength (not less than 20 MPa) and a total % regranulate content of more than 55%, as can be seen in Fig. 8 (field shaded in yellow). In the group of film,

Table 5. Summary of the results of the tear resistance measurements of the three-layer film, in which the leading raw material was LDPE regranulate

Formulation symbol	Tear strength [N/mm]	
	longitudinally (MD)	transversely (CD)
F004	113.93	195.83
F005	131.87	167.92
F006	122.77	167.05
F009	97.30	167.18
F010	128.46	199.63
F011	145.69	190.05
F012	144.08	192.40
F013	107.20	204.55
F014	83.16	179.87
F007	139.39	181.35
F015	109.26	176.55
F016	135.70	175.13
F017	106.69	150.50
F024	145.66	179.73
F025	144.98	179.57
F026	144.55	182.89
F030	110.86	162.21
F031	133.20	188.91
F032	126.21	194.47
F033	120.14	186.10
F034	119.26	162.31
F035	123.19	178.93
F036	120.05	168.15
F037	104.32	172.65
F038	139.09	188.13
F039	115.88	170.26
F041	107.58	229.83
F044	182.99	201.08
F045	160.77	205.62
F050	142.66	243.43
F051	93.02	268.27

Table 6. Summary of the results of the tear resistance measurements of the three-layer film, in which the leading raw material was HDPE regranulate

Formulation symbol	Tear strength [N/mm]	
	longitudinally (MD)	transversely (CD)
F046	137.86	193.02
F047	153.32	297.57
F048	183.54	278.21
F052	145.97	261.91
F054	196.52	317.80
F055	111.96	310.65
F056	108.46	214.41
F057	91.78	204.69
F058	137.22	278.77
F059	138.05	273.60

Table 7. Summary of the results of the puncture resistance measurements of the film, in which the leading raw material was LDPE regranulate

Formulation symbol	The lowest puncture mass of the film M0 [g]
F004	185.0
F005	155.0
F006	165.0
F009	125.0
F010	75.0
F011	150.0
F012	145.0
F013	150.0
F014	145.0
F007	110.0
F015	150.0
F016	135.0
F017	150.0
F024	80.0
F025	95.0
F026	110.0
F030	175.0
F031	170.0
F032	175.0
F033	150.0
F034	400.0
F035	185,0
F036	295.0
F037	190.0
F038	165.0
F039	160.0
F041	145.0
F044	115.0
F045	160.0
F050	75.0
F051	30.0

Table 8. Summary of the results of the puncture resistance measurements of the film, in which the leading raw material was HDPE regranulate

Formulation symbol	The lowest puncture mass of the film M0 [g]
F046	165.0
F047	65.0
F048	45.0
F052	50.0
F054	30.0
F055	30.0
F056	180.0
F057	120.0
F058	80.0
F059	70.0

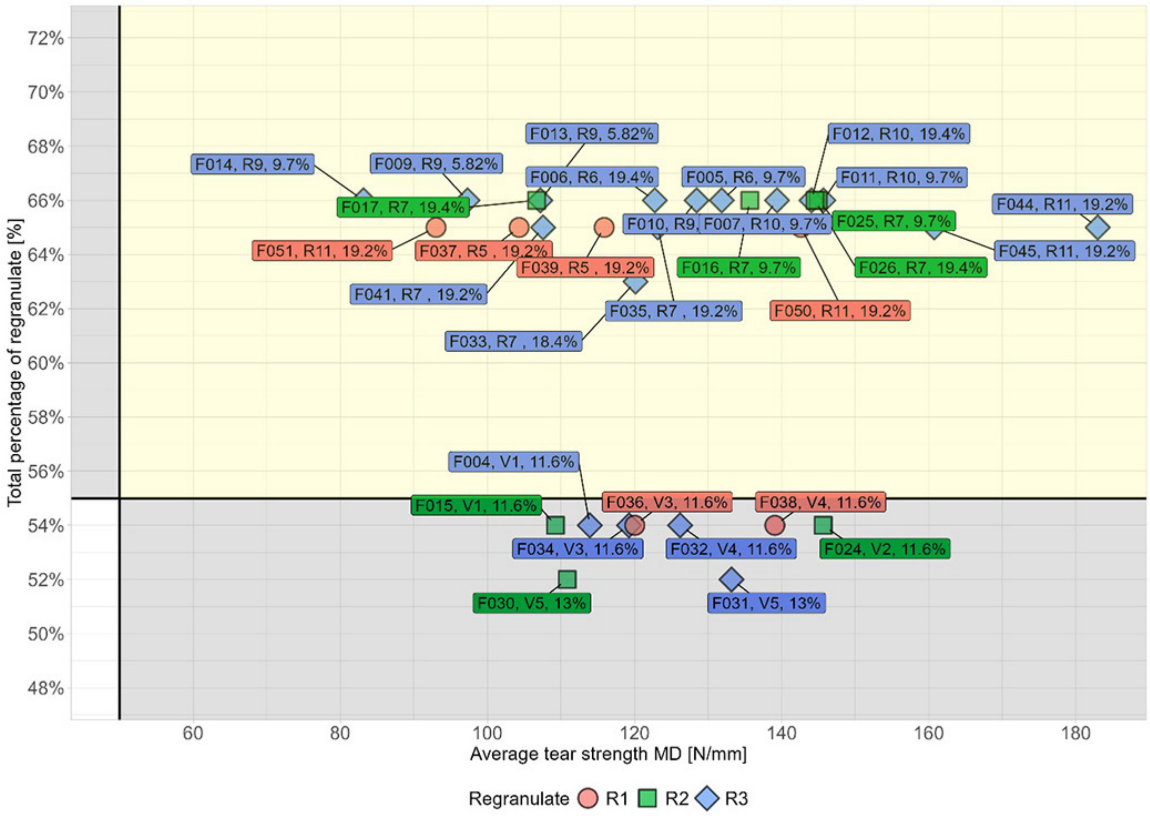

Fig. 11. Values of tear resistance in the longitudinal direction of three-layer film, in which the dominant raw materials were LDPE regranulates

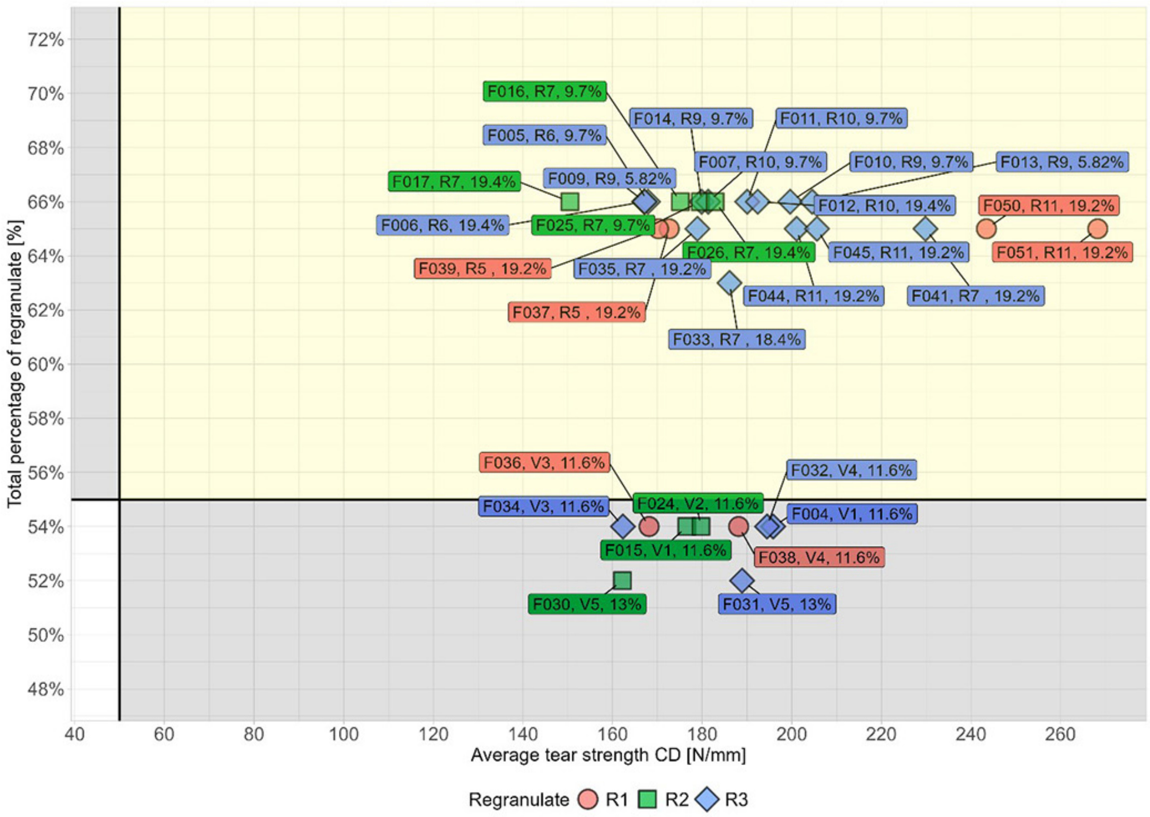

Fig. 12. Values of tear resistance in the transverse direction of three-layer film, in which the dominant raw materials were LDPE regranulates

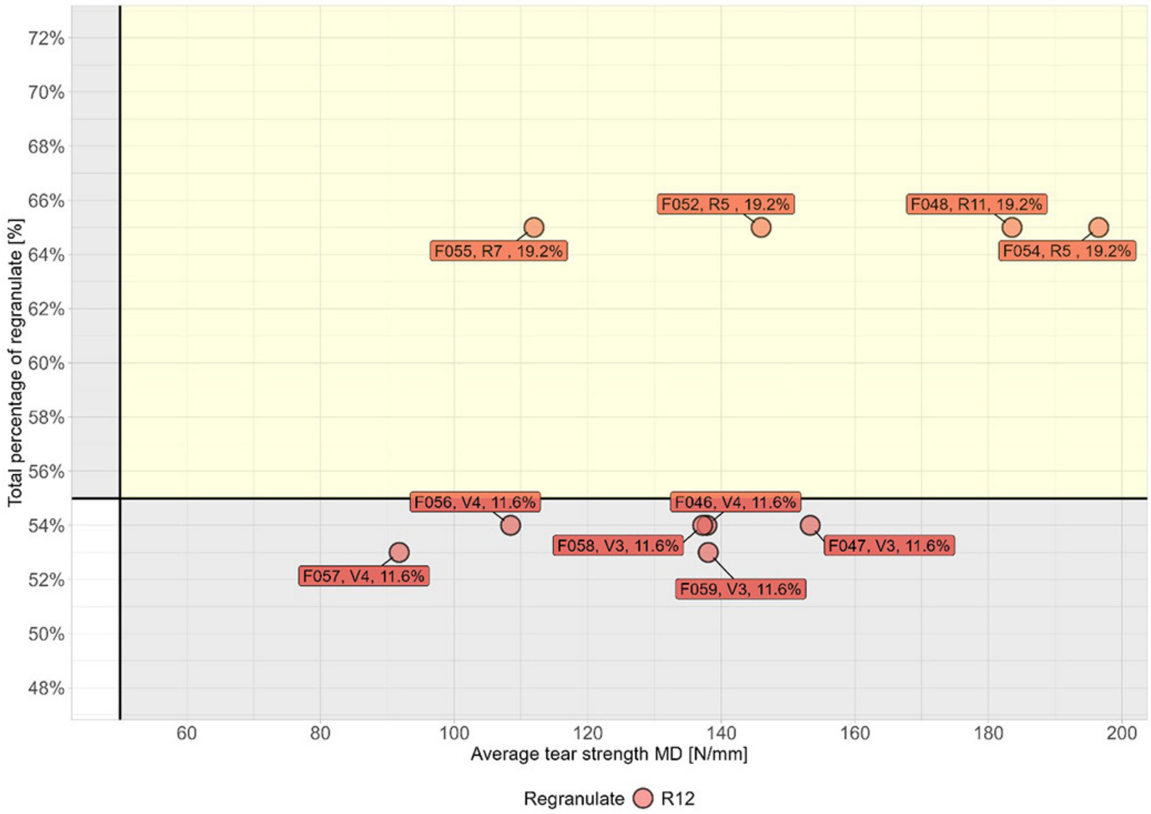

Fig. 13. Values of tear resistance in the longitudinal direction of three-layer film, in which the dominant raw materials were HDPE regranulates

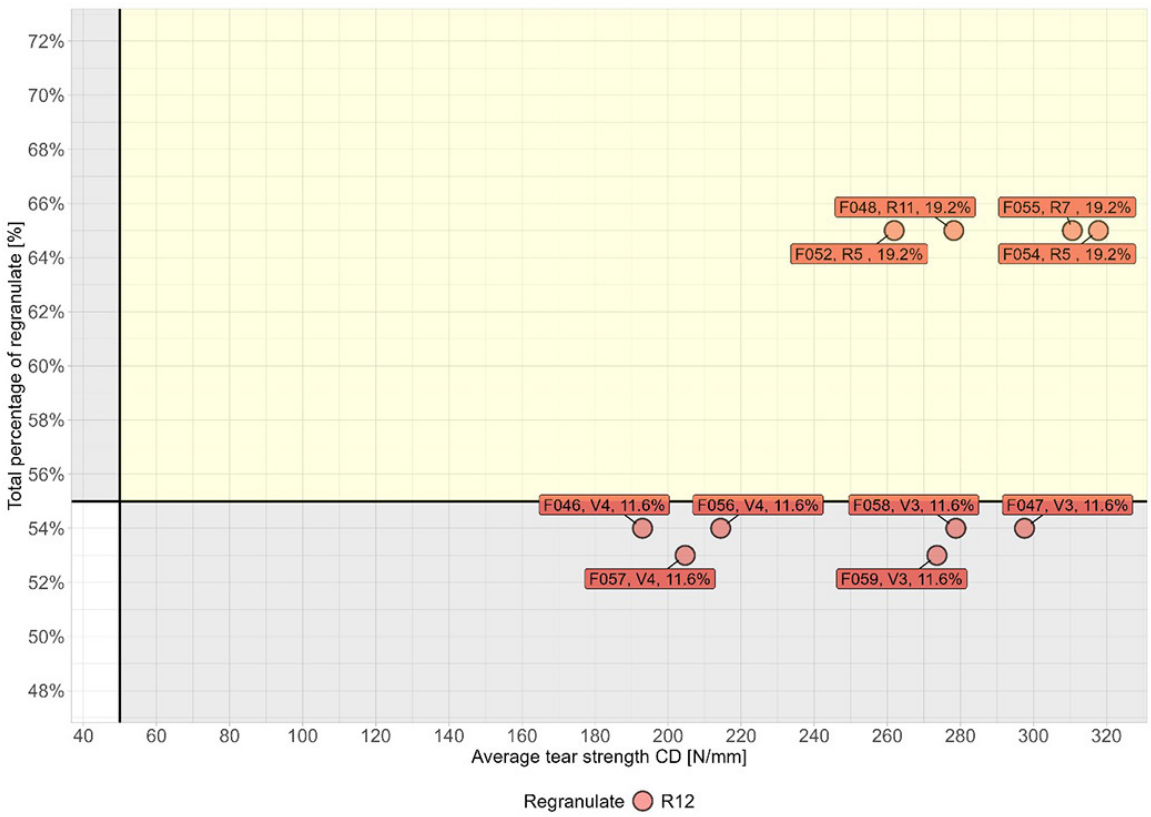

Fig. 14. Values of tear resistance in the transverse direction of three-layer film, in which the dominant raw materials were HDPE regranulates

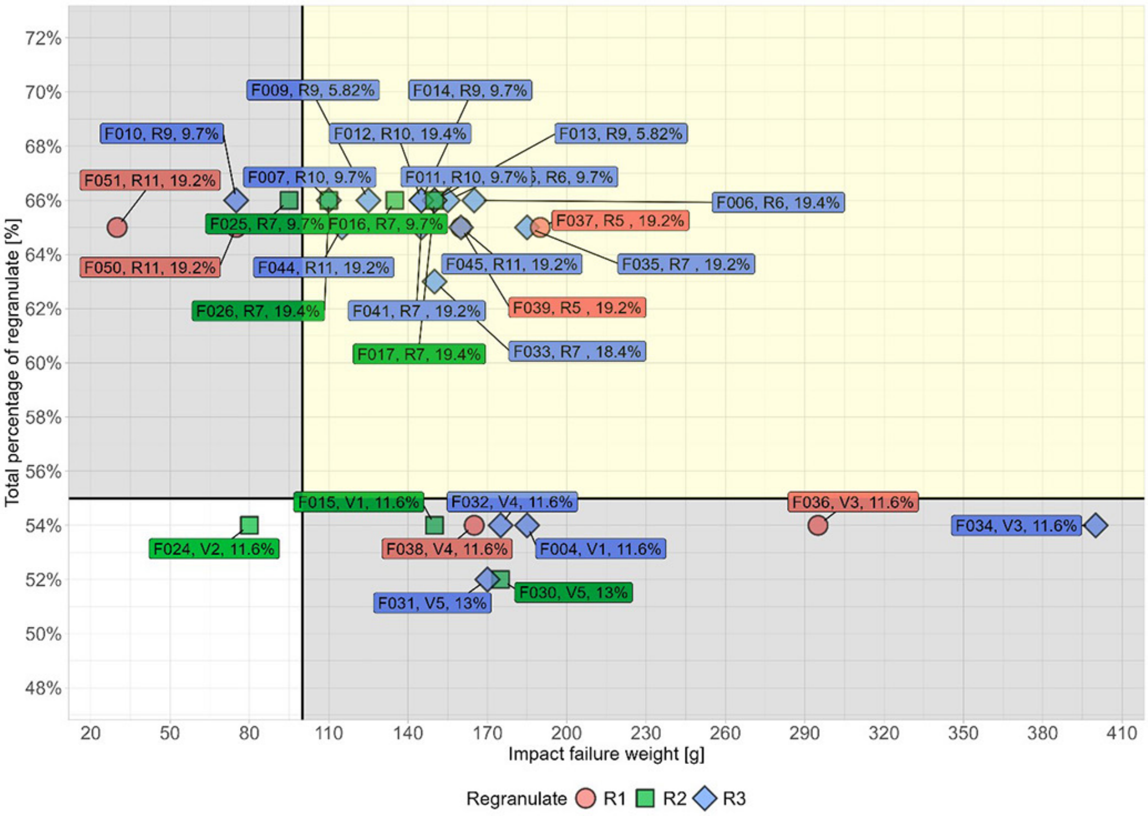

Fig. 15. Values of puncture resistance in three-layer film, in which the dominant raw materials were LDPE regranulates

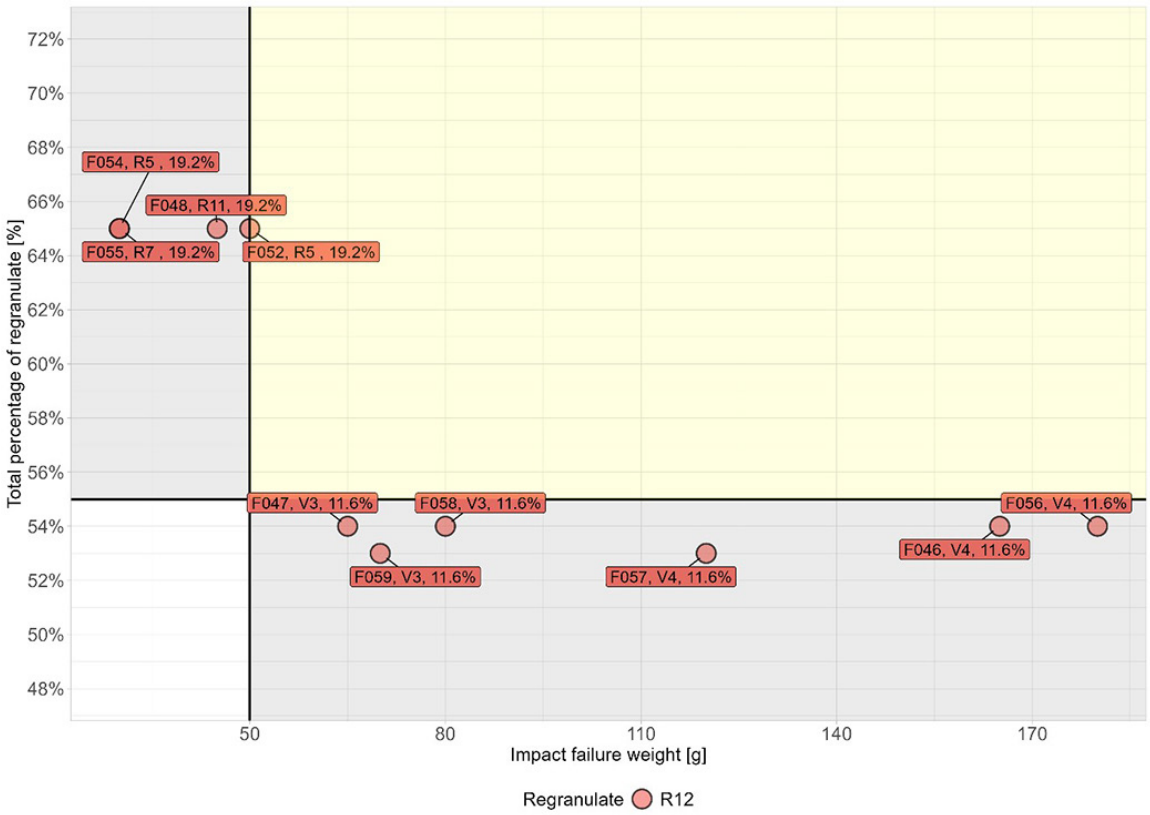

Fig. 16. Values of puncture resistance in three-layer film, in which the dominant raw materials were HDPE regranulates

Table 9. Summary of the number of formulations meeting individual research objectives

Number of formulations	Type of regranulate	Regranulate content >55%	Research objective 1	Research objective 2	Research objective 3	Research objective 4	Research objective 5
31	LDPE	22	3	21	26	31	31
10	HDPE	5	2	0	7	10	10

produced with HDPE leading regranulate (R12), it can be seen that formulations F048, F052, F054 and F055 achieved a total % regranulate share of 65%. In the MD strength tests (Figure 9), only the film produced from recipe F054 achieved a value above the target, at no less than 35 MPa, while CD strength (at no less than 30 MPa) was not achieved for any of the recipes tested (Figure 10).

In the tensile strength test, in the group of formulations involving LDPE regranulate, the target minimum value in the MD direction of not less than 50 [N/mm] was achieved by all the film tested made from the three leading regranulates R1, R2 and R3 (Figure 11). The same was true for the CD transverse survey - Figure 12. In both cases, it was observed that 9 formulations did not have a total regranulate percentage above the 55% level. These are graphically represented in a field shaded in grey. In the tear test, in the group of formulations involving HDPE regranulate, also all the film tested achieved a strength of not less than 50 N/mm in the longitudinal MD and transverse CD directions (Figures 13 and 14). However, only four of them (F048, F052, F054 and F055) have a total regranulate % share higher than 55%. The last test carried out was the measurement of the film's puncture strength. The final test carried out was a measurement of the film's puncture resistance. The minimum film damage weight values of not less than 100 g for formulations with LDPE leading regranulate (Figure 15) and 50 g for formulations with HDPE leading regranulate (Figure 16), assumed for the research objectives, were achieved for most of the film tested. It can be noted, however, that formulations F007, F010, F025 and F026 (LDPE) and F048 and F055 (HDPE), which did not meet these conditions, also failed to achieve their targets for the static tensile strength test in the longitudinal direction (Fig. 7 and 9) and transversely (Fig. 8 and 10). Table 9 summarises the number of formulations tested (LDPE and HDPE) and, in further columns, the number of formulations meeting each research objective.

CONCLUSIONS

The research developed and carried out has led to the following conclusions:

The instability of the processing properties of commercially available regranulates made it necessary to classify them in order to develop recipes for the weight dosage of waste plastics and to combine them with the LLDPE virgin material in the corresponding layers of the three-layer film produced;

Due to the mechanical properties of the waste material, different proportions were used in the individual layers, taking into account the principle of using at least 50% regranulate in the inner and outer layers and experimentally increasing its content in the middle layer;

The construction of a three-stage classifier of available regranulates based on MFR values was sufficient and enabled the development of a database of 41 formulations, from which more than 55% of the unstable waste material was introduced into 27 formulations;

Increasing the degree of use of waste plastics, by increasing their share of the unit product to 55% (a company-wide innovation), allowed the mechanical parameters of the film to be maintained, meeting defined industry expectations in terms of static tensile and tear strength and puncture resistance.

Regranulate share should be increased in finished products in line with the zero-waste principle in a circular economy. As demonstrated by industrial research, it is possible to produce three-layer film with the desired performance characteristics using a significant number of different LDPE and HDPE regranulates, which differ in their mechanical and processing properties.

Acknowledgements

This publication was created as part of the implementation of the project under Measure 1.1 R&D Projects of enterprises; Sub-measure 1.1.1 industrial research and development works carried out by enterprises of the Smart Growth Operational Programme. Contract number POIR.01.01.01-00-0379/19. Project title: Development and implementation of high-effective automatised and robotised technology for making knurled ecological rubbish bags from three-layered film produced by blow film extrusion, financed by the National Centre for Research and Development.

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