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*Original scientific paper*

# Changes in Mechanical and Rheological Properties of Recycled Thermoplastics

Zoran Janjuš[1]
City Administration Banja Luka, Municipal Police Department, Banja Luka
Predrag Miletić
University in Banja Luka, Faculty of Forestry, Banja Luka
Predrag Ilić
Institution Protection, ecology and computer science, Banja Luka

**Abstract:** Polymer materials have extremely wide application in various fields of technology and human needs (jobs, food industry, production of various packaging, furniture and household goods, Electrical and electronics, agriculture, chemical, automotive, aerospace and military industry and so on. This paper presents the results of changes in mechanical and rheological properties of recycled thermoplastics. The purpose is to show the polymer waste as raw material, or otherwise utilizing the material. Specific information will be given the possibility of forming a form to calculate the size observed through cycles of recycling, which may help in further research and analysis.

**Key words:** recycling, termoplast, properties.

## Introduction

Last century was marked by extremely rapid development of industry. As technology improved, the human needs for various types of products increased too. This led to a large and uncontrolled exploitation of raw materials. Today, in a consumer society, it is characteristic that the product is changed with a new, modern, functional and aesthetically likeble, and before the expiration of his exploitation century. Among other things, a characteristic example is mobile phones that users change due to fewer new modern look even if cellulars are still correct.

In order to get new product satisfactory technical properties, pleasing appearance, a short lifetime and low market prices, it is necessary to optimize all stages of its creation to the development and sale of (Janjuš; Janjuš and Šljivić, 2006).

One of the measures of optimization is using of cheaper technical materials. Plastics as an engineering material appeared in the first half of last century. Their use has continued an upward trend. Until today, any other material is not recorded by rapid development thanks to a very strong relative performance-quality-price and the fact that the characteristics of plastic can be easily adapted to different requirements of exploitation, as well as the relatively simple processing procedures. When you create a product should not ignore the fact that its exploatical deadline limited and will sooner or later found in a landfill for waste disposal. Important place in landfills have products made of plastic. A characteristic of modern society is a large amount of waste Certainly, the trend of production increase and its use follows the trend of waste increase. Large quantities of waste are disposed outside the places provided for disposal of waste.

A special problem for the ecological environment are materials that have extremely long period of decay and bioundegraded materials, so it represents as pollutants of nature for a long time. In recent years, through propaganda and appeals of environmental organizations, growing awareness of citizens and society as a whole to preserve the natural environment. In addition to this increase in prices of raw materials and energy requires a

[1] Correspoding author: E-mail: p.miletic@blic.net

new approach to creating a product, material selection, lifetime and especially to prescribe measures to be taken when product run out. Further problem in using polymeric materials is the fact that there are many different types of polymers and that each of them requires a different approach to further exploitation.

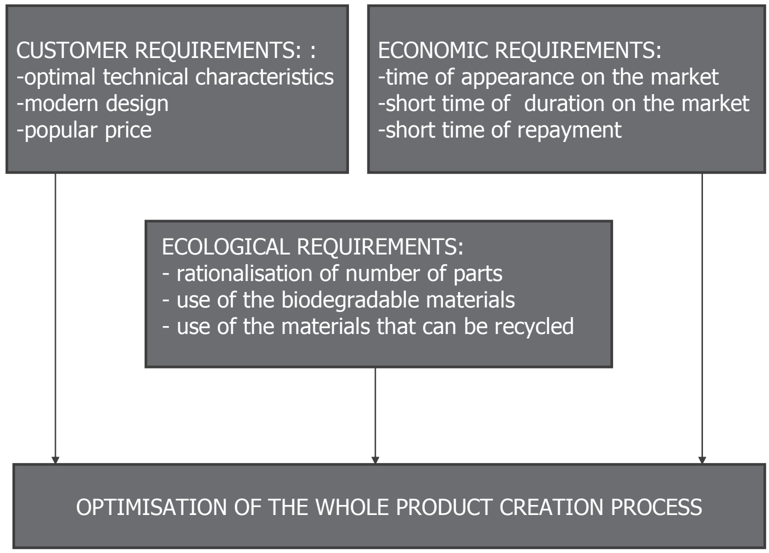



Figure1. Important requirements that should be satisfied for the product creation process

## Material and Methods

### *Description of experiment*

The experiment studied the impact of multiple recycling of thermoplastics mechanical properties change through the cycle. The aim is to bring the conclusions of the features, quality and reliability of the use of recycled materials. The changes in mechanical properties are accompanied by the three materials simultaneously through five cycles of recycling.

Three main materials were used for experiment:

1. Polyethylene (polyethylene PE-LD, Chemopetrol,a,s, Litvinov Czech Republic),
2. Polystyrene (polystyrene PS, DiOKI d.d., Zagreb Croatia),
3. Polypropylene (polypropylene PP, Hipol, Chemical industry, Odžaci SiCG).

All materials are refined on the same machine:

1. Title: ARBURG-MASCHINENFABRIK, HEHL & SOHNE, D 7298 LOSSBURG 1
2. Type: allrounder 305-210-700
3. Series: 115068
4. Motor force: 280 Nm
5. production year:1980

Processing is done in private enterprise Dita Banja Luka, March 2005. year. After each cycle of processing a certain amount of material is kept, and the rest comminuted and prepared for the next stage of processing So we get a tube of basic materials and test tubes I, II, III, IV and V of the cycle of recycling. Test tubes were obtained by extrusion process.

Processing temperature was constant and amounted to the190°C, 210°C, 220°C for the first, second and third heater.

Test tube cooling was done slowly at room temperature, without additional funds.

Crushing of materials (preparation for re-processing) was done in industrial mill.

The material is processed in the test tube in which tightening and hardness was measured, and from them were cop prepared examples for measuring the toughness and shear stress.

Test tubes were cast in a specially designed mold..

Experiment included the following measurements:

1. Maximum tensile force in,
2. Elongation at tensile,
3. Maximum force in shear i
4. Hardness by Rockwel.

The calculation of other mechanical quantities was performed which can be obtained using the measured values.

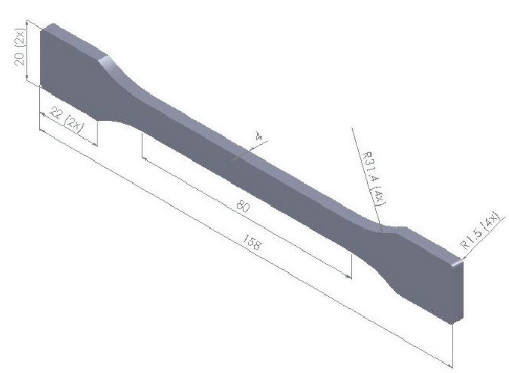
### *Testing tightening*

Testing tightening was done in the laboratory for quality control of "Jelšingrad - steel foundries "A. D. Banja Luka, April 2005.

Measuring machine data:

1. Name of the criteria: A device for measuring the strength - Testing Machine,
2. The manufacturer measures: Alfred Amsler, Germany,
3. Serial Number: 699/473,
4. Basic metrology characteristics: type SZBDA-699, measuring range 0-350kN, class of accuracy + / - 1%

Measure fulfilled machines condition for measuring the mechanical properties of the standard EN ISO 10 002 - 2




Picture 1. standard test tube

A device for measuring tensile strength - Testing Machine is shown in photo.

Test tube test (shown in the following figure) is with standard shape and dimensions.

The test is carried out by german standard DIN 50114 - for striped tube (Picture 1).

Dimensions of test tubes for testing:

1. The total length of the tube l1=156mm
2. First part of the length measurement l0=80mm,
3. Width of the clamping portion B=20mm,
4. Home for measuring cross-section width b0=10mm,
5. Home thickness cut-off for measuring s0=4mm.

For one cycle test was conducted four test tubes of the same series, and as a measurable result the mean measured size is adopted.

Test results are:

1. Max power F[N],
2. Tensile strength Rm[N/mm2],
3. Elongation Dl [mm],
4. Unit strain e 8 $\varepsilon = \frac{\Delta l}{b}$

Way of obtaining data:

1. Maximum force - with a direct reading instrument measure,
2. Tensile strength - calculating the pattern: $Rm = \frac{F\,max}{Ao}$ , where: $Ao = bo \cdot so$ – initial transverse cross-section area,
3. Elongation - measuring floating measure,
4. Unit strain - calculating the form: $\varepsilon = \frac{\Delta l}{lo}$ ,

Description of transferring measure:

1. Title: Transferring or bottlenose measure,
2. Purpose: Measurement of external and internal linear measures ,
3. Features: - reading - analogue;
   - measuring range: 0 – 150 mm;
   - accuracy measures: 0,05 mm.

## *Shear Testing*

Examination of tightening was done in the laboratory for quality control of "Jelšingrad - TAS"A. D. Banja Luka, April 2005. year.

1. Data on measuring machine:
2. Name of the criteria: A device for testing pressure and stretching,
3. Manufacturer criteria: MIP, USSR,
4. Serial Number: N 122, year 1978,
5. basic metrology characteristics: type 100-2 T 4.1. , measuring range 0-100kN, Accuracy class

+/- 1%

Dimensions of working area of the sample for testing:

1. Length l = 22mm,
2. Width B = 20mm,
3. Thickness s = 4mm,
4. Diameter hole d = 5,20mm.

For one cycle test was carried out four samples, and as a result it is adopt the mean maximum force. The holes are punched in samples d = 5.20 mm, and the value of breaking force was read by measuring instrument.

Test results are:

1. Max shear force F[N] 8
2. Shear strength τm [N/mm2],

Way of obtaining data:

1. Maximum shear force - with a direct reading measuring instrument ,
2. Shear strength - calculating the pattern: $\tau max = \frac{F max}{L \cdot s}$ , where: $L = d\pi$ - volume hole that penetrates and $d = 5,20mm$ diameter hole.

The results are shown in tables and graphs (Skakić, 2001).

## *Examination of hardness by Rockwel*

Data on the scale:

1. Name of criteria: WOLPERT-PROBAT,
2. Type of criteria: Harteprufer HT 2001, RB
3. serial number: 89 00386/0001,1990. god.,
4. Standard: DIN 51 224

Measurement procedure is carried out by the cone element (top of the cone angle is 1200) impressed with the diamond head tube to the surface, and the scale reads value. Measuring points are chosen on the wider part of the tube.

Hardness testing was performed in the laboratory for quality control of "Jelšingrad - TAS" A. D. Banja Luka, April 2005.

## *Rheological Properties of Thermoplastics*

Rheological properties of thermoplastics will be tables and graphs presented and analyzed results of measurements:

1. Strength index DIN 53735 – JUS G.S2.601
2. Relative density DIN 53420 – JUS G.S2.510

Characteristics of the experimental section:

1. Measurements were made on equipment „Zwick",
2. Processing is carried out injection molding,
3. Machine: „Belmatik 50/28R",

4. Processing system used to search „IBM-PC-AT-Mikronic“,
5. Materials: polyethylene -PE, polystyrene -PS and polypropylene -PP,
6. Temperature processing: 190ºC- PE i 210ºC-PS,PP.
7. Pressure processing: 80 bar-PS i 100 bar-PE,PP. [24]

### *Processing and analysis of results*

The analysis is done in Microsoft Excel using the regression method. All curves presented approximate polynomial functions are the fourth or fifth degree. Graphical and tabular presentation will illustrate the analysis of characteristics of polyethylene, while for other materials function changes are shown tables

## Results and Discussion

### *Mechanical properties*

**Tensile strength [$R_m$]** lightweight polyethylene recorded an upward trend compared to the base material. Maximum value is at the third cycle of recycling, followed by slow decline until the fifth cycle. Maximum change in tensile strength does not exceed 6.5%.

**Tensile strength [Rm]** lightweight polystyrene recorded an upward trend compared to the basic material. Maximum value is at the first cycle of recycling, followed by nearly constant value up to the fifth cycle. Maximum change in tensile strength does not exceed 25%.

**Shear strength [τm]** polyethylene recorded slow downward trend of recycled material in relation to the basic material. The overall decrease in shear strength to the base material does not exceed 12.2%.

**Shear strength [τm]** recorded a lightweight polystyrene downward trend of recycled material in relation to the basic material. The overall decrease in shear strength to the base material does not exceed 16%.

**Shear strength [τm]** Polypropylene recorded slow downward trend of recycled material in relation to the basic material. The overall decrease in shear strength to the base material does not exceed 8.1%.

**Hardness** polyethylene recorded an upward trend in recycled materials to the base. Were also a much wider range of imbalances measured hardness measured at different locations tube of recycled materials.

**The hardness** of polystyrene recorded consistent results, no significant deviation (less than 5%) of recycled material in relation to the base.

Polypropylene recorded a uniform hardness result, no significant deviation (less than 5%) of recycled material in relation to the base.

The highest tensile strength has polypropylene. Polypropylene and polyethylene have a mild almost insignificant changes in tensile strength. Lowest tensile strength and harshest change has polystyrene.

The highest shear strength has polystyrene. The lowest shear strength has polyethylene. All three recorded material shear strength lightweight pad.

All three materials have a slight difference in hardness observed cycles of recycling and a wide range of imbalances measured hardness (Šljivić and Stojanović, 2001).

Mechanical properties of recycled materials (thermoplastics) can vary from basic material properties.

In polyethylene and polypropylene tensile strength had a slight change. Shear strength in all three materials have a constant slight decrease. Hardness is slightly exchanges with all the material, and has a relatively wide range of steps observed for the different measuring points.

Table 1 Experiment results for some materials (Janjuš, 2006)

| **Polyethylene** | | | | | | |
|---|---|---|---|---|---|---|
| CYCLE RECYCLING | **0** | **I** | **II** | **III** | **IV** | **V** |
| Max force ist F[N] | 917,235 | 941,76 | 951,57 | 976,095 | 958,93 | 946,665 |
| Tensile strength $R_m$[N/mm$^2$] | 22,93 | 23,544 | 23,789 | 24,402 | 23,97 | 23,667 |
| Elongation Δl[mm] | 2,76 | 2,41 | 3,69 | 2,3 | 3,33 | 2,03 |
| UNIT deformation ε | 0,035 | 0,031 | 0,046 | 0,029 | 0,042 | 0,025 |
| Max force cм F[N] | 2029,88 | 1922,76 | 1867,83 | 1867,83 | 1804,65 | 1782,67 |
| STRENGTH shearing τm[N/mm$^2$] | 31,08 | 29,44 | 28,6 | 28,6 | 27,63 | 27,29 |
| Hardness HRC(Rockwel) | 75-80 | 80 | 77 | 78-80 | 75-81 | 76-79 |
| **Polystyrene** | | | | | | |
| Max force ist F[N] | 694,06 | 917,235 | 836,253 | 801,968 | 836,302 | 819,135 |
| Tensile strength $R_m$[N/mm$^2$] | 17,352 | 22,931 | 20,91 | 20,049 | 20,908 | 20,478 |
| Elongation Δl[mm] | 5,98 | 18,7 | 13,43 | 10,55 | 11,13 | 9,33 |
| Unit deformation ε | 0,075 | 0,234 | 0,168 | 0,132 | 0,139 | 0,117 |
| Max force cм F[N] | 2408,94 | 2323,8 | 2285,34 | 2252,38 | 2224,91 | 2065,6 |
| Strength shearing τm[N/mm$^2$] | 36,88 | 35,58 | 34,99 | 34,49 | 34,07 | 31,63 |
| Hardness HRC(Rockwel) | 90-93 | 90-92 | 87-92 | 85-90 | 85-91 | 85-90 |
| **Polypropylene** | | | | | | |
| Max force ist F[N] | 1172,295 | 1177,20 | 1184,56 | 1187,01 | 1211,535 | 1226,25 |
| Tensile strength $R_m$[N/mm$^2$] | 29,307 | 29,43 | 29,614 | 29,675 | 30,288 | 30,656 |
| elongation Δl[mm] | 6,36 | 6,83 | 5,03 | 4,13 | 4,26 | 5,23 |
| Unit deformation ε | 0,08 | 0,085 | 0,063 | 0,052 | 0,053 | 0,065 |
| Max force cм F[N] | 2351,26 | 2312,81 | 2321,05 | 2282,59 | 2263,36 | 2161,73 |
| Strength shearing τm[N/mm$^2$] | 36,0 | 35,41 | 35,54 | 34,95 | 34,65 | 33,1 |
| hardness HRC(Rockwel) | 83-84 | 85-86 | 82-86 | 80-82 | 80-83 | 82-87 |

Observed mechanical sizes through five cycles of recycling have not radically changed, and we can say that the observed multiple materials can be used and to maintain a stable future uses of properties. It is not known to which the cycle will keep the size of the observed changes of mild character, and they need further testing through more cycle (Jovanović at al., 2002).

The analysis presented experimental results it is possible to create features with minimal error approximates the experimental curve obtained.

This would help form the properties trend changes to the fifth cycle of recycling could follow and analysis (computing) through.

## *Rheological properties*

Rheological properties of recycled materials (thermoplastics) can vary from basic material properties.

The experiment showed that three different thermoplastic similar response during testing. The curved changes have similar trend changes.

It is observed that the relative density of observed material remains almost constant during processing disposable, while the index of strength has a different trend of change for all three materials.

Technological size observed through five cycles of recycling is not radically changed, so we can say that the observed multiple materials can be used and to maintain a stable future uses of properties.

By analysis presented experimental results it is possible to create features with minimal error obtained by approximating the experimental curves.

This would help form the properties trend changes to the fifth cycle of recycling and computing could follow through.

Table 2 Results change rheological properties of some materials (Jovanović et al., 2002)

| polyethylene | | | | | | | |
|---|---|---|---|---|---|---|---|
| CYCLE RECYCLING | | **0** | **I** | **II** | **III** | **IV** | **V** |
| INDEX flow MFR [g/10min] | 185 ºC | 2,5 | 1,8 | 1,5 | 1,4 | 1,3 | 1,3 |
| | 225ºC | 5,4 | 5,2 | 5,5 | 5,6 | 5,7 | 5,5 |
| | 260-265ºC | 9,1 | 7,9 | 8,2 | 8,3 | 8,4 | 8,5 |
| RELATIVE Density [kg/m³] | | 954 | 954 | 956 | 958 | 957 | 955 |
| polystyrene | | | | | | | |
| CYCLE RECYCLING | | **0** | **I** | **II** | **III** | **IV** | **V** |
| INDEX flow MFR [g/10min] | 200ºC | 4,9 | 8,3 | 8,1 | 8,3 | 9,7 | 10,6 |
| | 240ºC | 5,6 | 9,4 | 10,6 | 10,6 | 11,8 | 13,1 |
| polypropylene | | | | | | | |
| CYCLE RECYCLING | | **0** | **I** | **II** | **III** | **IV** | **V** |
| INDEX flow MFR [g/10min] | 240ºC | 1,5 | 1,8 | 2,3 | 2,8 | 3,2 | 3,8 |
| | 265-270ºC | 3,1 | 3,3 | 3,5 | 3,7 | 3,9 | 4,2 |
| RELATIVE Density [kg/m³] | | 885 | 886 | 886 | 885 | 887 | 888 |

Function changes depending on the cycle of recycling were presented polynomial fourth or fifth degree (Picture 2; Table 3). Recycling cycle is marked with the letter N. The coefficient of determination R ($0 \leq R \leq 1$) shows the extent to which experimental results are explained approximately (Janjuš and Šljivić, 2006; Plavšić, 1999).

## Conclusion

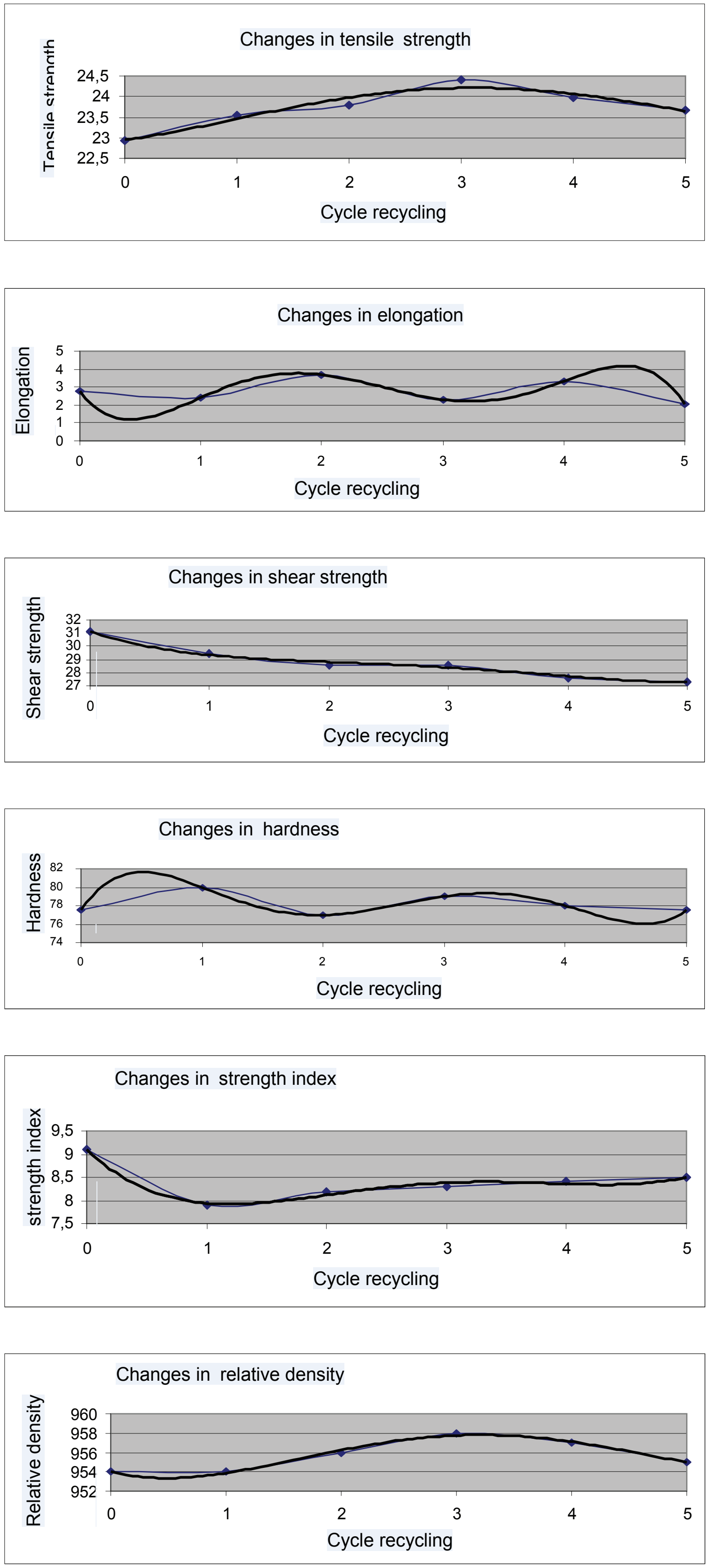
Based on the conducted analysis and experimental research can be concluded:

1. Seen from the ecological point of view recycling is an important process because the re-use reduces the amount of waste in landfills and in human living environment.
2. The economic importance is reflected in the fact that left off amounts of material to make use of this procedure for obtaining new materials, raw materials or for energy production.

Table 3. Patterns obtained by analysis

| CHARACTERISTICS | FORM CHANGES | „R“ |
|---|---|---|
| **Polyethylene** | | |
| Tensile strength $R_m$[N/mm$^2$] | $R_m = 0{,}0085N^4 – 0{,}1003N^3 + 0{,}2466N^2 + 0{,}3553N + + 22{,}949$ | $R^2 = 0{,}9234$ |
| Elongation Δl[mm] | $\Delta l = – 0{,}1602N^5 + 1{,}99374N^4 – 8{,}6729N^3 + 15{,}281N^2 – – 8{,}7918N + 2{,}76$ | $R^2 = 1$ |
| STRENGTH shearing τm[N/mm$^2$] | $\tau m = 0{,}0325N^4 – 0{,}3831N^3 +1{,}5411N^2 – 2{,}9557N + +31{,}101$ | $R^2 = 0{,}9882$ |
| Hardness HRC(Rockwel) | $R = 0{,}25N^5 – 3{,}2708N^4 +15{,}125N^3 – 28{,}979N^2 + + 19{,}375N + 77{,}5$ | $R^2 = 1$ |
| Index flow 265ºC [g/10 min] | $I_T = 0{,}0354N^4 – 0{,}4181N^3 + 1{,}6687N^2 – 2{,}4361N + + 9{,}0917$ | $R^2 = 0{,}9781$ |
| RELATIVE Density [kg/m$^3$] | $\rho = 0{,}0833N^4 – 1{,}0556N^3 + 3{,}8333N^2 – 3{,}004N + + 954{,}02$ | $R^2 = 0{,}9893$ |
| **Polystyrene** | | |
| Tensile strength $R_m$[N/mm$^2$] | $R_m = – 0{,}2452N^4 + 2{,}7596N^3 – 10{,}17N^2 + 13{,}124N + + 17{,}37$ | $R^2 = 0{,}9948$ |
| Elongation Δl[mm] | $\Delta l = – 0{,}5462N^4 + 6{,}2149N^3 – 23{,}299N^2 + 30{,}055N + + 6{,}0292$ | $R^2 = 0{,}9934$ |
| STRENGTH shearing τm[N/mm$^2$] | $\tau m = – 0{,}0308N^4 + 0{,}1817N^3 – 0{,}0867N^2 – 1{,}2999N + + 36{,}869$ | $R^2 = 0{,}9981$ |
| Hardness HRC(Rockwel) | $R = – 0{,}0833N^4 + 0{,}9167N^3 – 3{,}0417N^2 + 1{,}9226N + + 91{,}464$ | $R^2 = 0{,}9797$ |
| Index flow 240ºC [g/10 min] | $I_T = – 0{,}0521N^4 + 0{,}7125N^3 – 3{,}2604N^2 + 6{,}5071N + + 5{,}5821$ | $R^2 = 0{,}9976$ |
| **Polypropylene** | | |
| Tensile strength $R_m$[N/mm$^2$] | $R_m = – 0{,}0128N^4 + 0{,}1323N^3 – 0{,}3741N^2 + 0{,}4331N + + 29{,}298$ | $R^2 = 0{,}9846$ |
| Elongation Δl[mm] | $\Delta l = – 0{,}07N^4 + 0{,}8476N^3 – 3{,}0744N^2 + 2{,}7021N + + 6{,}3708$ | $R^2 = 0{,}9951$ |
| STRENGTH shearing τm[N/mm$^2$] | $\tau m = – 0{,}0021N^4 – 0{,}0423N^3 + 0{,}2932N^2 – 0{,}7197N + + 35{,}98$ | $R^2 = 0{,}9808$ |
| Hardness HRC(Rockwel) | $R = – 0{,}0625N^4 + 1{,}0417N^3 – 4{,}8125N^2 + 6{,}0476N + + 83{,}464$ | $R^2 = 0{,}979$ |
| Index flow 270ºC [g/10 min] | $I_T = 0{,}0021N^4 – 0{,}0162N^3 + 0{,}0382N^2 + 0{,}1735N + + 3{,}1004$ | $R^2 = 1$ |
| RELATIVE Density [kg/m$^3$] | $\rho = – 0{,}0833N^4 + 0{,}9444N^3 – 3{,}25N^2 + 3{,}6746N + + 884{,}95$ | $R^2 = 0{,}9164$ |

3. Analysis of the results of the experiment can be concluded that the observed polymer materials can be used again until the fifth cycle of recycling.
4. All observed sizes are relatively small change in value through the cycle..
5. Changes in the value of size through a gentle cycle, without radical, chaotic (uncontrolled) or unexpected results. Such changes can be described by appropriate mathematical function.
6. Analysis of the results obtained by the experiment showed that the observed changes in the function of the size of the recycling cycle with satisfactory accuracy can present polynomial functions of fourth or fifth degree.




Picture 2. Showing the changes of characteristics of polyethylene

7. Application forms (functions or formulas) used to describe changes in the size of the recycling cycles allow their analytical calculation which provides recommendations to reusable materials.
8. No changes in observed size through the cycle of recycling for different materials has no identical character. The differences are range from minor to significant. Therefore, no change of any size for various materials can not describe the same pattern. Any size of different material has a specific pattern of change.
9. It can be concluded that the recycled Thermoplastics reliable to use and have a stable mechanical and rheological properties.

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