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Studies on the Development of New Sustainable Materials Based on Recycled Low-density Polyethylene

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Abstract: *The paper presents our studies regarding the superior valorization of recycled low-density polyethylene (rLDPE) by compounding with thermoplastic starch (TPS) and ethylene propylene terpolymer elastomer (EPDM). Low-density polyethylene post-consumer waste from foil packaging was used for the experiments. The waste was mechanically recycled and the rLDPE granules obtained were characterized both from a physical-mechanical and structural point of view. In order to obtain new sustainable materials, rLDPE granules were mixed with TPS, EPDM, compatibilizers and crosslinking agents. The mixtures were obtained in a PlastiCorder Brabender mixer at 140°C, 30-80 rpm, working time 7 minutes. The obtained samples show very good resistance to abrasion, have very good values of Charpy impact strength and tensile strength, show very good behaviour to accelerated aging and to the action of some liquids, they have high hardness (51-53) °ShD and a Vicat softening point of 93-96°C. The new materials can be processed by methods specific to plastic materials (extrusion, injection, compression) in order to obtain finished products, and their fields of application can include: the footwear industry, the automotive industry, construction, packaging, agriculture, etc. The thermal analyses showed that up to a temperature of 130°C, the samples have good thermal stability, the mass loss being 0.33-0.79%. The LCA analysis of the composites shows a low environmental impact. The values of the carbon footprint range between 0.58 Kg CO_2 eq/kg and 0.75 Kg CO_2 eq /kg due to the use of recycled low-density polyethylene and optimised efficient production process.*

Keywords*: recycled low-density polyethylene, physical-mechanical properties, LCA, carbon footprint*

1. Introduction

In the last decades, plastic materials have experienced a remarkable development due to their low-cost price and very good properties that have led to their use in many domains. Because these materials are not biodegradable, plastic waste represents a danger to the environment. For these reasons, measures have been taken to reduce the amount of plastic waste through various methods such as mechanical recycling, incineration, the use of biodegradable plastic materials, etc. The production of plastic materials worldwide in 2021 was 390.7 million tonnes (Mt), and had an increase of about 4% compared to 2020. This was composed of: 352.3 Mt (90.2%) fossil-based plastics, 32.5 Mt (8.3%) post-consumer recycled plastics and 5.9 Mt (1.5%) bio-based plastics. Of this, 44% was used only for obtaining packaging. The European plastic production increased to 57.2 Mt in 2021 (with an increase of 6.1% compared to 2020), of which: 50.1 Mt fossil-based plastics, 5.8 Mt post-consumer recycled plastics and 1.3 Mt bio-based plastics [1, 2].

Mechanical recycling is one of the ecological methods of reducing the amount of plastic waste [1, 3]. For this, the waste is collected and sorted by types of categories of plastic materials in the collection centres. Then it is transported to mechanical plastic recycling companies.

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The process involves going through several technological stages. The first operation is the sorting into production batches depending on the type of polymer, colour, etc. Then it is moved on to the operation of grinding into flakes, followed by washing and drying the flakes. Dry flakes, if they are homogeneous, are directed to storage and then to processing by extrusion-granulation. Otherwise, the washing and drying operation is repeated or, if necessary, additional operations are used such as advanced separation to eliminate some contaminants, or separation of waste according to the type of polymer. Among the methods used, we mention: spectroscopic method [4, 5], selective dissolution of polymers [6], thermal adhesion method [7], froth flotation method [8], electrostatic separation methods [9-10] and others. At the same time, in order to improve the physical, mechanical, biodegradation or processing properties, the following can be added to the recycled plastic materials: compatibility agents, mineral additives, other polymers, natural fibres, plasticizers, antioxidants, etc. [1, 11].

Our work is part of these trends because it aims to reuse post-consumer plastic waste obtained from packaging and develop new materials that are partially biodegradable by introducing a fully biodegradable component, namely thermoplastic starch (TPS), into the composition. There are several specialized works already published that studied the possibilities of obtaining partially biodegradable materials based on low-density polyethylene (LDPE) [12-14]. Compared to the existing studies, in our work, recycled low-density polyethylene (rLDPE) is used as a polymer matrix, the mixtures contain a small amount of ethylene-propylene terpolymer rubber (EPDM), crosslinking agents and compatibilizers. The mixtures are obtained in a PlastiCorder Brabender mixer at 140°C, by mixing the elastomer and the other ingredients with the plastic material in a molten state, under strong shearing forces. The purpose of using this method is to obtain new materials with improved properties as a result of a good homogenization of the existing ingredients and especially those resulting from the contamination of rLDPE with other polymers during the mechanical recycling process [15, 16]. The samples were obtained in laboratory conditions and were characterized from a physical-mechanical and chemical point of view.

2. Materials and methods

2.1. Materials

The following materials were used to obtain the mixtures: (1) mechanically recycled low density polyethylene, in the form of yellow granules, with a humidity of 0.001% and an amount of ash of 2.2%; (2) thermoplastic starch obtained from soluble starch and glycerine from Lach-Ner, Czech Republic, to which citric acid anhydrous from Reanal Laborvegyszer Kft., Budapest, Hungary, is added as compatibilizer (3) Nordel 47310 ethylene-propylene terpolymer rubber (EPDM) from Dupont Elastomer; (4) compatibilizer for polymer mixtures: Admer NF 468E polyethylene grafted with maleic anhydride (LDPE-g-MA) from Mitsui Chemicals Europe GmbH, Germany, (5) a crosslinking agent, Luperox F40 di-2-tert-butylisopropyl benzene from Alkema and a crosslinking coagent, trimethylolpropane trimethacrylate (TMPTMA) Alcanpoudre TMPT MA 70 from Safic Alcan.

2.2. Mechanical recycling of low-density polyethylene post-consumer waste

For the experiments, sorted low-density polyethylene (LDPE) post-consumer waste from foil-type packaging was used. This type of waste was mechanically recycled using a "state-of-the-art" mechanical recycling facility at S.C. Monofil SRL, Săvinești, Romania. The stages of the technological process were: re-sorting – depending on the type of polymer or the colour of the packaging, shredding, washing and drying of the sorted waste in the shredding, washing and drying installation, followed by re-granulation, checking the rLDPE quality and packaging.

2.3. Obtaining thermoplastic starch (TPS)

Two types of thermoplastic starch were obtained. For the first type, starch and plasticizer in a starch:glycerol mass ratio of 70:30 were mixed and then introduced into the PlastiCorder Brabender internal mixer at 100-120°C, mixing speed of 30-80 rotations per minute (rpm), for 7 min, where the

process of starch gelatinization took place in order to obtain thermoplastic starch. The second type of thermoplastic starch additionally contains a compatibilizer, namely citric acid. In this case, the mass ratio of the starch:glycerol:citric acid mixture components was 70:30:2. The work method is similar to the one presented above.

2.4. Obtaining mixtures and test samples

The composition of the blends made is presented in Table 1. The polymeric mixtures were obtained using a Plasti-Corder Brabender internal mixer, at 140°C for 7 min. The rotor speed was 30 rpm in the first 3 min and increased to 80 rpm for the next 4 min. Figure 1 shows the variation of the torque and the temperature over time when obtaining the mixture PA 2-2 and Figure 2 shows the variation of the torque versus time when obtaining the blends on Plasti-Corder Brabender. The processing characteristics obtained on the Brabender Plasticorder are presented in Table 2. The variations in temperature and torque are due to introducing the ingredients (the first 3 min), as well as the increase in temperature with the reduction of viscosity under the influence of shear forces. The specific energy of the mixtures is higher in the mixtures containing a larger amount of rLDPE. They confirm the working method specified previously and indicate a good mixing of the ingredients [17-18].

Table 1. Composition of mixtures

Ingredients	**Sample code**			
	PA1-1	**PA1-2**	**PA2-1**	**PA2-2**
rLDPE, g	150	200	150	200
Plasticized starch – type 1, g	75	25	-	-
Plasticized starch – type 2, g	-	-	76.5	25.5
LDPE-g-AM, g	10	10	10	10
EDPM, g	25	25	25	25
Luperox F40, g	0.2	0.1	0.2	0.1
TMPTMA, g	0.2	0.1	0.2	0.1

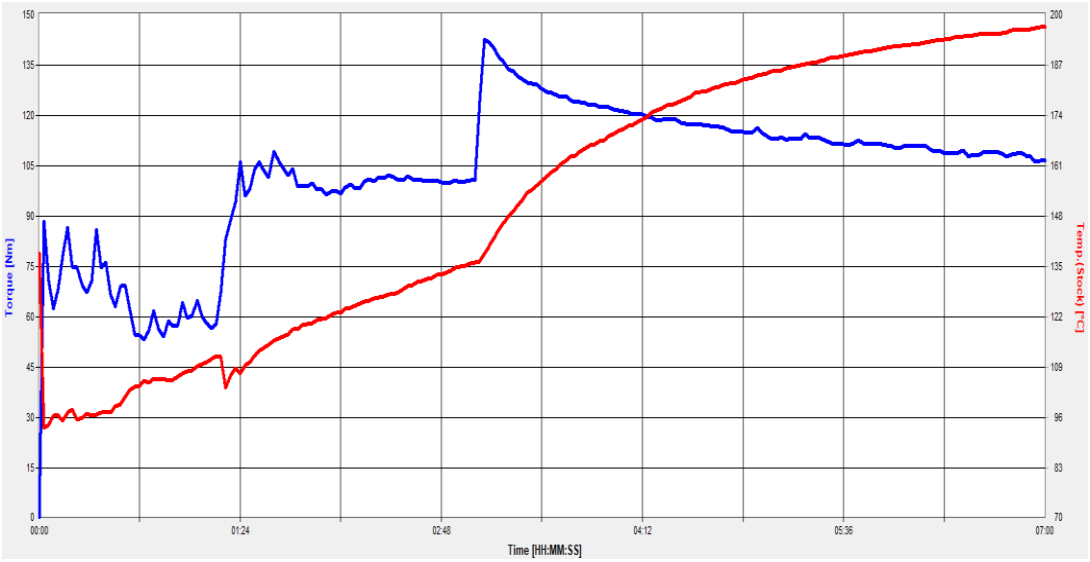


Figure 1. Torque and temperature variation for sample PA2-2

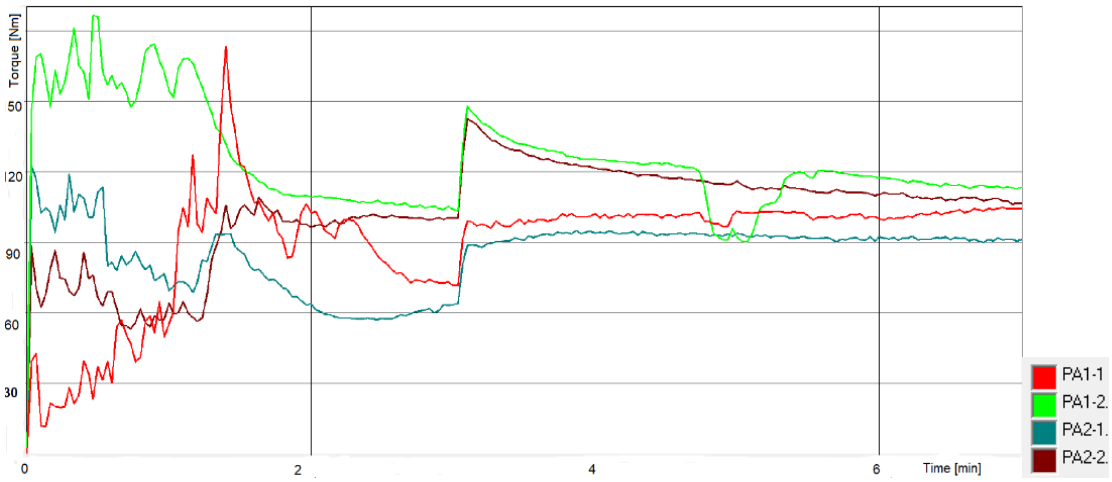


Figure 2. Diagram showing the variation of the torque versus time when obtaining the blends on Plasti-Corder Brabender

Obtaining the samples necessary for the physical-mechanical determinations was carried out on a laboratory press and took place in three stages: (1) preheating for 2 min at a temperature of 150°C; (2) modelling for 5 min at 150°C and 300 kN compression force; (3) cooling for 8 min at 45°C and 300 kN compression force. Plates with a thickness of 2 mm and 6 mm were made, from which the specimens necessary for physical-mechanical determinations were obtained by punching. The samples were conditioned for 16 h at room temperature.

Table 2. The characteristics obtained on Plasti-Corder Brabender during the preparation of the samples

Sample code	Specific energy, [kNm/g]	Torque variation range, [Nm]	Temperature variation range, [°C]
PA1-1	1.1	0-147.9	97-145
PA1-2	1.4	109.7-186.6	111-205
PA2-1	1	89.9-122.6	103-181
PA2-2	1.3	53.2-140.2	93-200

2.5. Specimen characterization

Tensile strength and elongation at break were carried out using dumb-bell shaped specimens according to ISO 37, using a Schopper strength tester at a crosshead speed of 50 mm/min. Residual elongation is the elongation of a specimen measured 1 min after rupture in a tensile test.

Hardness was measured in °Sh D (scale specific for hard materials), using 6-mm thick samples according to ISO 48-4.

Resilience was determined according to ISO 46662 using a Schoob test machine and 6 mm thick samples. Measurements were performed in triplicates and the resulting values were the average of 3 measurements.

Accelerated ageing trial was carried out according to ISO188 using the hot air circulation oven method. The test took 7 days at a temperature of 70 ± 1°C, subsequently comparing the results with those from samples not subjected to ageing.

Melt flow index (MFI) of the samples was measured by extruding the mixtures through a 2mm die capillary rheometer - Haake Melt Flow MT at 190°C or 200°C, and a 5 kg force was used, according to ISO 1133. Each data point was the result of three measurements.

The densities of samples were measured according to ISO 2781.

Abrasion resistance was determined according to ISO 4649, the cylinder method, using a force of 10 N. The cylindrical shaped samples with a 16 mm diameter and minimum 6 mm height were obtained by rolling and pressing the mixtures, and cutting with a rotating die. Abrasion resistance was expressed by relative volume loss in relation to calibrated abrasive paper.

The action of liquids was determined according to ISO 1817, monitoring volume and mass variation using the volumetric and gravimetric methods. Immersion time was 22±0.25 h. The samples used had a volume of 1-3 cm^3 and a uniform thickness of 2±0.2 mm.

Charpy shock resistance was determined on type 1 specimens, notch type A, using pendulum 5J (according to ISO 179) and determination of flexural properties was achieved according to ISO 178 - stress at the maximum test speed (2 mm/min).

Vicat softening temperature was determined according to ISO 306 – method A50 – 1kg.

Fourier Transform Infrared Spectroscopy (FTIR) spectra of samples were obtained using Nicolet iS50 FT-IR spectrophotometer in the wavenumber ranging from 400 cm^{-1} to 4000 cm^{-1}, using attenuated total reflection (ATR).

Thermal stability of the samples was investigated with a STA 449C Jupiter from Netzsch. Samples of ~ 15 mg were placed in an Al_2O_3 crucible and heated up to 600ºC in air with a speed of 10ºC/min.

In order to perform LCA, GaBi ts (Professional) software and ecoinvent database were used, that incorporate emission factors associated to processes and materials, as well as data provided by the producer and collected in the Life Cycle Inventory (Figure 3). For the analysed composite a new project and a Plan were created.

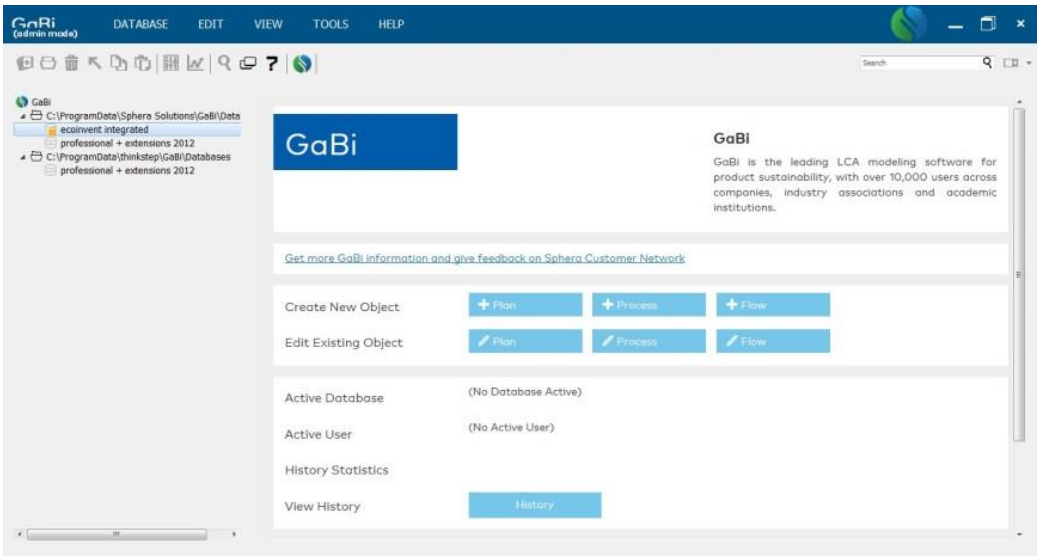


Figure 3. GaBi software interface

The functional unit is a very important element in the study of carbon footprint and water footprint, all inventory data (inputs and outputs of the system) are related to it. In addition, the results obtained must be reported by functional unit. For this study, the functional unit refers to 1 Kg of composite.

3. Results and discussions

3.1. Physical-mechanical characteristics of rLDPE

The rLDPE granules were analysed from the point of view of physical-mechanical properties and the results obtained are presented in Table 3. The results obtained for the characterization of mechanically recycled polyethylene waste - rLDPE, according to data from the literature [19-21] are close in value to those of virgin polymers.

Table 3. Physical-mechanical characteristics of rLDPE

Characteristic	**rLDPE**
Hardness, ^{0}ShD	53±0.58
Resilience, %	22±1.00
Tensile strength, N/mm^2	12.7±0.75
Elongation at break, %	660±50.20

Vicat softening temperature, ^{0}C	97±2.00
Charpy impact strength, J/m^2	48.39±2.04
Flexural Strength, MPa	No breaking
Flow index, g/10'(at 190°C with a load of 2.16 kg)	0.620±0.02

3.2. FTIR analysis of rLDPE

The structural characterization (FTIR) of rLDPE was carried out, and the FTIR spectra obtained (Figure 4), according to the literature [22-25], confirm the fact that the recycled granules come from LDPE waste because they contain the absorption bands at 2915 cm^{-1} and 2848 cm^{-1} which are attributed to the symmetric and asymmetric stretching vibrations of the methylene group, the double absorption bands specific to the amorphous and crystalline phase located at 1463 cm^{-1} and at 719-730 cm^{-1} attributed to the bending and in-plane (rotation) deformation vibration of the methylene group (-CH_2-). In addition, compared to virgin LDPE, in rLDPE we can observe the existence of two low intensity absorption bands at 1696 cm^{-1} and 1258 cm^{-1}, which could be due either to the existence of dyes, compatibilizers or other ingredients, or to possible degradation reactions with the formation of aldehydes, ketones, carboxylic acids, etc. [26, 27].

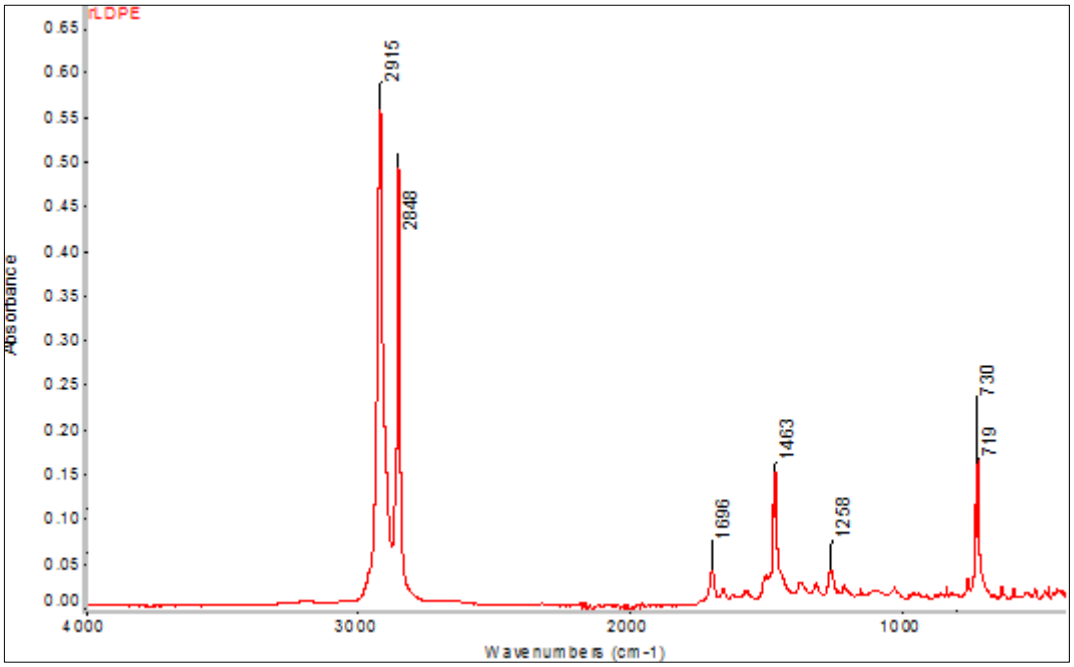


Figure 4. FTIR spectrum of rLDPE

3.3. Physico-mechanical and chemical characterization of the samples

The characteristics of the samples with the rLDPE polymer matrix are presented in Tables 4-5 and Figures 5, 6. From the obtained results, it can be seen that they are influenced by both the composition and the production technology.

Most of the samples show a behaviour specific to plastic materials, with the formation of a "neck" under the action of stretching forces (high residual elongation) [28] The density of the samples is low (0.97-1.05 g/cm^3) and the abrasion resistance has high-performance values (below 52.07 mm^3), specific to composites with polymer matrix based on polyethylene [29].

The samples with a higher TPS content (PA1-1 and PA2-1) have weaker tensile strength and abrasion resistance in normal state than those with a lower TPS content, indicating that these properties are additive and depend on the properties of the components of the mixtures. The samples containing citric acid (PE2-1 and PA2-2) show an improvement in tensile strength and abrasion resistance, and are more compact (higher specific gravity) than those with the same composition (PE1-1 and PA1-2) but which

do not contain citric acid, indicating that it led to an improvement in the compatibility between the phases [30].

All mixtures can be processed by methods specific to plastic materials (compression, injection, extrusion), flow index at 190°C with 5 kg load for the analysed samples had values of 0.235-1.09 g/10' (Table 4), with the exception of sample PA1-1 which requires different work parameters for processing in the molten state (namely, at 200°C with 5 kg load, the value of 3.54±1.07 g/10' was obtained).

Hardness and tensile strength increase after accelerated aging. The variation of hardness is max +4 °ShD and the variation of breaking strength is -2.09% – +59.63% (Figure 5). This behaviour may indicate a continuation of the process of crosslinking and stabilization of the sample during maintenance at a temperature of 70°C for 168 hours, obtaining higher values for these characteristics than those obtained for rLDPE.

The behaviour of the samples when immersed in water and toluene is very good, with mass variations below 10% for toluene and below 0.5% for distilled water and volume variations of a maximum of 23.71% in water and 35.63% in toluene (Figure 5).

The samples have a softening temperature of 93-96°C and good values of Charpy impact strength (36-39 J/m^2), but values are 1-4°C, respectively 20.1-23.8% lower than those obtained in the case of rLDPE. The flexural strength (ability to resist deformation under load) of the analysed samples was very good, the samples did not break under the test conditions (test speed 2 mm/min) (Table 5).

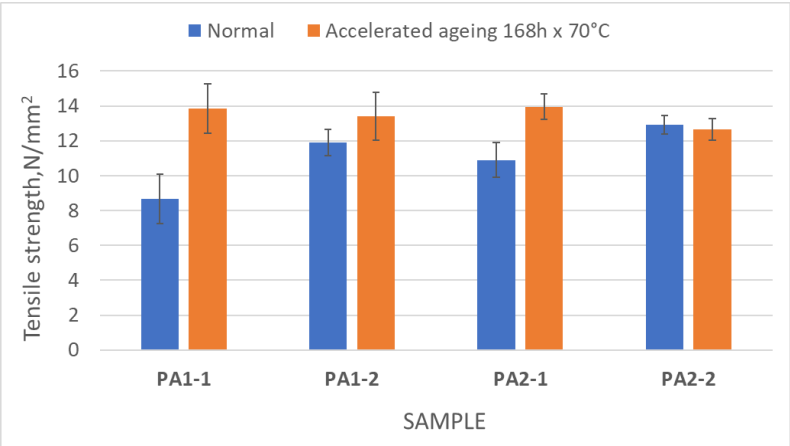
Table 4. Values of density, resistance to abrasion and flow index for the analysed samples

Property	**Sample code**			
	PA1-1	**PA1-2**	**PA2-1**	**PA2-2**
Density, *g/cm^3*	1.00±0.015	0.97±0.008	1.05±0.008	0.98±0.005
Abrasion resistance, *mm^3*	52.07±3.840	30.51±0.290	51.38±3.970	25.72±1.460
Flow index, *g/10 min.* (at 190°C with a load of 5 kg)	3.54±1.07*	1.09±0.260	0.377±0.200	0.938±0.091

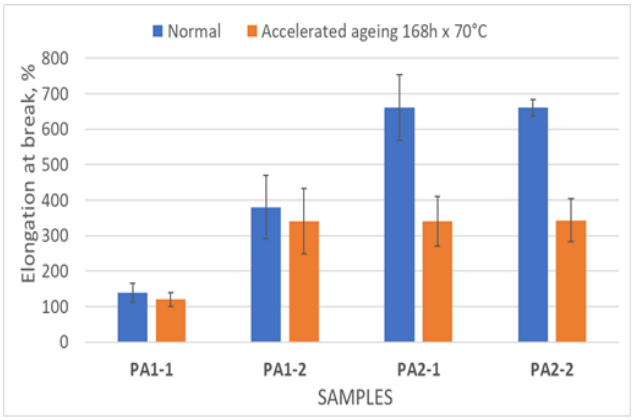
* Flow index at 200°C with 5 kg load

Table 5. Values of Charpy impact strength, flexural strength and Vicat softening temperature of samples

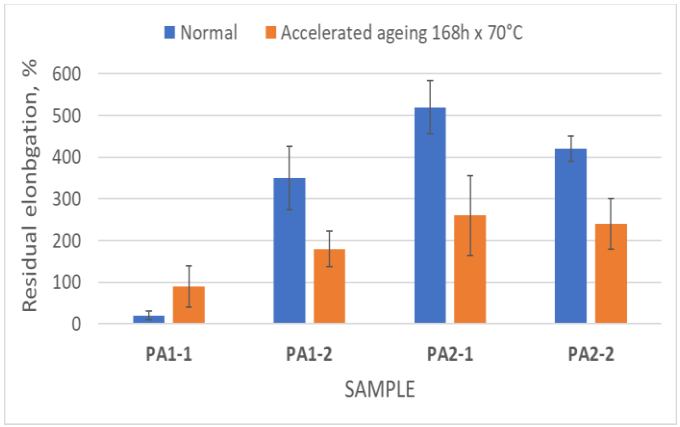
Sample code	*Charpy impact strength, J/m^2*	*Vicat softening temperature, ^{0}C*	*Flexural Strength, MPa*
PA2-1	36.85±5.65	93±2.00	No breaking
PA2-2	38.68±3.08	96±2.00	No breaking



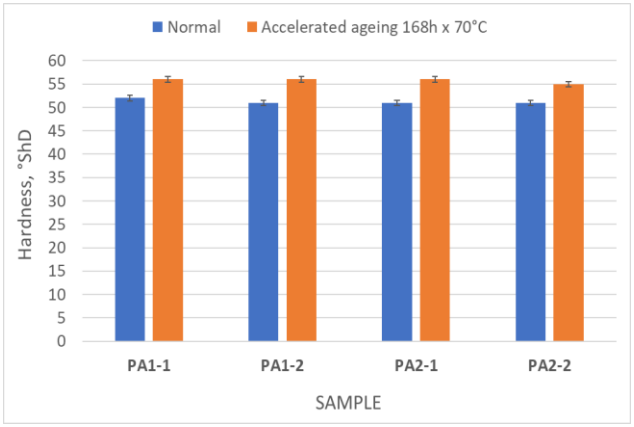
a.



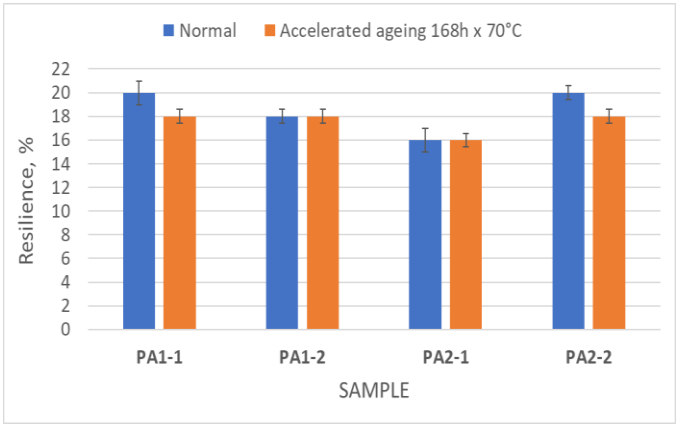
b.



c.

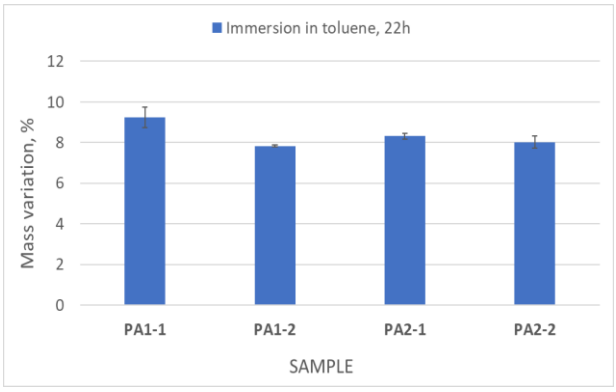


d.

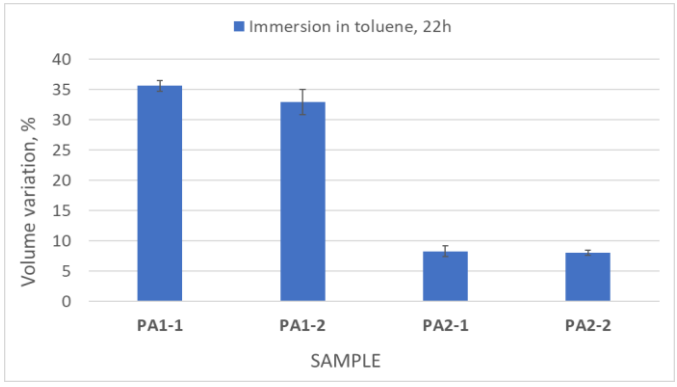


e.

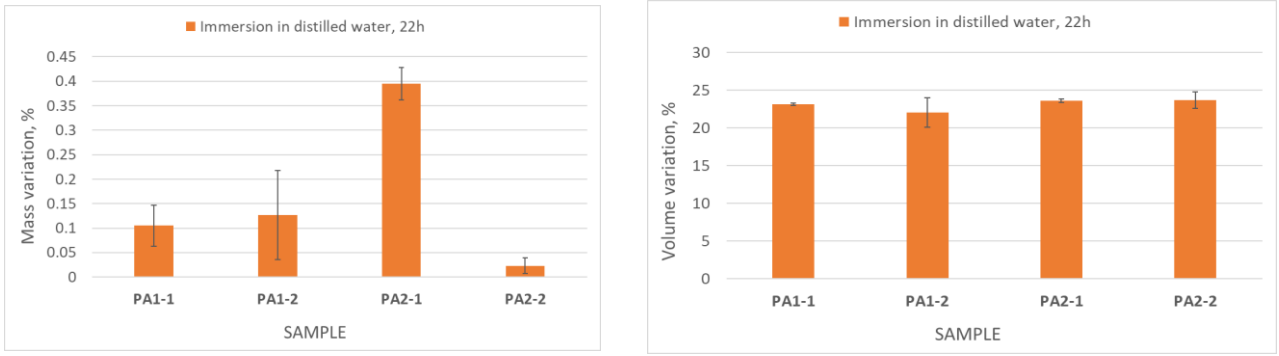
Figure 5. Physical-mechanical characterisation of samples in normal state and after accelerated ageing for 168 hours at 70°C: tensile strength (**a**); elongation at break (**b**); residual elongation (**c**); hardness (**d**); resilience (**e**)



a1



a2



b₁ ***b₂***

Figure 6. Chemical characterization of samples: (a) Resistance to immersion in toluene, 22 h: mass variation (a_1) and volume variation (a_2), (b) Resistance to immersion in distilled water, 22 h: mass variation (b_1) and volume variation (b_2)

3.4. Thermal analysis (TG-DSC)

The thermogravimetric (TG) and differential scanning calorimetry (DSC) curves are presented in Figure 7. The endothermic effect with minimum at ~110ºC that is observable on the DSC curves is attributed to the sample melting, the onset being around 97ºC. The higher amount of plasticized starch from samples PA1-1 and PA2-1 leads to slightly lower temperatures for the endothermic peak, with ~ 2ºC, indicating the faster completion of the melting process. Samples have a good stability up to 130ºC, with a mass loss of under 1% (Table 6). This is due to elimination of moisture (residual water absorbed by the plasticized starch present in the samples) [31]. The samples PA1-1 and PA2-1 that have larger amounts of plasticized starch exhibit a corresponding larger mass loss when compared with PA1-2 and PA2-2.

Table 6. Principal data from thermal analysis

Sample	T5%	T10%	Mass loss RT-130ºC	Mass loss 130-285ºC	Mass loss 285-375ºC	Melting onset/peak ºC	Exo I ºC
PA1-1	222ºC	292ºC	0.79%	8.12%	13.25%	96.9/109.9	253.4
PA1-2	293ºC	332ºC	0.33%	4.08%	14.70%	96.9/112.1	255.6
PA2-1	205ºC	259ºC	0.77%	12.55%	23.54%	97.4/111.1	260.8
PA2-2	271ºC	311ºC	0.39%	5.68%	16.38%	96.9/113.0	251.0

The mass loss in the temperature interval 130-285ºC is also correlated to the plasticized starch quantity from the samples. The high starch content of PA1-1 and PA2-1 leads to larger mass loss in case of these samples when comparing to PA1-2 and PA2-2, indicating that in this temperature interval the oxidative-degradation of the plasticized starch takes place (as indicated by the small exothermic effect on the DSC curves) [32].

In conclusion, the presence of larger amounts of plasticized starch leads to lower thermal stabilities as indicated also by $T_{5\%}$ and $T_{10\%}$ values. Nevertheless, these differences are sizable at high temperatures, over 200ºC, where the polymeric blends are already melted and do not impact the usability of items made for ambient temperature use. The differences between the two plasticized starch types leads to lower thermal stability when citric acid is used, the corresponding mass loss for PA2 series being larger than for PA1 series. By contrast, the PA2 series containing citric acid exhibit a slower melting process, requiring about 1ºC for completing.

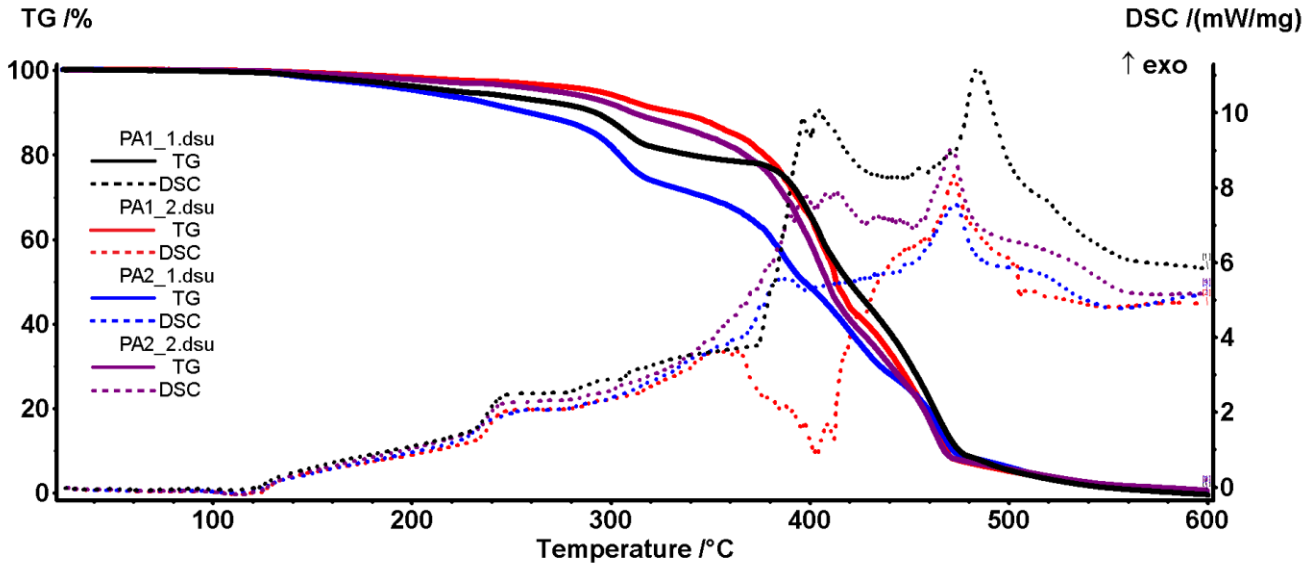


Figure 7. The thermal analysis (TG and DSC curves) for PA1-1, PA1-2, PA2-1 and PA2-2 samples

3.5. Life Cycle Assessment of the composites

Figure 8 shows the flow diagram of studied types of composite PA2-2. The LCA study was performed for 4 types of composites based on rLDPE and EPDM.

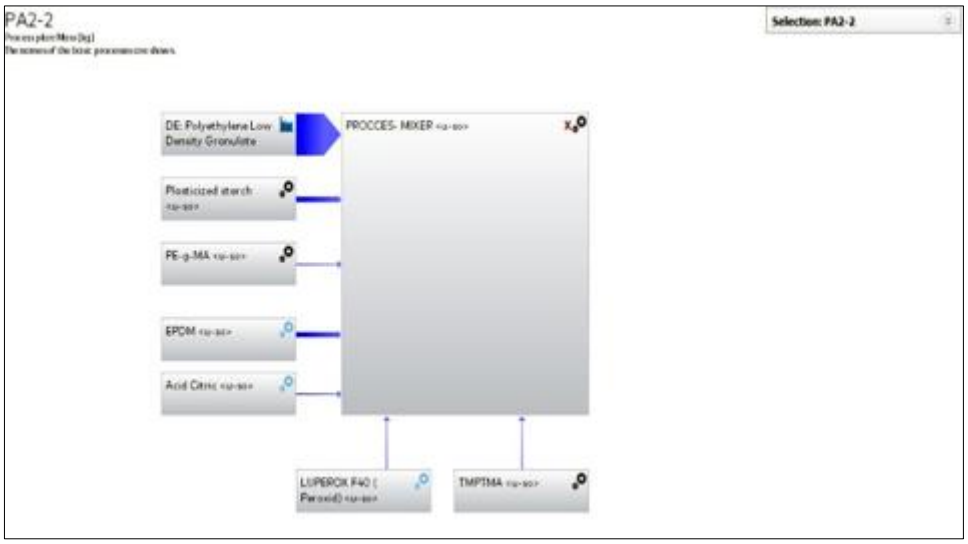


Figure 8. Flow diagram of the system under study

Values of carbon footprint for the obtained composites fall between 0.75 Kg CO_2eq/kg for PA2-1 and 0.58 Kg CO_2eq /kg composite PA1-2, based on rLDPE and EPDM (Figure 9). The analysis of the results indicates a low environmental impact for all studied polymer blends, mainly due to the selection of sustainable materials and the efficient production process [33-35]. The lowest carbon footprint is achieved, mainly by using the recycled LDPE in large amounts and reducing the amount of other chemicals, like starch.

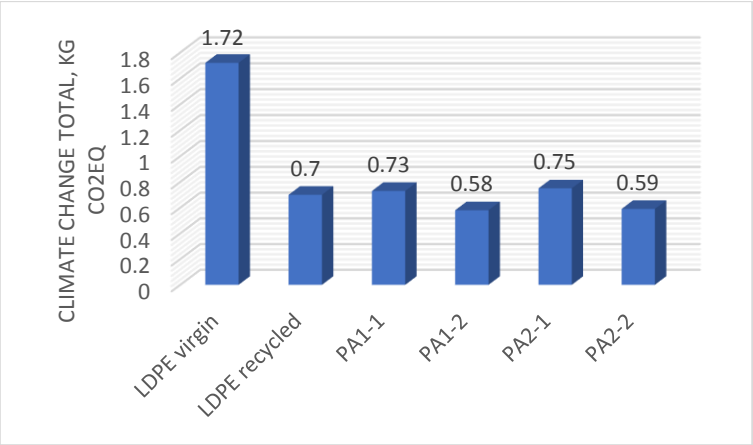


Figure 9. Carbon footprint for the composites

4. Conclusions

Research was carried out to obtain new sustainable materials that can be used in the footwear industry, the automotive industry, construction, packaging, agriculture, etc. Starting from the existing data in the literature, mechanically recycled low-density polyethylene waste was selected as the polymeric matrix. To obtain a partially biodegradable material, thermoplastic starch was added to the mixture, which is known to be completely biodegradable. In order to obtain products with high-performance properties, suitable for the intended fields of use, compatibilizers and other ingredients were added to the samples. The use of rLDPE instead of virgin LDPE reduced the amounts of other chemicals, thus contributing to obtaining low carbon footprint composites, having low environmental impact.

Due to the composition and the selected working method, the samples obtained have very good characteristics, namely: hardness of 51-56°D, resilience of 16-20%, tensile strength of 8.67-13.95 N/mm^2, elongation at break of 100-660 %, low specific weight of 0.97-1.05 g/cm^3, very good abrasion resistance of max. 52.07 mm^3, mass variation after 22 h of immersion in water of max 0.395%, mass variation after 22 h of immersion in toluene of max 35.63%, Vicat softening temperature of 93-96°C, Charpy impact strength of 38.85-36.68 J/m^2, very good flexural strength (without breaking) and a very good behaviour after accelerated aging for 168 h at 70°C. The thermal properties are influenced by the TPS content, but the samples show good thermal stability up to a temperature of 130°C, with a mass loss of 0.33-0.79%. They can be processed into finished products by methods specific to plastic materials, and the resulting pre-consumer waste can be shredded and re-introduced into the technological flow without significantly changing the properties of the finished products. Due to their physical-mechanical and chemical properties, the new materials can be used in making several types of finished products with applicability in various fields.

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References

1. CONSTANTINESCU, D., BOATA, B., IORDACHE, M., STELESCU, M.D., GEORGESCU, M/, SÖNMEZ, M., Technological considerations regarding the mechanical recycling of waste from polyethylene and polypropylene packaging, *Proceedings of the 9th International Conference on Advanced Materials and Systems* Bucharest, Romania, 26-28 October, 2022, pp.401-406, https://doi.org/10.24264/icams-2022.IV.3
2.***https://plasticseurope.org/wp-content/uploads/2022/10/PE-PLASTICS-THE-FACTS_V7-Tue_19-10-1.pdf, (accessed on 20.07.2023).

3. AL-SALEM, S., LETTIERI, P., BAEYENS, J., Recycling and Recovery Routes of Plastic Solid Waste (PSW): A Review, *Waste Manage.*, 29, 2009, 2625–2643, https://doi.org/10.1016/j.wasman.2009.06.004
4. ADARSH, U.K., BHOJE GOWD, E., BANKAPUR, A, KARTHA, V.B., CHIDANGIL, S., UNNIKRISHNAN, V.K, Development of an Inter-confirmatory Plastic Characterization System Using Spectroscopic Techniques for Waste Management, *Waste Manage.*, 150, 2022, 339–351, https://doi.org/10.1016/j.wasman.2022.07.025.
5. BONIFAZI, G., CAPOBIANCO, G. AND SERRANTI, S., A Hierarchical Classification Approach for Recognition of Low-Density (LDPE) and High-Density Polyethylene (HDPE) in Mixed Plastic Waste Based on Short-Wave Infrared (SWIR) Hyperspectral Imaging, *Spectrochim. Acta A Mol. Biomol. Spectrosc*. 198, 2018, 115–122, https://doi.org/10.1016/j.saa.2018.03.006
6. PAPPA, G., BOUKOUVALAS, C., GIANNARIS, C., NTARAS, N., ZOGRAFOS, V., MAGOULAS, K., LYGEROS, A., TASSIOS, D., The Selective Dissolution/Precipitation Technique for Polymer Recycling: A Pilot Unit Application, *Resources, Conservation and Recycling*, 34(1), 2001, 33-44, https://doi.org/10.1016/S0921-3449(01)00092-1
7. TALL, S., "Recycling of Mixed Plastic Waste – Is Separation Worthwhile?", PhD Thesis, Department of Polymer Technology, Royal Institute of Technology, 2000, Stockholm, Sweden.
8. KOKKILIÇ, O., MOHAMMADI-JAM, S., CHU, P., MARION, C., YANG, Y. AND WATERS, K.E., Separation of Plastic Wastes Using Froth Flotation – An Overview, *ADV COLLOID INTERFAC*, 308, 2022, 102769, https://doi.org/10.1016/j.cis.2022.102769.
9. SILVEIRA, A.V.M., CELLA, M., TANABE, E.H. and BERTUOL, D.A., Application of Tribo-Electrostatic Separation in the Recycling of Plastic Wastes, *PROCESS SAF ENVIRON*, 114, 2018, 219–228, https://doi.org/10.1016/j.psep.2017.12.019.
10.WU, G., LI, J. and XU, Z., Triboelectrostatic Separation for Granular Plastic Waste Recycling: A Review, *Waste Management*, 33, 2013, 585–597, https://doi.org/10.1016/j.wasman.2012.10.014
11. AZEEZ, T.O. (2019), "Thermoplastic Recycling: Properties, Modifications, and Applications", in: EVINGÜR G.A., PEKCAN Ö. and ACHILIAS D.S. (eds.), *Thermosoftening Plastics*, https://doi.org/10.5772/intechopen.81614.
12. KABOORANI, A., GRAY, N., HAMZEH, Y., ABDULKHANI, A., SHIRMOHAMMADLI, Y. Tailor-ing the low density PE - thermoplastic starch composites using cellulose nanocrystals and compatibilizer, *Polymer Testing*, 93, 2020, 107007, doi.org/10.1016/j.polymertesting.2020.107007
13. PERES, A. M., PIRES, R.R., ORÉFICE, L.R. Evaluation of the effect of reprocessing on the structure and properties of low density polyethylene/thermoplastic starch blends, *Carbohydrate Polymers*, 136, 2016, 210-215, https://doi.org/10.1016/j.carbpol.2015.09.047.
14. MAZEROLLES, T., HEUZEY, M.C., SOLIMAN, M., MARTENS, H., KLEPPINGER, R., HUNEAULT M.A., Development of co-continuous morphology in blends of thermoplastic starch and low-density polyethylene, *Carbohydrate Polymers* 206, 2018, 757-766, doi.org/10.1016/j.carbpol.2018.11.038.
15. VAN DUIN, M., MACHADO, A. V., EPDM-based thermoplastic vulcanisates: Crosslinking chemistry and dynamic vulcanisation along the extruder axis, *Polymer Degradation and Stability*, 90(2), 2005, 340-345, doi.org/10.1016/j.polymdegradstab.2005.04.050
16. MACHADO, A. V., M. VAN DUIN, Dynamic vulcanisation of EPDM/PE-based thermoplastic vulcanisates studied along the extruder axis, *Polymer*, 46(17), 2005, 6575-6586, doi:10.1016/j.polymer.2005.05.011
17. STELESCU, M.D., SONMEZ, M., ALEXANDRESCU, L. NIȚUICĂ, M., GURAU, D.F., GEORGESCU, M., Structure and properties of blends based on vulcanized rubber waste and styrene–butadiene–styrene thermoplastic elastomer. *J Rubber Res* 25, 2022, 421–434. https://doi.org/10.1007/s42464-022-00187-y

18. MISHRA S., CHATTERJEE A., RANA V. K., Polymer nanoparticles: their effect on rheological, thermal, and mechanical properties of linear low-density polyethylene (LLDPE), *Polym. Adv.Technol.*, 22(12), 2011, 1802-1811, DOI:10.1002/pat.1674
19. WARD, I.M., Mechanical properties of solid polymers, John Wiley, 2nd edition 1990, p. 357
20. PEREZ, J. O., Physique et mécanique des polymères amorphes", Lavoisier, Tech. et Doc., Paris, 1992, p. 360
21. GHITA, E., RAINER-GILLICH, G., BORDEAU, I., VODA, M., TROI, C. (2007), Aspects concerning polymers behaviour under tension, *Mater. Plasr.*, **44**(2), 2007, 158-162.
22. DEPAN, D., CHIRDON, W., KHATTAB, A., Morphological and Chemical Analysis of Low-Density Polyethylene Crystallized on Carbon and Clay Nanofillers, *Polymers*, 13, 2021, 1558, https://doi.org/10.3390/ polym13101558
23. AGOSTI E., ZERBI G., WARD I.M., Structure of the skin and core of ultra drawn polyethylene films by vibrational spectroscopy, *Polymer*, 33, 1992, 4219–4229.
24. PAINTER, P.C., HAVENS, J., HART, W.W., KOENIG, J.L., Fourier-transform IR spectroscopic investigation of polyethylene single-crystals. 1. Fine structure of CH_2 rocking mode. *J. Polym. Sci. Pol. Phys*, 15, 1997, 1237–1246.
25. HUANG, J.B., HONG, J.W., URBAN, M.W., Attenuated total reflectance Fourier transform infrared studies of crystalline amorphous content on polyethylene surfaces. *Polymer*, 33, 1992, 5173–5178, https://doi.org/10.1016/0032-3861(92)90797-Z.
26. SMITH B.C., The Infrared Spectra of Polymers II: Polyethylene, *Spectroscopy* 36(9), 2021, 24–29, https://doi.org/10.56530/spectroscopy.xp7081p7
27. COATES J., Interpretation of Infrared Spectra, A Practical Approach, in Encyclopedia of Analytical Chemistry, Editor Meyers R.A., pp 10815-10837, John Wiley & Sons Ltd, Chinchester, 2000.
28. ANDRIANOVA G.P., KARGIN V.A., Theory of neck formation in the stretching of polymers, *Polymer Science U.S.S.R.*, 12(1), 1970, 1-8, https://doi.org/10.1016/0032-3950(70)90271-6
29. NWAPA, C., OKUNWAYE, O. J., OKONKWO, C. L., CHIMEZIE, O. W, Mechanical Properties of High Density Polyethylene and Linear Low Density Polyethylene Blend, *SSRG International Journal of Polymer and Textile Engineering* (SSRG-IJPTE), 7(1), 2020, 23-28
30. KAHVAND F., FASIHI M., Plasticizing and anti-plasticizing effects of polyvinyl alcohol in blendwith thermoplastic starch, *Int J Biol Macromol*, 140, 2019, 775–781, https://doi.org/10.1016/j.ijbiomac.2019.08.185
31. STELESCU M.D., OPREA O.C., MOTELICA L., FICAI A., TRUSCA R.D., SONMEZ M., NITUICA M., GEORGESCU M., Obtaining and Characterizing New Types of Materials Based on Low-Density Polyethylene and Thermoplastic Starch, *J Compos Sci* 8(4). 2024. 134.
32. NITUICA M., OPREA O., STELESCU M.D., SONMEZ M., GEORGESCU M., ALEXANDRESCU L., MOTELICA L., Polymeric Biocomposite Based on Thermoplastic Polyurethane (TPU) and Protein and Elastomeric Waste Mixture, *Materials* 16(15), 2023, 5279.
33. GEORGESCU M., SONMEZ M., ALEXANDRESCU L., NITUICA (VILSAN) M., STELESCU M. D., GURĂU D., DRUSAN D.-A., CIOBANU A.-M, CHELARU C., Low Carbon Footprint Composite Based on Chloroprene Rubber and Elastomer Waste, *Proceedings of the 9th International Conference on Advanced Materials and Systems* Bucharest, Romania, 26-28 October, 2022, pp. 421-426, https://doi.org/10.24264/icams-2022.IV.6
34. SCHWARZ A.E., LIGTHART T.N., GODOI BIZARRO D., DE WILD P., VREUGDENHIL B., VAN HARMELEN T., Plastic recycling in a circular economy; determining environmental performance through an LCA matrix model approach, *Waste Management*, 121, 2021, 331-342, https://doi.org/10.1016/j.wasman.2020.12.020.
35. https://www.recyclingtoday.com/news/recycled-pp-hdpe-lower-carbon-footprint-pet/ (accessed on 01.08.2023)

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