

BIODEGRADABLE POLYMERIC COMPOSITE BASED ON RECYCLED POLYURETHANE AND RUBBER WASTES: MATERIAL FOR GREEN SHOE MANUFACTURING

Laurenția ALEXANDRESCU*, Mihai GEORGESCU, Maria SÖNMEZ, Mihaela NIȚUIĂ

INCDTP - Division: Leather and Footwear Research Institute, 93 Ion Minulescu St., sector 3, Bucharest, Romania, laurentia.

alexandrescu@icpi.ro

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BIODEGRADABLE POLYMERIC COMPOSITE BASED ON RECYCLED POLYURETHANE AND RUBBER WASTES: MATERIAL FOR GREEN SHOE MANUFACTURING

ABSTRACT. This paper presents the process of obtaining biodegradable polymeric composites based on post-consumption thermoplastic polyurethane and post-consumption vulcanized rubber waste, in various ratios (5, 10, 20, 30, 50%), cryogenically ground to min. 100 nm, and maleic anhydride grafted polyethylene as compatibilizer between the two phases. This composite will be formed into a low-density product, with low cost, recovery and reuse of waste, containing post-consumption polymers, and last but not least, biodegradable. The methodology for making the new materials involves the following steps: sorting waste, grinding, functionalization and compounding. These operations are easy to manage and do not involve new equipment. Compounding, the most important operation, will be carried out on a corotating twin-screw extruder-granulator with the possibility to adjust parameters such as temperature and speed depending on the material, having a screw ratio (L/D-35). The tested biodegradable composites were characterized structurally and physico-mechanically. Waste transformation (ground and functionalized) into new value-added products will lead to remarkable improvements in the life cycle of raw materials and the responsible use of this waste, contributing to sustainability, improving eco-efficiency and economic efficiency and reducing the "pressure" of waste on the environment.

KEY WORDS: biodegradability, polymeric composite, post-consumption, green footwear

COMPOZIT POLIMERIC BIODEGRADABIL PE BAZĂ DE POLIURETAN RECICLAT ȘI DEȘEURI DE CAUCIUC: MATERIAL PENTRU FABRICAREA ÎNCĂLȚĂMINTEI ECOLOGICE

REZUMAT. Această lucrare prezintă procesul de obținere a unor compozite polimerice biodegradabile pe bază de poliuretan termoplastic post-consum și deșeurii post-consum de cauciuc vulcanizate, în proporții variate (5, 10, 20, 30, 50%), măcinat criogenic la dimensiuni de min. 100 nm și compatibilizator, polietilenă grefată cu anhidridă maleică, între cele două faze. Acest compozit va fi utilizat la realizarea unui produs cu densitate scăzută, cu cost redus, cu recuperarea și reutilizarea deșeurilor, cu conținut de polimeri post-consum, și nu în ultimul rând, biodegradabil. Metodologia de obținere a noilor materiale implică următorii pași: sortarea deșeurilor, măcinarea, funcționalizarea și compundarea. Aceste operațiuni sunt ușor de gestionat și nu presupun echipamente noi. Compundarea, cea mai importantă operațiune, va fi realizată pe un extruder-granulator cu dublu șneac cu posibilitatea de a adapta parametri ca temperatura și viteza în funcție de material, având un raport al șneacului (L/D-35). Compozitele biodegradabile experimentate au fost caracterizate structural și fizico-mecanic. Transformarea deșeurilor (măcinate și funcționalizate) în produse noi cu valoare adăugată va conduce la îmbunătățiri remarcabile ale ciclului de viață a materiilor prime și la utilizarea responsabilă a acestui deșeu, contribuind la sustenabilitate, la îmbunătățirea eco-eficienței și eficienței economice și la reducerea „presiunii” deșeurilor asupra mediului.

CUVINTE CHEIE: biodegradabilitate, compozit polimeric, post-consum, încălțăminte ecologică

COMPOSITE POLYMÉRIQUE BIODÉGRADABLE À BASE DE POLYURÉTHANE RECYCLÉE ET DE DÉCHETS DE CAOUTCHOUC : MATÉRIEL POUR LA FABRICATION DES CHAUSSURES ÉCOLOGIQUES

RÉSUMÉ. Cet article présente le processus d'obtention des composites polymères biodégradables à base de polyuréthane thermoplastique post-consommation et de déchets post-consommation de caoutchouc vulcanisé, dans diverses proportions (5, 10, 20, 30, 50%), broyés cryogéniquement à min. 100 nm et polyéthylène greffé anhydride maléique comme compatibilisant entre les deux phases. Ce composite sera transformé en un produit de faible densité, avec un faible coût, en récupérant et réutilisant des déchets, contenant des polymères post-consommation, et enfin et non des moindres, biodégradable. La méthodologie d'obtention des nouveaux matériaux comprend les étapes suivantes : tri des déchets, broyage, fonctionnalisation et compoundage. Ces opérations sont faciles à gérer et n'impliquent pas de nouveaux équipements. Le compoundage, l'opération la plus importante, sera réalisé sur une extrudeuse bi-vis corotative avec la possibilité d'ajuster des paramètres tels que la température et la vitesse en fonction du matériau, ayant un rapport de vis (L/D-35). Les composites biodégradables expérimentés ont été caractérisés structurellement et physico-mécaniquement. La transformation des déchets (broyés et fonctionnalisés) en nouveaux produits à valeur ajoutée conduira à des améliorations remarquables du cycle de vie des matières premières et à l'utilisation responsable de ces déchets, contribuant à la durabilité, améliorant l'éco-efficacité et l'efficacité économique et réduisant la « pression » des déchets sur l'environnement.

MOTS CLÉS : biodégradabilité, composite polymère, post-consommation, chaussures écologiques

* Correspondence to: Laurenția ALEXANDRESCU, INCDTP - Division: Leather and Footwear Research Institute, 93 Ion Minulescu St., sector 3, Bucharest, Romania, laurentia.alexandrescu@icpi.ro, laura_alexandrescu@yahoo.com

INTRODUCTION

In 2015, the European Commission adopted the new Circular Economy Action Plan to stimulate Europe's transition from a linear towards a circular economy. This ambitious initiative: "Closing the loop - An EU action plan for the Circular Economy" is promoting the transition to a more circular economy, where the value of products, materials and resources is maintained in the economy for as long as possible and the generation of waste is minimized, being an essential contribution to the EU's efforts to develop a sustainable, low carbon, resource efficient and competitive economy. By maintaining the value of the materials and energy used in products in the value chain for the optimal duration and by minimising waste and resource use, the circular economy can promote competitiveness, innovation, a high level of protection for humans and the environment, and bring major economic benefits, thus contributing to growth and job creation. It can also provide consumers with more durable and innovative products that lead to monetary savings and an increased quality of life. Nowadays, the circular economy is an irreversible, global trend. It was foreseen that the proposed actions would contribute to "closing the loop" of product lifecycles through greater recycling and re-use, and bring benefits for both the environment and the economy [1-4].

The paper presents a recycling route for post consumption shoe soles and wood wastes in order to obtain biodegradable composites for green shoe manufacturing. Reusing and recycling of polymer waste are valid options to reduce the amount of waste and, implicitly, the environmental impact, as required by Directive 2008/98/EC. A possible alternative is the transformation of polymer waste with a long lifespan into biodegradable polymeric composites. The worldwide trend toward using cheap, atoxic and durable materials from renewable resources contributes to sustainable development. Thus, the investigation of the potential use of rubber wastes as reinforcing agent in polymeric composites has gained new significance [5]. The global overuse of the synthetic polymer materials has brought some significant issues due to their negative impact on the environment during the past years [6].

Most synthetic polymers made from petroleum and its allied components are not degradable after they are transformed into waste. Today, more and more scientists have shifted their attention to the eco-friendly polymeric materials [7]. Advances in polymer waste management in recent years have made used tires be perceived as a potential source of energy or new raw materials. [8]. Analysis reports of the tire recycling market show that vulcanized rubber powder from tires is mainly used as a filler agent and modifiers in various types of compositions and polymer composites [9-12]. Discarded tires and other rubber waste (containing high quality natural and synthetic rubbers) have become the source of materials for obtaining "environmentally-friendly" composites with useful properties. The benefits of using such materials include the sustainable management of large quantities of used polymer items that are currently burdening the environment and the cost reduction of composite materials produced from cheap polymeric waste [13, 14]. The increase in the level of consumption of rubber recycling products shows that rubber powder is used in the rubber industry as a raw material for obtaining composites with useful practical properties and in an economical way. For high quality and durable rubber products, the use of rubber powder is limited. Therefore, rubber composites obtained by using rubber powder are used in practice for the manufacture of cheap items, where strength is not a priority, such as: flooring materials, windshield wipers, washers, conveyor belts, moulds, cable insulators and shoe soles [15]. Powders obtained from rubber waste can be used to obtain various types of new rubber-polymer composites with elastomeric and thermoplastic matrices. It is known in the literature that the properties of rubber powder depend on the method of obtaining it (cryogenic grinding or at ambient temperature), granule size, crosslinking, filler content, and the type of natural or synthetic rubber that the original products had. The mechanical properties and performance of polymer composites manufactured from rubber powder obtained from waste depend on the type of polymer matrix used and therefore the nature of the interactions between the matrix, the type and size of rubber granules and their quantity in

composites [14, 15]. Thermoplastic polyurethane (TPU) is a preferred choice of polymer to produce engineering products because it has good physical and chemical characteristics, TPU can be processed with extrusion, injection, blow and compression molding equipment. In addition to that, TPU is well suited for a wide variety of fabrication processes [16]. There are previous studies reporting on TPU composites used with several different fibres, for instance TPU reinforced with synthetic fibres such as glass [17] and carbon fibre [18], with other natural fibres [19] such as curaua fibre [20], kenaf fibre [21] and roselle fibres [22]. The literature shows that no study has been carried out on post-consumption thermoplastic polyurethane (WTPU) / post-consumption rubber waste composites. The aim of this paper is to study the properties of WR reinforced WTPU composites with differential rubber waste content (5, 10, 20, 30 and 50 wt%). Tensile strength, tear strength, elasticity, hardness, elongation of break, attrition, melt flow index and morphological study (FT-IR) of WTPU/WR composites were examined.

EXPERIMENTAL

Materials and Methods

Materials

Materials used to obtain the polymer composites based on recycled polyurethane and rubber wastes are as follows: all composites contain post-consumption thermoplastic polyurethane waste (WTPU) compounded with compatibilizing agent, polyethylene grafted with maleic anhydride (PE-G-MA) from Sigma Aldrich and post-consumption butadiene-styrene rubber waste (WR), cryogenically milled at 10,000 rpm for 15 s and sieved through a 100 nm mesh sieve.

Preparation of Various Types of Biodegradable Polymer Composite

WTPU, PE-g-MA and WR were mechanically mixed in Brabender Plasti-Corder PLE 360 at 175°C and 80 rpm for 3 minutes, to melt the plastomer, and for 2 min. at 170°C for homogenisation. The total time was 7 min. Table 1 shows tested formulations from the Brabender mixing diagrams presented in Figure 1.

Table 1: Formulations of TPU waste with butadiene-styrene rubber waste and compatibilizing agent

Components	TPU waste + SBR powder + PE-g-MA					
	TPU 0	TPU 1 (5%)	TPU 2 (10%)	TPU 3 (20%)	TPU 4 (30%)	TPU 5 (50%)
TR waste	300	285	270	240	210	150
SBR Powder	0	15	30	60	90	150
PE-g-MA	0	15	15	15	15	15

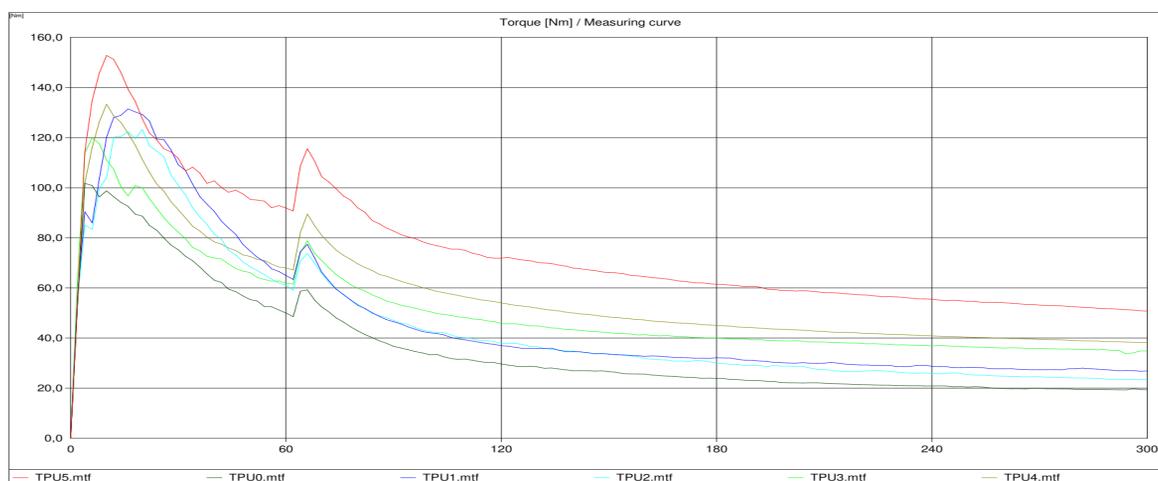


Figure 1. The overlapping Brabender mixing diagrams of control sample (WTPU – TPU0) and TPU-1-5 polymeric composites of TPU waste with butadiene-styrene rubber waste and compatibilizing agent

From the diagrams shown in Figure 1 it is observed that the temperature in the mixing chamber increases, depending on the percentage of rubber waste added to the polymer composite (starting from 175°C, it decreases to 120-144°C then increases as follows: TPU1-183°C, TPU2-185°C, TPU3-187°C, TPU4 -192°C and TPU5-205°C, the latter being the composite with a percentage of 50% elastomeric waste). Also, the mixing forces increase proportionally with the percentage of elastomeric waste in the composite starting at a temperature of 175°C

and reaching a maximum at 30 seconds of mixing of 120 Nm for the TPU1 sample and increasing proportionally up to 157 Nm for the TPU5 sample. The maximum force is reached when the WTPU plasticizes and the dispersion of WR particles begins. After reaching the maximum force, it decreases evenly, the last minute being constant, which indicates the homogenization of the mixture. Table 2 shows the processing characteristics presented in the Brabender diagrams, for each processed composite.

Table 2: The characteristics, presented in the Brabender processing diagrams, for WTPU/PE-g-MA/WR polymer composites

Compound/ Characteristics	TPU1 (5%WR)	TPU2 (10%WR)	TPU3 (20%WR)	TPU4 (30%WR)	TPU5 (50%WR)
Loading peak temperature, °C	123	127	134	138	144
Inflection point temperature, °C	178	183	157	159	165
Maximum temperature, °C	178	183	160	162	168
Loading peak energy, Nm	67.2	77.0	18.0	18.1	22.4
Maximum energy, kNm	36,2	19.8	102,7	116.6	156.8
Gelation area energy, J	1,3	0.0	0.8	1.1	1.2
Specific energy (W/Sample mass), kJ	0.3	0.3.5	0.4	0.4	0.6
Gelation speed, Nm/min.	8.1	7.1	372,5	447.6	537.6

The compounds were then compression-molded (using an electrically heated laboratory press) to obtain a sheet of about 2 mm thick. Press parameters: preheating 3 min.; pressing 4 min.; cooling 13 min.; pressure 300 kN; temperature 170°C. The sheet was then cooled down to room temperature under the same pressure. The specimens were die-cut from the compression molded sheet and used for testing after 24 hours of storage at room temperature.

Testing Methods

Tensile tests of the samples were carried out according to SR ISO 37:2012 using a Schopper Tensile Testing machine 1445, at a constant crosshead speed of 500 ± 5 mm/min.

Hardness of the samples was measured by Shore "A" Durometer according to SR ISO 7619-1:2011.

Abrasion resistance was carried out according to ISO 4649/2010, the cylinder method, using a pressure of 10 N. Abrasion resistance was expressed by relative volume loss in relation to calibrated abrasive paper. A wearing tester with abrasive cloth having granulation of 212–80 mm

(PE 80). The samples used were obtained from rolled blends and pressed into sheets, then cutting with a rotating die and have cylindrical shape, with a diameter of 16 mm and height of min. 6 mm.

Repeated flexions - Ross Flex - SR ISO 132/2018 is the test that determines the resistance of the specimens to the appearance and propagation of cracks when they are subjected to repeated flexions at an angle of 90°, on a mandrel with a diameter of 10 mm, up to 30,000 cycles or until the crack appears or the material breaks.

FT-IR spectroscopy was done using the FT-IR 4200 JASCO, Herschel series instrument, equipped with ATR having diamond crystal and sapphire head within the spectrometric range 2000-530 cm^{-1} .

RESULTS AND DISCUSSIONS

Physical-mechanical tests were carried out in the Investigation laboratory from INCDTP - Division ICPI, accredited by RENAR, and materialized in the determination of hardness,

elasticity, tensile and tear strength, attrition, residual elongation and elongation at break for thermo-oxidative aging (168h x 100°C) and normal state, and melt flow index (see Tables 3 and 4).

Table 3: Physical-mechanical test results of formulations: TPU waste with butadiene-styrene rubber waste and compatibilizing agent

Property / test method	TPU0	TPU1	TPU2	TPU3	TPU4	TPU5
Hardness, °Sh A, SR ISO 7619-1:2011	79	80	81	81	81	78
Elasticity, %, ISO 4662:2009	24	24	24	24	24	20
Modulus, 100 %, ISO 4662:2009	3.53	3.53	3.57	3.43	3.31	2.84
Modulus, 300 %, ISO 4662:2009	6.23	5.29	-	-	-	-
Tensile strength, N/mm ² , SR ISO 37:2012	7.9	5.64	4.82	4.29	3.8	3.52
Elongation at break, %, SR ISO 37:2012	480	340	280	220	200	180
Residual elongation, %, SR ISO 37:2012	96	56	34	24	20	18
Tear strength, N/mm, SR EN 12771:2003	49.8	52.6	44.67	41	32.22	25.84
Density, g/cm ³ , SR ISO 2781:2010	1.18	1.17	1.17	1.17	1.16	1.16
Abrasion, mm ³ , SR ISO 4649/2010	90.42	100.68	153.3	177.04	189.06	200.25
Ross Flex - repeated flexions SR ISO 132/2018	Withstands up to 150,000 cycles			90000 cycles-cracks appear, withstands cracks up to 109.000 cycles	3600 cycles-cracks appear, withstands cracks up to 25300 cycles	

Table 4: Physical-mechanical test results of formulations: TPU waste with butadiene-styrene rubber waste and compatibilizing agent, after accelerated ageing 70°C x 168 h

Sample/Characteristic	TPU0	TPU1	TPU2	TPU3	TPU4	TPU5
Hardness, °Sh A, SR ISO 7619-1:2011	77	78	79	80	81	79
Elasticity %, ISO 4662:2009	30	26	26	24	24	22
Modulus, 100 % ISO 4662:2009	3.41	-	-	-	-	3.64
Modulus, 300 % ISO 4662:2009	5.81	3.61	3.52	3.57	3.45	-
Tensile strength, N/mm ² , SR ISO 37:2012	6.65	5.05	4.28	3.57	3.62	3.64
Elongation at break, %, SR ISO 37:2012	460	300	280	180	160	140
Residual elongation, %, SR ISO 37:2012	60	34	28	20	20	20
Tear strength, N/mm, SR EN 12771:2003	58.5	50.5	45	38	34	25.5

Hardness

In the initial state of the control sample, TPU1, hardness value is 79-80°Sh A and increases to 81°Sh A for TPU3 and TPU4 samples, proportional to the rubber waste content. The increase of 2°Sh A is due to the loss of plasticizer in the compounding process at temperatures of 175°C. Regarding the TPU5 sample, with 50% elastomeric waste, the decrease to 78°Sh A occurs due to the large amount of waste, which initially had a hardness below 70°Sh A. After accelerated aging, in the control sample and the first three samples (5-20% elastomeric waste content) the hardness decreases by 1°Sh A due to the plasticization of the WTPU matrix and in those with high content of elastomeric waste, increases by 1°Sh A, due to losses of plasticizer

from the elastomeric waste dispersed in the plastic mass.

Tensile Strength

The tensile strength falls in the range from 5.64-3.52 N/mm², for normal state and 5.05-3.64 N/mm² after aging. The values are comparable to those of thermoplastic polyurethane waste (TPU0) -7,9 N/mm² and decreases proportionally with the WR content introduced into the composite. Compared to the normal state, accelerated aging modifies the tensile strength values only by 1-2%.

Elasticity

The elasticity values are the same with the control sample (TPU0), respectively 24%

in all samples, except the composite with 50% elastomeric waste, for which the value is four units lower than the others, due to the high percentage of waste dispersed in the mass of WTPU. The values fall within the specific standards. At the accelerated aging, the values are lower than characteristic of the control sample - 30%, but higher than the characteristic values of the composites tested in normal state and non-uniform.

Tear Strength

Tear strength is an index with performance values, maintaining the same values (52.6-25.84 N/mm) due to tested elastic-plastic composites morphology (Table 3) and decreases proportionally with the amount of composite rubber waste compared to the control sample value of 49.8 N/mm. In the case of accelerated aging, the value of the control sample increases by approximately 20% (from 49.8 N/mm to 58.5 N/mm) compared to the values obtained for the composites that have an increase of 1-2 units, the composite with the higher proportion of rubber waste being practically equal. This is due to the rubber waste which is already aged and no longer influences the test values.

Abrasion Resistance

The values of abrasion resistance increase slightly from 90.42 mm³ for the control sample up to 100.68 mm³ for the sample with the 5% rubber wastes and increases with the amount of rubber waste up to 200.25 mm³ for the TPU5 sample with 50% rubber waste. The values fall within the requirements imposed by the standards.

Density

Density values do not change when the rubber waste particles are added to the compound.

Repeated Flexions – Ross Flex

The current footwear standards specify for thermoplastic polyurethane footwear values of 30,000 cycles when determining repeated flexions. The values presented in Table 3 show that only the TPU5 composite with a rubber waste content of 50% does not fall into this value

(25,300 cycles). The others have values five and three times higher, respectively, than the values imposed by the standard.

FT-IT Spectroscopy

IR spectrum represents the radiant energy absorption curve in the IR domain by the sample molecule, depending on the wave length or radiation frequency. The infrared domain of the electromagnetic radiation is between 0.8 and 200 μm. IR domain for usual organic chemistry is between 2.5 and 25 μm. The structural determinations were carried out on an IR molecular absorption spectrometer with double beam, in the range of 4000-600 cm⁻¹, using 4200 FT-IR equipped with ATR diamond crystal and sapphire head. The solid-state samples were set in the ATR and the equipment recorded the transmittance spectra of the sample and then compared it with the background spectra previously recorded. The recorded spectra of the samples were compared with the polyurethane waste and elastomeric waste spectrum. The FTIR spectra of the analyzed materials are presented in Fig. 2.

After the tests were carried out, the following were found:

- These figures clearly show the interaction between urethane groups and WF. The FTIR analysis confirmed the presence of functional groups characteristic for the TPU. The urethane moieties of WTPUs are confirmed by the presence of the main characteristic absorption bands. They are well represented by the characteristic $\nu(\text{C}=\text{O})$ vibration region (1725 cm⁻¹), $\nu(\text{N}-\text{H})$ stretching vibration region (3326 cm⁻¹) [15] and $\delta(\text{N}-\text{H})$ bending vibration absorption peak at 1529 cm⁻¹ [16]. It should be pointing out that with increasing content of WR in the reaction mixture the carbonyl peak at 1725 cm⁻¹ decreases (Figure 2).
- The decrease of the absorption band from 1725 cm⁻¹ specific to WTPU is associated with the decrease of the amount of WTPU from the tested composites and the increase of the elastomeric waste content. As the WR content in the polymer mixture increases, the WR-specific signal strength increases.

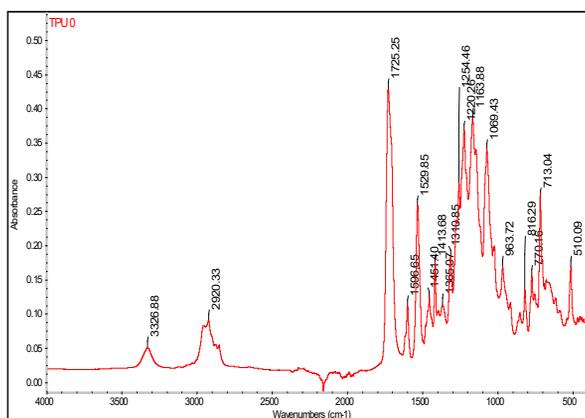


Figure 2. FTIR spectra for WTPU

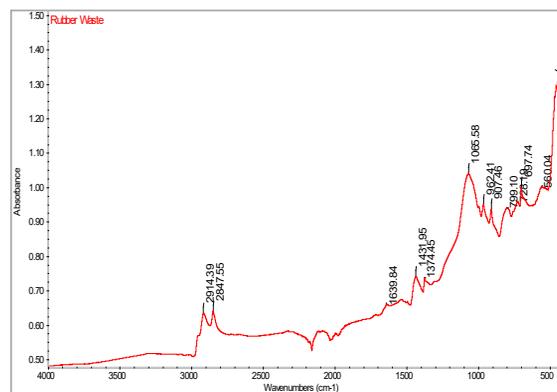


Figure 3. FTIR spectra for WR

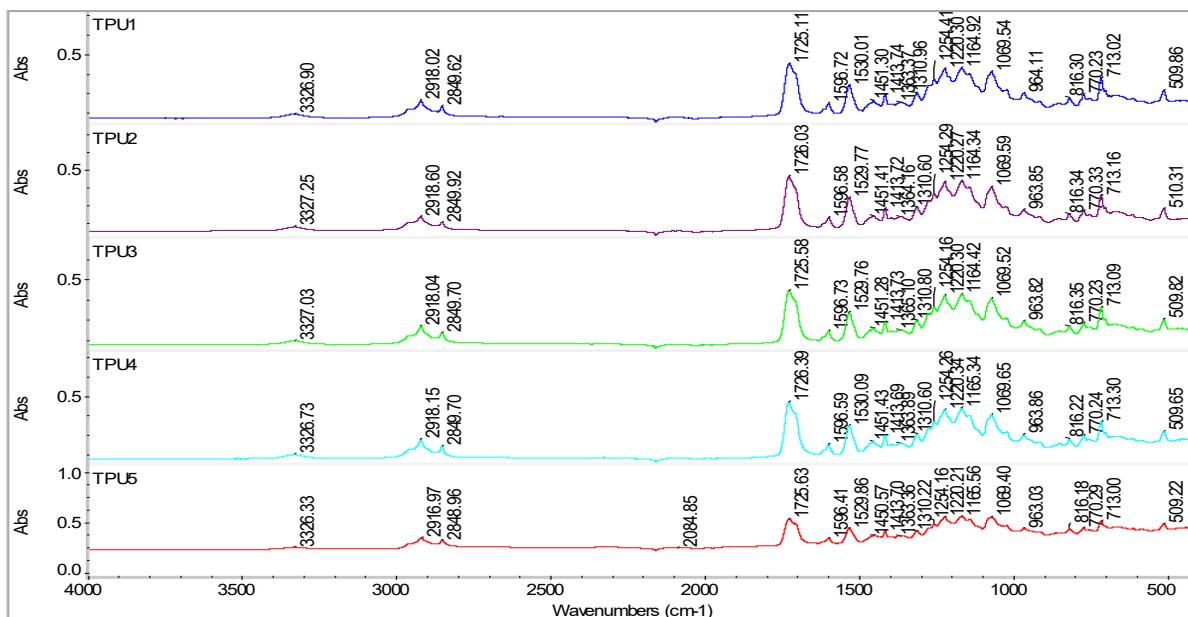


Figure 4. FTIR spectra for WTPU/PE-g-MA/WR polymer composites

- The presence of specific PE-g-MA groups does not appear in the spectra due to the small amount of material.

CONCLUSIONS

Post-consumption thermoplastic polyurethane (WTPU) waste is compounded with cryogenically ground post-consumption rubber waste (WR). These polymeric composites were processed in a twin-screw extruder-granulator using elastomeric waste of different proportions (5, 10, 20, 30, 50%) and compatibilizer based on maleic anhydride grafted polyethylene (PE-g-MA). Polymer composite samples with

percentages of 5, 10, 20% post-consumer elastomeric waste showed the best physical and mechanical performance compared to those with higher percentages of elastomeric waste (30, 50%), characteristics that fall in the specific values of standardized thermoplastic polyurethane. Properties such as elongation at break and elasticity showed a slight reduction compared to WTPU, and the hardness increased by 1-2 units. The tensile strength of the composition decreased by half compared to WTPU. Thus, the WTPU / PE-G-MA / WR composite has a great potential for use in the footwear industry. Thus, the use of thermoplastic polyurethane and post-consumer rubber waste in polymer composites

could contribute to sustainable development in the near future.

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