



Development of thermoplastic composites based on recycled polypropylene and waste printed circuit boards



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ABSTRACT

In the last several years, the electronic waste, especially printed circuit boards have significantly increased over the world, generating one of the highest rates of solid waste. The recycling process of the printed circuit boards implies mainly the recovery of metals and glass fibers, while the reuse of the polymeric support has remained largely in the phase of research. In this paper, the non-metallic part of printed circuit boards was used as filler (up to 30%), but also to improve the fire resistance of thermoplastic composites based on recycled polypropylene and diene block-copolymers. The synergy between the elastic effect of elastomers and the reinforcing effect of the waste powder into the thermoplastic matrix was studied by mechanical and dynamo-mechanical analysis, X-ray diffraction, optical microscopy, micro-calorimetry and thermo-gravimetric analysis. Improved mechanical properties, especially impact strength was observed. The compatibilization of components considering the interactions between the ethylene-butylene blocks from the hydrogenated and maleinized styrene-butadiene block-copolymer and recycled polypropylene, respectively between the MA groups and the functionalities of the waste powder, evidenced by FTIR, was highlighted by changes in the X-ray pattern and an increased fire resistance and thermal stability.

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1. Introduction

In the last decades, the electronic waste stream has increased substantially, in consequence generating one of the highest rates of solid waste. The accelerated development of the economy, the life time of the electronics, or the diversified supply of these products lead to the increase of the quantities of electronic waste (Bazargan et al., 2014; Li et al., 2012; Sahajwalla et al., 2015; Suresh et al., 2018). Also, the obsolescence of electronic equipment and the growing demand of consumers for an advanced equipment lead to significant quantities of out-of-date electronics. With an overall recycling rate of only 10–18%, most of the outdated electronic equipment is deposited in landfills, incinerated or relocated to the economic circuit of developing countries. The overall envi-

ronmental impact of electronic waste can be reduced by proper initiatives regarding the energy consumption, the reduction of disposed solid and wastewater by e-waste recycling etc. (Fathi and Hanifi, 2007; Hong et al., 2015; Ning et al., 2017; Sohaili et al., 2012; Vermeşan et al., 2020).

Waste printed circuit board (WPCB) recycling is an increasingly debated issue in the last two decades at government level, worldwide, as they have a particularly harmful polluting effect (heavy metals, bromide compounds used as fire retardants, non-biodegradability). The increasing share of about 10% of WPCB in waste electrical and electronic equipment (WEEE) exacerbates the problem of their recycling (Korf et al., 2019; Li et al., 2012). The economic profitability is the decisive factor in recycling WPCB. Consequently, the recycling procedures applied on industrial scale refer to the recovery of metals and glass fibers. The reuse of the polymeric support – a thermosetting resin (usually epoxy) that is impossible to be used in a conventional melt technology of recy-

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cling, has remained largely in the phase of research (Anshu and Hait, 2018; Cui and Zhang, 2008; Gu et al., 2019; Hadi et al., 2015a, 2015b; Wang and Xu, 2014), the waste being a value-added material for sustainable rehabilitation. Hence, several proposed applications of this fraction have been comprehensively examined (Luda, 2011). The high content (of about 30–70%) of glass fibers contained by WPCB (Muniyandi et al., 2013a, 2014) has a great reinforcing effect on the thermoplastic composites due to their excellent mechanical characteristics like L/D ratio, elastic modulus etc. Beneficial effects of WPCB on the polyolefin matrix were remarked. Filling high density polyethylene (HDPE) or polypropylene (PP) with rigid particles like the fine-sized WPCB is a convenient, effective and economic method to enhance their strength and rigidity (Biswal et al., 2015; Muniyandi et al., 2013a, 2014; Yang et al., 2016b; Zheng et al., 2009). In case of PP, the most important mechanical property that must be improved is its impact strength. This property, for recycled polypropylene (rPP), becomes even more deficient due to the degradation of the products during their exploitation. WPCB powder was used as reinforcing agent for rPP with or without maleic anhydride grafted polypropylene (MAPP) and an increase of 12% of the polyolefin impact strength was obtained (Biswal et al., 2015). By adding styrene-diene block-copolymers, the increase of the impact strength of rPP can reach to 90% (Ghioca et al., 2017).

Besides the environmental benefits of both rPP and WPCB recycling, economic advantages can be obtained mainly considering the costs of: landfill or other method of disposal; inorganic fillers that have the same reinforcing effect on the polyolefin matrix, production process of virgin polypropylene. Thus, increasing the market demand for obtaining both higher performance and cheaper materials can lead to an increase of WPCB, a byproduct of printed circuit boards recycling industry, that can be used in nonmetallic/PP composites (Zheng et al., 2009).

In this study, the non-metallic part of printed circuit boards was used both as filler and fire resistance modifier in composites based on recycled polypropylene and styrene-butadiene block-copolymers. The synergic effect of the elastomers and the waste powder added to the thermoplastic matrix on mechanical and dynamic-mechanical properties, thermal stability and flammability of composites was studied. The compatibility between the waste powder and polypropylene was improved using a functionalized block-copolymer, containing maleic groups. Also, morphological and elemental characterization of rPP-elastomer-WPCB compounds was performed. There is no information, to the best of our knowledge, on the effect of these components synergy and on the micro-calorimetry tests of rPP/elastomers/WPCB composites.

As compared to previous researches, the paper aims a simultaneous reuse of printed circuit boards and polypropylene wastes with various benefits, like economic (low costs of obtaining, WPCB powder with dimensions below 1 mm whose less advanced grinding is achieved with low energy consumption), environmental (higher quantities of waste), and respectively technical advantages (polypropylene composites with high-performance impact strengths, comparable to those of existing assortments on the market based on virgin polyolefin).

2. Experimental part

2.1. Materials

As matrix, recycled polypropylene (rPP) as grind material (of about 0.05x0.6 mm) from industrial transport shuttles was supplied by a local recycling company (Romcarbon SA) and used as received. The main characteristics of homogeneous rPP were: den-

sity 0.96–0.99 g/cm³, melt flow index (at 190 °C, 5 kg) 6 g/10 min; the tensile strength of 20.6 MPa, elongation at break of 2.83% and Izod impact strength (at 23 °C) of 10 kJ/m² were measured after melt processing the thermoplastic waste on the Brabender plastometer.

To improve the impact strength of polypropylene, a radial styrene-butadiene block-copolymer (SBS, Europrene SOL T 161 C, 30% bounded polystyrene, molecular mass 198,000 g/mole, melt flow index at 190 °C, 5 kg ≤ 1 and density 0.94 g/cm³) was used. A hydrogenated and maleinized styrene-butadiene block-copolymer (SEBS-MA, Kraton FG 1901X, 1.4% chemically bound maleic anhydride, 30% polystyrene content, molecular mass 58000 g/mole, melt flow index at 230 °C, 5 kg of 17.2 and density 0.91 g/cm³) was added during the melt processing of rPP as compatibilizer.

The non-metallic powder with dimensions below 1 mm was obtained by leaching the WPCB using a simple mild oxidation system. Copper was removed from the grounded boards using sulfuric acid, copper sulphate and sodium chloride. After desolvation, a light blue-green powder was obtained. The method was described in detail by (Cocchiara et al., 2019).

A sterically hindered phenolic primary antioxidant compatible with polypropylene – Irganox 1076 (octadecyl-3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate) - Basf was used against thermo-oxidative degradation (1% of total mass).

2.2. The composites obtaining

The composites were obtained by melt compounding the components on a Brabender plastometer equipped with a 30 cm³ tank, at a temperature of 185 °C and a mixing speed of 60 rpm for 7 min, Fig. 1. The obtained materials were pressed using a Platen Press P 200 E (Dr. Collin GmbH, Germany) at 190 °C with 200 MPa pressure for 15 min. After cooling the material for 20 min under pressure to room temperature, plates of 1 mm and 4 mm thicknesses were obtained from each mixture, for tensile test, respectively the impact strength determinations.

The amount of WPCB powder added to the polymeric matrix (rPP/rPP + elastomers) ranged from 5 to 30 wt%. A higher filler quantity to the recycled polypropylene damaged the processability of the composites, and the materials could not be obtained by this technique. The blends composition is presented in Table 1.

Chemical reactions between WPCB, epoxy resin, rPP and elastomers, especially SEBS-MA by their functionalities were not excluded, but the paper mainly focused on the compatibility between components with similar structure, and strengthening the interfacial adhesion by physical interactions.

2.3. Methods

Fourier-Transform Infrared Spectroscopy (FTIR) was used to determine the functionality on the non-metallic waste powder with a Bruker Optik GMBH spectrometer, equipped with DRIFT accessory and ATR mode, in the wavelength range of 4000–400 cm⁻¹, 32 scans, 4 cm⁻¹ resolution.

The IZOD impact strength was measured on notched samples using a Zwick-Roell HIT 5.5P Impact Tester Pendulum (Zwick-Roell, Germany) according to ISO 180. A FPZ 100 Testing Machine was used to determine the elongation at break and tensile stress of the composites at a testing speed of 50 mm/min according to ISO 527. Ten replications were achieved for every tensile and impact test.

The dynamic mechanical properties (DMA) were recorded using a Q800 dynamic mechanical analyzer from TA Instruments, (V20.24, New Castle, DE, USA), in air atmosphere, with 1 Hz frequency, from –105 °C to 155 °C with a speed of 5 °C/min and

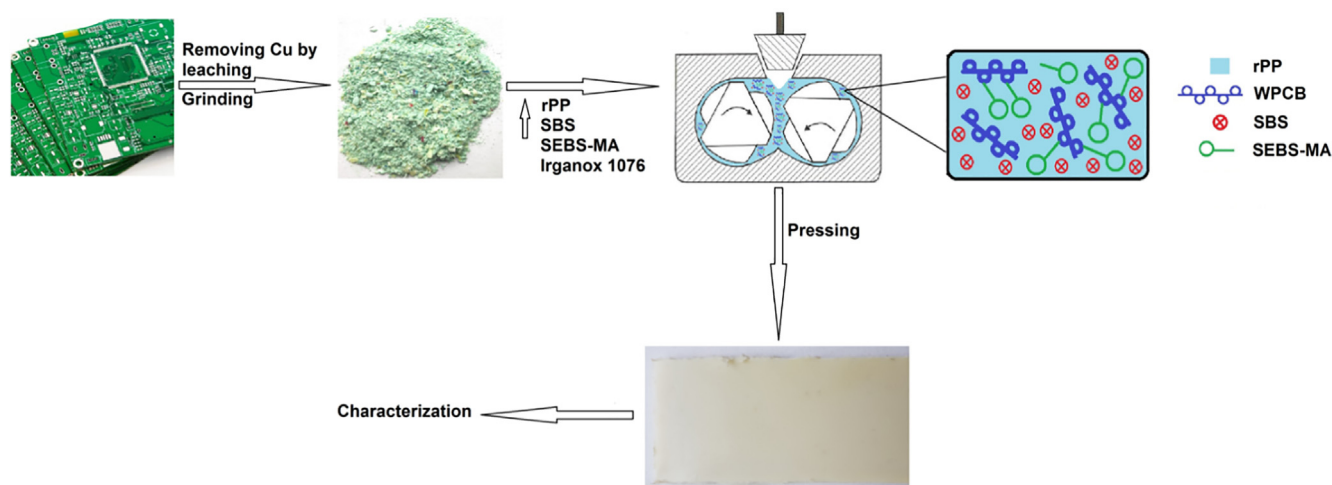


Fig. 1. Schematic representation of the process.

Table 1

The proportion between the components of the mixtures.

Sample	Composition, %				
	rPP	WPCB	SBS	SEBS-MA	Irganox 1076
rPP + WPCB	99–69	0–30	–	–	1
rPP + SBS + WPCB	84–54	0–30	15	–	1
rPP + SBS + SEBS-MA + WPCB	79–49	0–30	15	5	1

20 μm amplitude. Storage modulus and loss modulus dependence on temperature resulted.

The optical microscopy (OM) was performed with a Primo Star ZEISS optical microscope that offers the possibility to investigate the samples in transmitted light at a magnification between 4X and 100X. The equipment had attached a digital video camera (Axiocam 105) which, by the microscope software, allowed real-time data acquisition. The obtained images were achieved with a 40X magnitude.

The diffraction data (XRD) were recorded with an X-ray diffractometer Rigaku Ultima IV (Rigaku, Tokyo, Japan) with high-resolution multimodal and multifunctional system, vertical goniometer θ/θ (285 mm radius) in geometry G/9, X-ray tube – Cu anode (2 kW) and Bragg–Brentano, using $\text{Co K}\alpha$ radiation with $\lambda = 1.54 \text{ \AA}$. The profiles were recorded from 3 to 90°, with 1°/min. The d -spacing (d) for rPP composites was calculated from the Bragg equation (1) and their crystallinity (I_c) based on equation (2):

$$d(\text{\AA}) = \lambda / (2 \cdot \sin\theta) \quad (1)$$

$$I_c(\%) = \frac{A_c}{A_c + A_a} * 100 \quad (2)$$

where: θ is the diffraction angle ($^\circ$), λ is the wavelength of the incident wave (\AA) and A_c and A_a are the areas of crystalline and amorphous peaks.

The wavelength dispersive X-Ray Fluorescence Spectrometry (WDXRF) was used for a qualitative and quantitative elementary composition (elements ranging from 80 to 92U). Detection limit: 1 ppm–10 ppb; Accuracy less than 0.1–0.5%.

The thermal stability of the obtained materials was analyzed by thermogravimetric analysis (TGA) using a Q5000IR instrument (TA Instruments, New Castle, DE, USA) using 100 μL platinum pans. The samples were heated with 10 $^\circ\text{C}/\text{min}$ from room temperature to

700 $^\circ\text{C}$, the temperature was maintained constant for 5 min under air, and then under nitrogen atmosphere.

To investigate the fire performance of the materials based on rPP and WPCB, micro-scale combustion calorimetry (MCC) was performed with a FITT Micro-calorimeter using the following parameters: heating rate 1 $^\circ\text{C}/\text{min}$., the combustor temperature 900 $^\circ\text{C}$, N_2 flow rate 80 cm^3/min ., O_2 flow rate 20 cm^3/min ., according to method A (ASTM D7309-13). In the procedure of method A, the sample is subjected to controlled thermal decomposition (Wilkie et al., 2006) and then heated in an oxygen/anaerobic environment. The gases released from the sample during the test are cleaned from the test chamber with nitrogen, then mixed with the excess oxygen and completely oxidized in a high temperature combustion furnace. The volumetric flow rate and the volumetric oxygen concentration of the gas stream exiting the combustion furnace are continuously measured during the test to calculate the rate of heat release using oxygen consumption. Method A measures the combustion heat of the volatile component of the sample (sample gases), but not the heat of combustion of solid residues.

3. Results and discussion

The composition of the non-metallic WPCB powder is a complex one. According to literature (Muniyandi et al., 2013a, 2013c), the non-metallic part of WPCB consists mainly of epoxy resin, fiber glass and brominated flame retardants, which makes it quite difficult to reprocess them. The functionality of the filler can be established using FTIR, Fig. 2a. In the wavelength range between 3300 and 3500 cm^{-1} , the specific peaks for O-H groups or Si-OH on the glass fibers surface can be observed for WPCB and also for rPP composites, proportionally with WPCB dosage, Fig. 2b. The peak at 1729 cm^{-1} corresponds to C = O group (Yao et al., 2020). The bands from 1499 and 1597 cm^{-1} can be attributed to the stretching vibration of the benzene skeleton. The peak at 828 cm^{-1} can be attributed to the bending vibration of the substi-

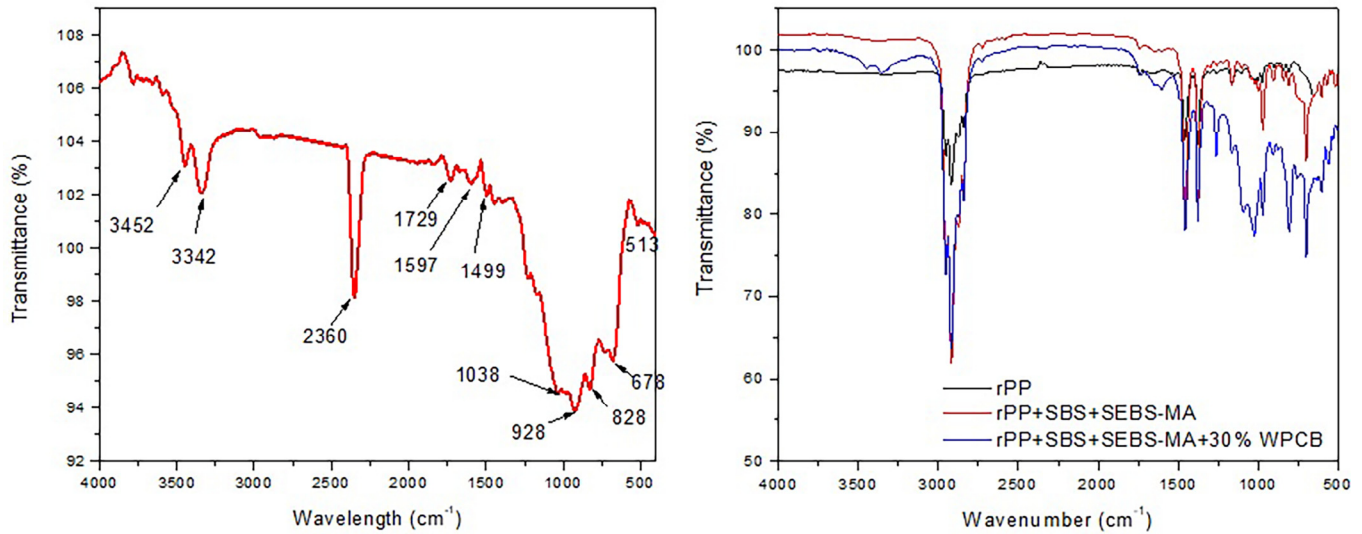


Fig. 2. FTIR spectrum of WPCB (a) and rPP composites (b).

tuted benzene skeleton. The characteristic band at 1038 cm⁻¹ indicates the presence of C-O-C groups. The characteristic C-Br bands that appear at 678 cm⁻¹ and 513 cm⁻¹ can highlight the presence of brominated flame retardants in the circuit board waste powder. The identified polar groups from WPCB can establish physical interactions with the polymers from the system. Peaks with higher intensities appear at shifted wavenumbers between 1700 and 700 cm⁻¹. The compatibility between rPP and the ethylene-butylene block from SEBS-MA is reflected by more intense bands at 3000–2800 cm⁻¹, 1466 and 1372 cm⁻¹ respectively (Fig. 2b).

The research was focused on using WPCB as filler in rPP modified with elastomers in order to improve its characteristics, especially the polyolefin impact strength and thermal stability. The brominated components, usually contained by WPCB, can impart fire retardancy to rPP composites.

The mechanical properties of a polymer composite containing powder fillers depend on the efficiency of the mechanical stress transfer from the matrix to the filler. The strength of a composite material depends on the synergy of the weakening effect due to a high mechanical effort and on the reinforcing effect when the filler creates a barrier against the cracks propagation on load stress; more micro-cracks are formed instead of deeper cracks. Moreover, the strength also depends on the interfacial adhesion between the components and the percentage of filler added (Móczó and Pukánszky, 2008).

The relationship between the impact strength values and the composition of polypropylene composites is shown in Fig. 3. The impact strength of rPP composites filled with the rigid powder decreases from 10 kJ/m² to 6.25 kJ/m² with the addition of WPCB until 15% of filler. The impact strength has an attenuated decrease at higher dosages, adding 30% WPCB results in an impact strength of 3.77 kJ/m². Combining the elasticity effect produced by SBS and SEBS-MA and the reinforcing effect of the WPCB powder, a more pronounced decrease of the impact strength is obtained until a dosage of 15%, or 20% for rPP + SBS, respectively for rPP + SBS + SEBS-MA, and then a tendency to limit the values can be observed.

The addition of high amount of WPCB involves adding many glass fibers into the rPP composites so the filler can agglomerate and create regions susceptible of stress concentration and a reduced dispersion of ingredients with poor interfacial adhesion (Guo et al., 2010; Muniyandi et al., 2013b). By adding elastomers, the resistance to crack propagation is improved when the material

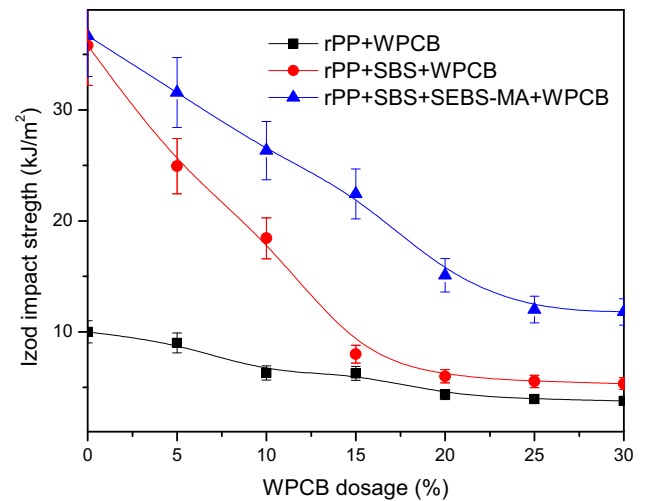


Fig. 3. The impact strength recorded for rPP composites depending on the WPCB dosage.

is subjected to mechanical stress, by a better transfer of the destructive force. When the content of filler increases up to 30%, the impact strength value decreases with 62% for rPP-WPCB composite compared to rPP, 85% by adding SBS and 68% when using also SEBS-MA related to the control samples (without filler). As can be seen from Fig. 3, a three times higher impact strength compared to filled rPP is obtained by the polyolefin compatibilization with both the elastomers, the maleic groups being able to physical bind with the WPCB functionality, and thus enhance the interfacial adhesion between the waste powder and rPP matrix. The impact strength improvement can be attributed to a better interfacial area between the components, the compatibilizer being adherent to the filler surface and thus an attenuation of the cracks dimensions appeared when the material is subjected to mechanical stress, by a better transfer of the destructive force (Arencon and Velasco, 2009; Zheng et al., 2008). These results indicate that combining the WPCB rigid filler with elastic modifiers results in a synergistic reinforcing effect of the recycled polypropylene matrix by obtaining higher values of the impact strength of rPP composites.

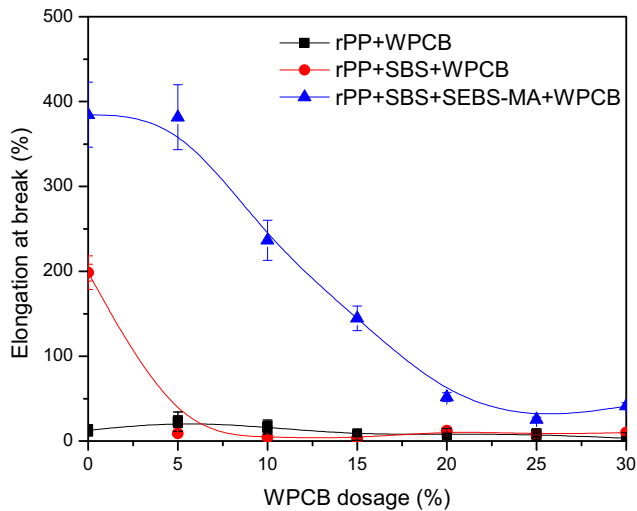


Fig. 4. Variation of elongation at break of composites with WPCB dosage.

The elongation at break recorded for the polypropylene composites is illustrated in Fig. 4. The effect of the thermoplastic elastomers with high elasticity on the rPP matrix is reflected in a significant increase in the elongation at break of the materials, the composites, initially brittle, becoming ductile. By increasing the dosage of rigid filler, the reinforcing effect produced by the rigid filler, especially due to the contained glass fibers, is more significant than that of elasticity, which leads to a decrease of elongation at break. At WPCB dosages of 20–30%, close values of elongation at break are observed, probably because of a low compatibility of the high quantity of WPCB with rPP. Higher elongation of break can be obtained by compatibilization with SEBS-MA. The elongation at break of the composites compatibilized with SEBS-MA increases by 30, 17, 14 and 19 times compared to rPP at 0, 5, 10 and 15% WPCB, respectively.

The higher elongation at break and improved IZOD impact strength of recycled polypropylene compounded with elastomers, with or without fillers, were also reported in literature (Biswal et al., 2015; Răpă et al., 2013; Răpă et al., 2020). Adding SBS, a significant increase in impact strength and elongation at break can be obtained. Also, SEBS-MA, through its polar groups, improves the compatibility between the WPCB powder and rPP with beneficial

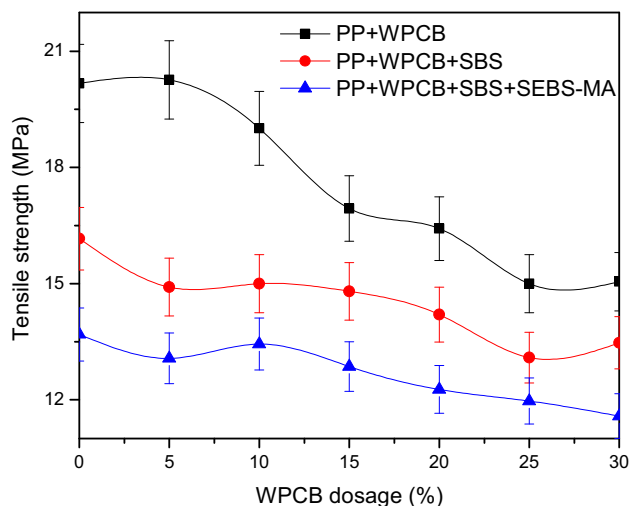


Fig. 5. Variation of tensile strength of composites with WPCB dosage.

effect on impact resistance, even at higher dosages of WPCB (20–30%).

For the obtained rPP composites, a decrease of tensile strength both with WPCB dosage and the addition of elastomers can be observed, Fig. 5. Dispersion of WPCB in the rPP matrix has a disruptive effect on the polypropylene crystal lattice, especially at higher dosages. The high amount of elastomers dilutes also the crystalline phase of rPP with decreasing the tensile strength. SBS is distributed into the polyolefin as domains smaller than SEBS-MA and thus the strength values are less low. In addition, the decrease in the tensile strength of composites may be due to a high molecular weight polydispersity of recovered polypropylene which may have a certain degree of degradation that usually leads to less resistant materials.

Fig. 6 presents the temperature dependence of storage (E') and loss modulus (E'') of rPP in the presence of SBS and SEBS-MA, and the waste powder. For economic and environmental reasons, the maximum WPCB load was considered. The results indicate improved storage modulus for the composites with 30% WPCB as compared to the matrix without filler. Thus, for E' measured at 30 °C, an increase with 35% was obtained for rPP + 30% WPCB composite and 42% for the sample that contains also SBS, in comparison with the control samples, due to the reinforcing effect of the high amount of glass fiber contained in the waste powder and SBS uniform distribution into the polyolefin. In case of the filled composite containing SEBS-MA, the synergic effect of elastomers and WPCB is evidenced by an increase of the material stiffness of only 12%. This value is influenced by the fact that the block-copolymer is in a rubbery phase having a low transition temperature and considering that interactions can be established between the components.

Similar observations can be sustained also from the loss modulus dependence on temperature. By adding the elastomers, the viscosity of samples, correlated with E'' , decreases and increases with the rigid filler, an expected effect of reinforcing. A shift of the maximum peak can be observed and can be attributed to an immobilization of the polymer chains at the interphase with WPCB powder. The more intense peaks recorded for rPP composites containing the waste can be due to a decrease in the polymers mobility.

Fig. 7 a-d shows the optical microscopy (OM) images of WPCB and the fracture surfaces of rPP composites. The high amount of glass fibers distributed into the epoxy matrix is better evidenced in the waste morphology (a). Single glass fibers of 0.5–1.5 mm detached from the matrix, but also small agglomerates of fibers can be observed, especially for the rPP-WPCB sample (b). The filler is relatively homogeneous dispersed into rPP, but micro-cracks are present. Many voids between the filler and matrix appear in the fractured surface due to the poor compatibility and weak adhesion of WPCB to the polyolefin.

By adding SBS, an improved aspect of the samples can be observed, but also the filler tendency to agglomerate (c). The improved compatibility obtained with SEBS-MA and better affinity of WPCB and the MA groups is revealed by less voids in the fractures surface of the composite and higher homogeneity (d). The better adhesion by physical interactions between the filler and the polymeric matrix is beneficial for the stress transfer and sustain the improvement in mechanical properties of compatibilized materials.

In order to study the effect of the compatibilizers and WPCB on the recycled polypropylene matrix, XRD diffraction pattern of rPP, and rPP-elastomers that contain 30% filler, respectively the pattern for WPCB and pure block-copolymers were recorded and presented in Fig. 8 a-c. Important peaks appear at 14, 16.8, 18.5, 21.6° being attributed to the (1 1 0), (0 4 0), (1 3 0), and (1 1 1) crystal diffraction peaks known for polypropylene (De Rosa and Auriemma, 2013; Lu et al., 2017; Spoerk et al., 2018). Similar results for all the samples

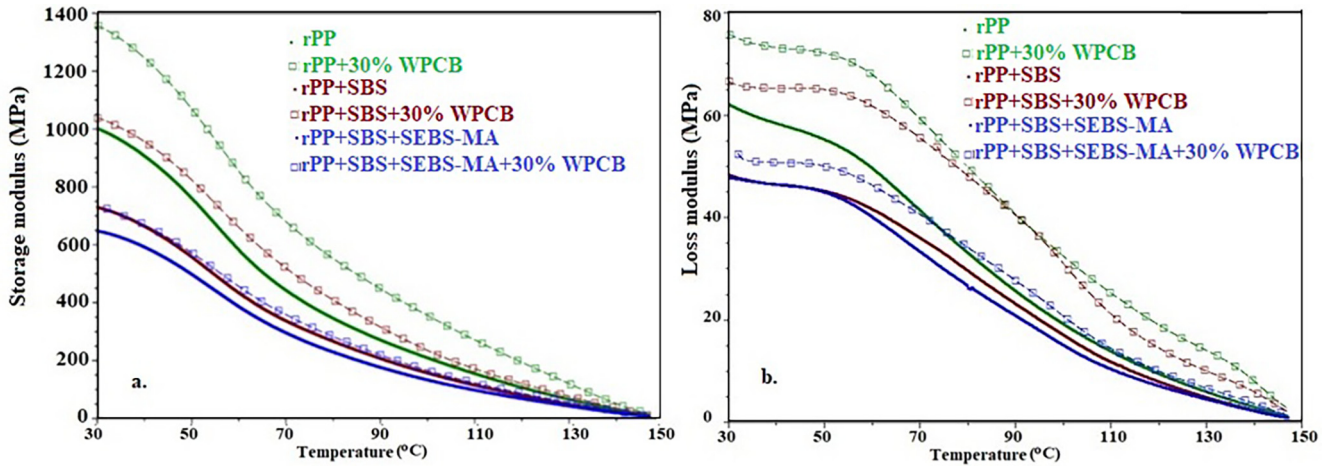


Fig. 6. Storage Modulus (a) and Loss Modulus (b) recorded for the rPP composites.

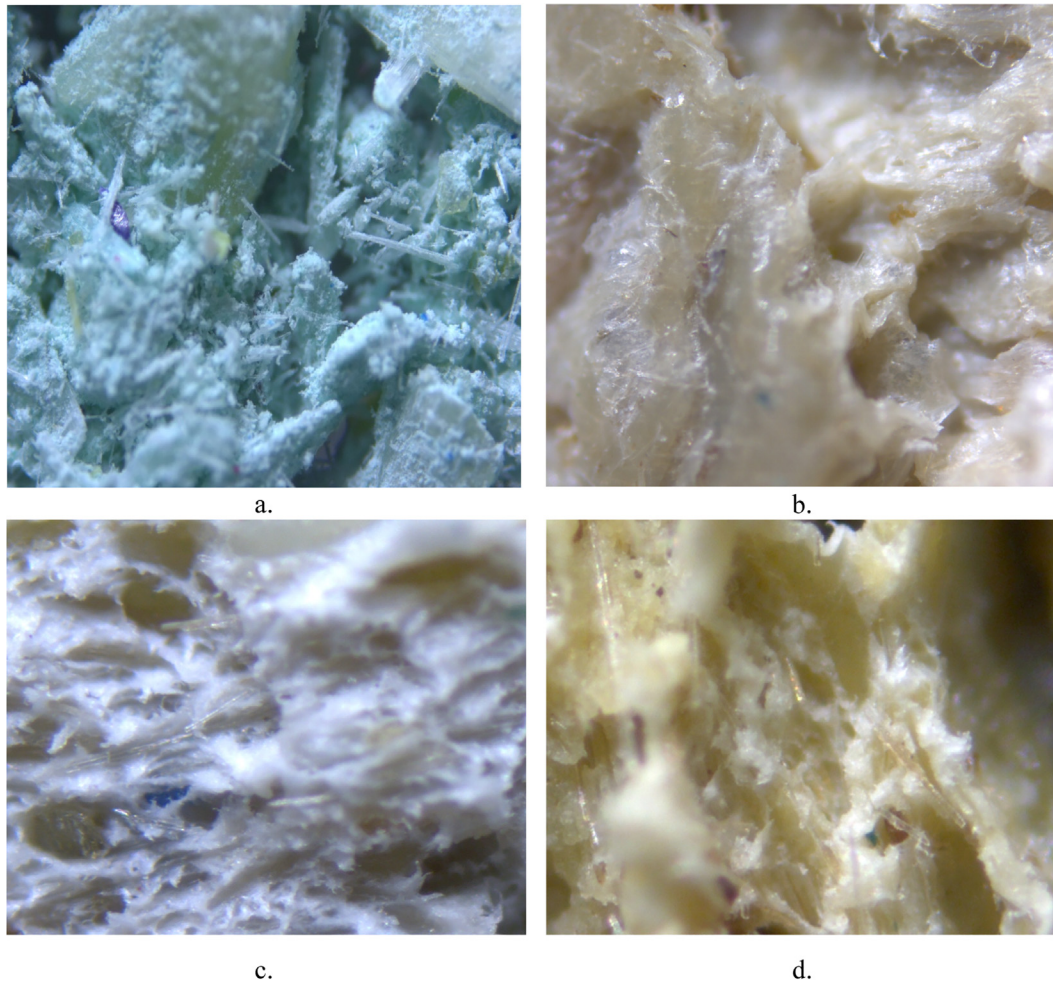


Fig. 7. Optical microscopy of WPCB (a), rPP + 30% WPCB (b), rPP + SBS + 30% WPCB (c), and rPP + SBS + SEBS-MA + 30% WPCB (d).

can be observed (Fig. 8a). The intensities of the rPP specific peaks are smaller with the addition of SBS and WPCB. A decrease of the crystallinity, as can be seen in Table 2, can lead to smaller peaks by rPP dilution with SBS. An increase in crystallinity and thus of the specific peaks of rPP when using SEBS-MA can be attributed to both of the compatibilization effect and the contribution of block-copolymer individual crystallinity.

The diffraction patterns of SBS and SEBS-MA (Fig. 8c) exhibit broad and amorphous reflexions of the block-copolymers at approximately $2\theta = 20^\circ$. The smaller intensity of the amorphous peak of SEBS-MA is due to a percent of the crystalline phase present in SEBS-MA.

Similar d-spacing for (110), (040) and (130) plane can be observed for rPP composites (Table 2): d_{110} from 5.921 to 5.982,

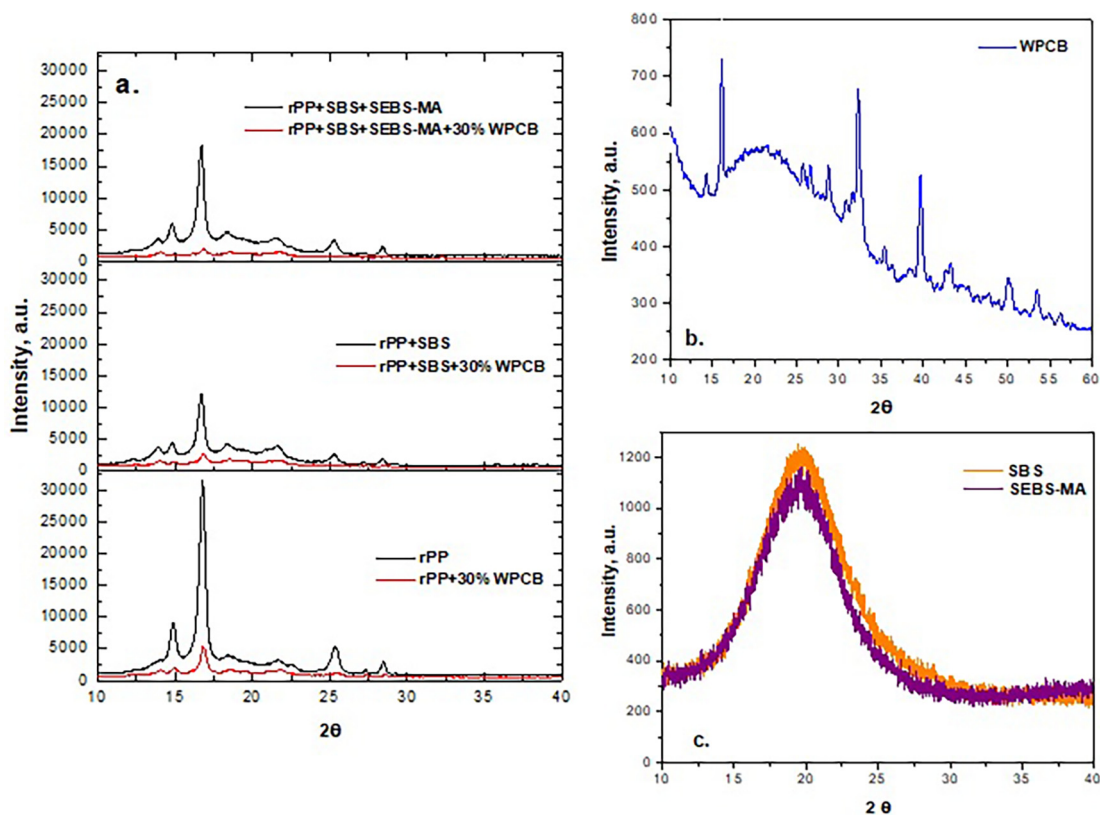


Fig. 8. XRD analyses for rPP/elastomeric composites (a) WPCB (b), and for pure block-copolymers (c).

Table 2

The crystallinity index (Ic) of the obtained composites.

Sample	rPP	rPP + WPCB	rPP + SBS	rPP + SBS + WPCB	rPP + SBS + SEBS-MA	rPP + SBS + SEBS-MA + WPCB
Ic, %	59.60	44.86	51.68	38.22	53.45	41.42
d_{110}	5.953	5.921	5.978	5.929	5.982	5.933
d_{040}	5.286	5.271	5.306	5.267	5.310	5.269
d_{130}	4.798	4.754	4.810	4.775	4.828	4.767
d_{111}	4.098	4.070	4.110	4.074	4.123	4.078

d_{040} from 5.269 to 5.309, d_{130} from 4.753 to 4.828. The distance between rPP planes presents a small increase when adding the elastomers, especially SEBS-MA, due to a compatibility improvement. The interphase modification in case of rPP + SBS + SEBS-MA + WPCB can be observed by comparing the d_{111} values for rPP composites (from 4.070 to 4.767) when a decrease of about 16% of the inter-planar distance resulted by adding both the block-copolymers and the waste powder as compared to rPP.

The crystallinity of all the samples was calculated and mentioned in Table 2. A decrease of Ic values was obtained by adding SBS (of about 13%) or both the block-copolymers (of 10%), as an expected influence of the elastomer phase that dilutes the crystalline rPP matrix. By adding 30%WPCB, the decrease in crystallinity is more pronounced, especially for the rPP + SBS + WPCB composite. The compatibility between polypropylene and the ethylene-butylene blocks from SEBS-MA and between the maleic groups and the waste powder leads to different interactions present into the system that could obstruct the crystallization process (Panaitescu et al., 2012; Vuluga et al., 2012).

The crystallinity of a material subjected to tensile stress is proportional with the mechanical resistance of the material so a smaller Ic can be correlated with a decreased tensile strength, as observed in Fig. 5. DMA analysis confirms this behavior by decreas-

ing the storage and loss modules in direct correlation with the composite crystallinity.

Analyzing the waste spectrum, peaks appeared in the range 10 and 25° on the XRD spectrum can be attributed to the polymer matrix. The peaks at higher angle suggest the presence of small quantities of metals due to their incomplete removal by leaching (Das et al., 2019; Hino et al., 2018), probably Cu, Fe, etc. Also, the complex composition of WPCB that contains fiberglass, silicates, fillers and additives, leads to numerous peaks on the diffraction patterns.

The elemental composition resulted from the WDXRF analysis is presented in Table 3. As expected, Si, Al, and Mg were found in WPCB that could be attributed to a high amount of fiber glass materials and mineral fillers. Fillers like calcium carbonate or titanium oxide can be present into the waste. Also, the presence of Br, Cl, and P could be attributed to fire retardants like polybrominated diphenyl ethers (PBDE), hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA) or chlorinated phosphate, the most used fire resistance modifiers for printed circuit boards. Similar values were also reported by other groups (Muniyandi et al., 2014; Yang et al., 2015). Metallic traces, as resulted also from XRD, can be observed, so the leaching must be more intense in order to eliminate all the quantity of Cu and Fe.

Table 3
WDXRF results for rPP, WPCB and their elastomeric composites.

Sample/Element, %	WPCB	rPP- 30% WPCB	rPP-SBS- 30% WPCB	rPP-SBS-SEBS-MA-30% WPCB	Fire retardant/mineral filler probably association
Br	7.7273	1.7907	1.3111	1.5025	PBDE, HBCD, TBBPA
Cl	2.0785	0.4264	0.1988	0.4012	Chlorinated phosphate
P	0.6312	0.0255	0.0176	0.0216	Phosphate
Si	19.177	3.2618	1.4617	2.3154	Glass fibers
Ca	6.0877	1.9342	0.8249	1.4068	Calcium carbonate
Mg	2.0012	0.0648	0.0401	0.0571	Magnesium hydroxide/magnesium silicate
Al	5.0593	0.6226	0.3591	0.4362	Aluminum silicate/Aluminum trihydrate
Ti	0.1185	0.504	0.1696	0.2537	Titanium oxide
Cu	0.1062	0.0243	0.0311	0.0210	-
Fe	0.0629	0.0245	0.0219	0.0274	-

All the additives remain into the composition after the thermal processing, in proportional quantities with the WPCB dosage into the composites. So, these small amounts of toxic compounds such as bromine, chlorine etc. that attend the security standard practice (GB5085.3-2007, 2007) for various applications can possible insure improved fire performance to the polyolefin composites.

Microscale combustion calorimetry (MCC) or pyrolysis combustion flow calorimetry (PCFC) has been developed in order to evaluate the flammability of materials using only milligrams of sample (DOT/FAA/TC-12/53, 2013; Lyon and Walters, 2004; Lyon et al., 2007). This small calorimeter can evaluate the heat release properties of pure polymers and has been used as a high capacity research tool for developing new materials (Wilkie et al., 2006). The method simulates flame burning by pyrolysis of the pure polymer in an N₂ medium to produce combustible gases, which are subsequently oxidized in an O₂-rich environment (Wath et al., 2015).

By MCC, important parameters regarding the fire risk of combustible materials can be measured: PHRR - maximum heat flux; T_{PHRR} - material ignition temperature, also known as temperature at maximum heat release flow; THR - total amount of the released heat; HRC - heat release capacity; HRR - heat release rate; Char Yield - the actual percentage of (solid) carbonized sample residue; * Decomposition rate - the sample decomposition rate. The tests were carried out under controlled conditions which assume that the temperature in the combustor is 900 °C and constant throughout the test and the gradual increase of the temperature at a rate of 1 °C/s from room temperature to 750 °C, in the pyrolyzer.

The WPCB powder contains a high amount of glass fiber that do not decompose, as can be seen in the char yield of 65.16%. HRC values indicate low flammability in the MCC test and low real-life fire hazard (Lyon et al., 2003). The HRC value is less than 100, so the material presents low ignition, as expected. The inflammability properties are good due to this glass fiber content, but also by another component which can delay ignition or reduce the rate of combustion contained by the waste that can improve the rPP fire performance.

The samples that contain elastomers and 30% waste powder presents the highest resistance to fire, with a HRC of 522 (J/(g*K)) and Char Yield of 18.75%. Compared to rPP, a reduction of 51.19% of HRC can be observed for rPP + SBS + SEBS-MA + WPCB as a result of the improved contact between the different phases of the components. Very close values were recorded for PP + SB

S + 30% WPCB. For rPP + WPCB the improvement is smaller, the HRC value is with 37% smaller than polypropylene due to the filler low flammability. As expected, 30% of WPCB added to the polyolefin matrix improves rPP fire resistance. Also, the total amount of the released heat during combustion and PHRR values are smaller for the composites in comparison with recycled polypropylene.

Because the percentage of carbonized residue is an indication of the amount of unburned fuel in a material treated with flammable substances, it is expected that the material with a higher char yield will produce a lower amount of heat during combustion (Cogen et al., 2009). This dependence can be also observed in case of the rPP composites.

The impact modifier can influence not only the mechanical properties of composites, but also the MCC results. As can be seen in Table 4, lower PHRR and THR are recorded for the neat elastomers, so the HRC value for composites is influenced also by the physical interaction between the components.

Differences can also be observed in HRR variation with time, where HRC is the maximum value of the release rate recorded in time. Thus, major discrepancies between the occurrences of HRC for the analyzed samples can be seen from Fig. 9. Even if WPCB has the lowest HRC, also the time is short, less than 120 s from

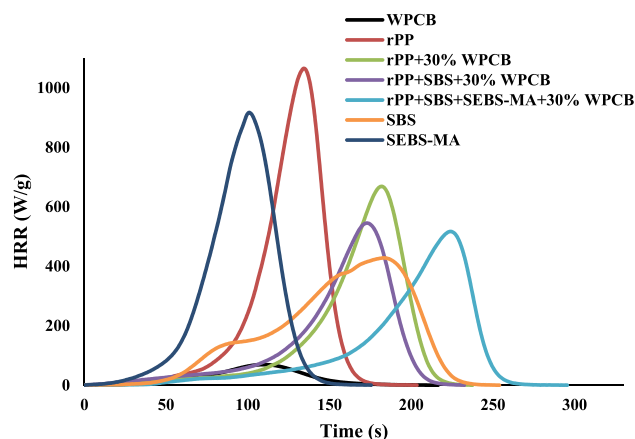


Fig. 9. HRR dependence on time recorded for rPP, WPCB and their elastomeric composites.

Table 4
MCC parameters recorded for rPP, WPCB and their elastomeric composites.

Sample	Char Yield (wt %)	THR (kJ/g)	PHRR (W/g)	T _{PHRR} (°C)	Time (s)	HRC (J/(g*K))
WPCB	65.16	4.94	69.09	393.27	110.00	93.03
rPP	0.00	42.26	1065.19	483.54	134.50	1071.18
SBS	0.00	40.20	427.94	479.83	182.50	727.09
SEBS-MA	0.00	41.54	916.84	468.50	101.00	929.20
rPP + 30% WPCB	15.30	34.17	668.47	481.81	182.00	675.36
rPP + SBS + 30% WPCB	18.25	30.24	545.08	480.01	173.00	553.07
rPP + SBS + SEBS-MA + 30% WPCB	18.75	31.39	516.79	479.39	224.50	522.84

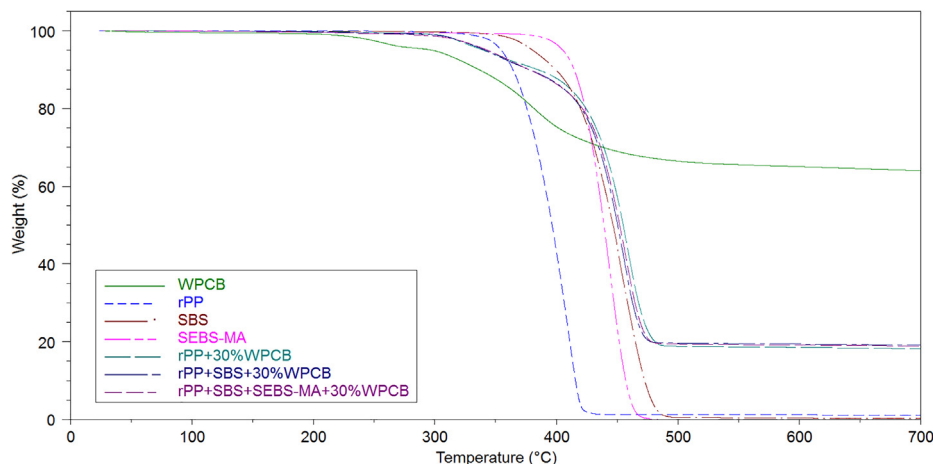


Fig. 10. TGA curves of components and rPP composites.

Table 5
Thermo-gravimetric analysis of components and PP blends.

Degradation step/Sample	35–120 °C		120–280 °C		280–355 °C		355–585 °C		585–700 °C		Residue/700 °C	
	Wt. loss %	Wt. loss %	T _{max} °C	Wt. loss %	T _{max} °C	Wt. loss %	T _{max} °C	Wt. loss %	Wt. loss %	N ₂ %	Air %	
WPCB	0.47	3.82	258.3	9.05	311.4	21.46	380.1	1.1		64.07	56.97	
Degradation step/Sample	35–230 °C			230–395 °C		395–525 °C		525–700 °C		Residue/700 °C		
rPP	0.06			–		98.74	459.8	0.12		1.08	1.08	
SBS	0.12			8.48	372.6	90.90	452.3	0.28		0.22	0.20	
SEBS-MA	0.03			1.69	364.8	98.11	444.4	0.03		0.14	0.13	
rPP + 30% WPCB	0.32			8.06	318.7	72.86	458.9	0.51		18.25	16.28	
rPP + SBS + 30%WPCB	0.41			12.12	323.7	67.88	455.6	0.44		19.16	16.85	
rPP + SBS + SEBS-MA + 30%WPCB	0.55			12.14	350.6	67.97	457.6	0.42		18.92	16.69	

the process start. In apposition with the inert filler, for the sample with 30% WPCB and two elastomers, the HRC appears after almost 240 s from the beginning of the process, being the most fire resistant. Probably during the obtaining process, due to the increase temperature, WPCB loses the contained volatiles and the glass fibers maintained in the system ensures a higher resistance to fire. The value of T_{PHRR} recorded for the rPP + SBS composite compared with rPP is not significantly change. HRC peak for the compatibilized sample appear at a time higher with 42 s in comparison with the polyolefin, so the fire resistance is improved.

The same thermal differences can be observed also in the thermo-gravimetric analysis, Fig. 10. The TG and DTG results obtained by heating the samples with a rate of 10 °C/min are presented in Table 5. For the non-metallic WPCB powder, the weight loss recorded at temperature below 280 °C can be attributed to an incomplete desolvation after the leaching. A 9–12% weight loss occurred from 280 °C to 355 °C due to the decomposition of bromine fire retardants (Tian et al., 2017). The most important degradation step was observed between 355 and 585 °C, with a maximum at 380.1 °C when most of the resins had decomposed, in accordance with literature (Quan et al., 2009). A 21.46% weight loss was recorded on this stage. The mass become stable from this temperature, meaning that the most of the volatiles were eliminated. The residue remained at 700 °C of about 57–64% is due to the high amount of glass fibers, in accordance with other studies (Kumar et al., 2018; Rajagopal et al., 2016). Thermal decomposition of epoxy resins containing brominated fire retardants takes place by secondary radical formation of phenols and their derivatives (Evangelopoulos et al., 2015).

The thermal stability of rPP composites show a slight improvement, the smaller weight loss on the most important stage of

degradation, of about 26–30%, is due to the glass fiber-reinforcing epoxy resins from WPCB. For the temperature interval from 280 to 355 °C, an increase of the maximum temperature is observed for the composite with 30% WPCB and compatibilized with SEBS-MA. The fire retardants improve the thermal stability of rPP by stopping the polyolefin degradation by radical scission of the macromolecular chain (Yang et al., 2016a). The residue obtained for the samples with 30% WPCB can be correlated with the powder residue.

4. Conclusions

The waste polypropylene and waste printed circuit boards were recovered as composites by elastomer compatibilization. Up to 30% WPCB was used and improved impact strength was obtained by adding two block-copolymers. Increasing the WPCB dosage from 5 to 30%, has decreased the impact strength value with 62–85% related to the control samples (without filler). By adding the elastomers, especially the malenized block-copolymer, a three times increased impact strength was obtained compared to rPP filled with the same dosage of WPCB due to the compatibility between the ethylene-butylene blocks from SEBS-MA and polypropylene and the MA groups and the functionalities of the waste powder, established by FTIR. The effect of the thermoplastic elastomers with high elasticity on the recycled polypropylene matrix was reflected in a significant increase in the elongation at break of the materials, the composites, initially brittle, becoming ductile. By increasing the filler dosage, the reinforcing effect was more significant than that of block-copolymer's elasticity, which leads to a decrease of elongation at break. The elongation at break increased by 30–19 times at the composites compatibilized with SEBS-MA

compared to rPP. The improvement of impact strength and elongation at break was obtained in detriment of tensile strength. The synergy between the elastic effect of elastomers and the reinforcing effect of the waste powder was evidenced also by the storage modulus and loss modulus increase.

The composites presented similar morphologies, as resulted from the XRD analysis. The possible interaction between SEBS-MA and rPP, respectively between MA groups and WPCB functionalities could be observed by the change in crystallinity and a decrease of about 16% of the inter-planar distance corresponding to (1 1 1) peak of rPP.

WPCB is composed, besides the thermosetting resin, mainly from fiber glass, mineral fillers, metallic traces and brominated and chlorinated compounds. The high amount of glass fiber that do not decomposed was highlighted also by thermogravimetry. The presence of Br, Cl, and P could be attributed most likely to usually used fire retardants that can modify the fire performance of rPP composites. By micro-calorimetry tests, and comparing the heat release capacity and its variation in time for components and rPP composites, the highest resistance could be observed for the polyolefin compatibilized with the two block-copolymers and with 30% waste powder. Also, a small increase of the thermal stability was observed when adding the elastomer component and the waste powder.

The improved mechanical characteristics and fire resistance of rPP elastomeric composites containing WPCB makes them suitable for various technical applications and industrial packaging.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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