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THERMAL BEHAVIOUR OF COMPOSITE MATERIALS OBTAINED FROM RECYCLED TETRA PAK® BY THERMOPLASTING FORMING

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ABSTRACT. Tetra Pak® (TP) multilayer packaging consisting of paper, polyethylene (PE) and aluminum, are widely used as aseptic packages in the food and beverage industry. Consequently, large quantity of packaging waste is generated, and many efforts have been made for recycling. The aim of this work was to develop new composite materials from TP cartons waste by thermoplastic method at different temperatures ranging from 120 ºC to 140 ºC. The resulting material was characterized by thermogravimetric/differential thermal analyses (TG/DTA) and differential scanning calorimetry (DSC) measurements to establish the thermal behavior of the prepared composites. Based on these results, the degradation pathway of the composites obtained from TP waste was proposed. The results of the study show that the preparation of thermoplastic composites represents a promising recycling process. The thermal degradation temperature of the prepared composites establishes a limit for practical use, which prohibits the utilization of these materials at temperatures exceeding this threshold.

Keywords: *Tetra Pak® waste, recycling, composite materials, thermal degradation*

INTRODUCTION

Tetra Pak® (TP) packaging is widely recognized as an aseptic solution in the beverage and liquid food industry. TP cartons are particularly popular for containing milk, wine, juice, and soft drinks and enable products to be

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distributed without refrigeration for extended periods without spoiling. Additionally, TP containers help maintain the nutritional quality and hygiene of the contents [1-3]. The development of milk packaging began in 1943, led by Swedish engineer Ruben Rausing. The brand name Tetra Pak® was established in 1944, inspired by the tetrahedral shape of the cartons. TP cartons gained popularity due to their ability to efficiently transport and store milk foams. In 1961, TP introduced aseptic sterilization technology, enabling the production of bacteria-free milk. This technology further evolved to allow products to be stored for up to six months without refrigeration using the ultra-high temperature (UHT) pasteurization process [4].

It is known that TP consists mainly of three materials, namely paper, polyethylene (PE), and aluminum (Al) foil, organized into six-layers. The type of paper used in composite beverage cartons varies based on the packaged product, the region where it will be sold, and the manufacturing conditions. Generally, paper constitutes 75% of the total weight of the package, while PE accounts for 20%, and Al makes up the remaining 5% [5]. The primary function of the paper is to ensure the package maintains its rigid shape and to enhance material strength. The Al foil is included due to its excellent barrier properties, protecting against light, oxygen, water vapor, and microorganisms, whereas PE serves several purposes, it protects the paper layer from water, safeguards the Al layer from direct contact with the contents, bonds the layers together, and allows for heat sealing [6].

Packaging waste leads to significant environmental and financial impacts, prompting various efforts aimed at recycling. The concept of a circular economy seeks to prolong the life of products through strategies such as reuse, energy recovery, and recycling. Depending on the level of processing involve, the recycling process encompasses various stages, as production recycling, material and feedstock recycling. [4, 7-9].

In such circumstances, it is essential to establish a TP recycling system and explore effective technologies for recycling TP materials [10]. The first method used to recycle TP was the particle board method. In this process, the cartons were not separated into their components before being processed through thermal compression, resulting in boards that could be used in furniture, civil construction, and packaging industries. In subsequent years, a new recycling method known as hydro-pulping was developed. This method allowed for the recovery of paper fibres, which make up 75% of the TP cartons. After hydro-pulping, the remaining PE and Al parts are processed using various technologies, including plastic product transformation, energy recovery, pyrolysis, and plasma technologies. However, these processes are quite expensive and complex. Nowadays, the collected TP are either accumulated in certified collection and separation facilities, placed in landfill areas, or burned in cement plants [11].

One of the most promising applications of the TP recycled cellulose is the preparation of thermoplastic composites [12]. Manny research groups have concentrated their efforts on the production of composite materials utilizing packaging waste in order to better validified such materials. Obtaining composite materials, particularly from TP waste, represent a valuable resource that can serve as an alternative for various structural and non-structural applications. The optimization of processing parameters for recycled TP is critical, as the properties of these composites are influenced by several factors, including material type, mixing ratio, mixing duration, material size, and the employed technology. A range of technologies is available to produce new composites, with compressive moulding being among the most prevalent. Research indicates that the Al layer present in TP can significantly minimize water absorption, while the high cellulose content in TP reduces the necessity for supplementary reinforcement materials. Furthermore, TP enhances multiple mechanical properties of composite materials, including density, thickness, swelling, elastic modulus, and compressive strength. Therefore, the development of new composite materials derived from packaging waste not only alleviates environmental pollution but also contributes to the reduction of waste management costs [13-15].

To enhance the utility and quality of composite materials derived from TP waste, it is crucial to study their thermal properties, as these determine potential applications. Due to their chemical composition, these materials are both thermally degradable and combustible. The characteristics of their thermal degradation significantly impact their performance and sustainability. The thermal degradation temperature establishes a definitive upper limit for practical use. Therefore, these materials cannot be utilized at temperatures beyond this threshold. Gaining a thorough understanding of thermal degradation behaviour is essential for various practical applications [16].

Many efforts are made to enhance the recycling capacity for carton packages across the European Union (EU), aligning with the proposed EU Packaging and Packaging Waste Regulation (PPWR), which are very important in facilitating the transition towards sustainable food packaging and to achieving greater material circularity. In this context, the novelty of this study consists in preparation and characterization of new composites materials obtained from TP waste by using an advantageous method with a low energy consumption and respecting the principles of the circular economy. Consequently, the aim of this work was to investigate the thermal behaviour of TP composite materials, and their production parameters influence on the critical temperatures and weight loss. Such approach gives important data for further establishing of these composites' application areas.

RESULTS AND DISCUSSION

In order to evaluate the thermal behavior of new composite materials based on TP waste as raw material, a series of samples were obtained using the methods developed in our research group [17]. The samples codes used in this study and the process parameters are presented in *Experimental section*.

Thermal analyses of the new composite materials were carried out under the same conditions to determine the thermal degradation behaviors. Thermogravimetry (TG), its derivative (DTG) and differential thermal analysis (DTA) profiles of composite materials prepared at different temperatures are displayed in Figures 2-6. TG, DTG and DTA curves of the composite thermoformed at 120 ºC (S1-120.5) are shown in **Figure 1**. The initial weight loss between 24.21 and 97.75 °C was only about 3.39%, suggesting the moisture loss. The primary weight loss of 59.95% occurred within the temperature range 213.16 - 382.65 °C, with the maximum weight loss rate peak at 332.61 °C, which was caused by the decomposition of cellulose fibers from the composite material. About 25.34% of the initial weight was lost between 382.65 and 495.64 °C, with a weight loss peak at 448.83 °C, which was caused by the decomposition of the PE layers. It was expected that the char is formed mainly from the cardboard part of the packaging used in the composite preparation, so the residue after 500 ºC probably consisted of the aluminum and the char formed. The results of the analysis were consistent with previously reported studies on TG analysis of waste TP packaging [18-20]. The behavior of the thermal decomposition of composite (S1-120.5) obtained from TP waste clearly shows two important different stages, with the maximum decomposition rates at 332.61 °C and 448.83 °C, in agreement with the results obtained by Haydary et al. [19]. The first stage corresponds with thermal decomposition of cellulose and hemicelluloses to an intermediate product and the second stage includes decomposition of low-density polyethylene (LDPE) and the intermediate product.

The decomposition profiles of the other composites (S2-125.5, S3-130.5, S4-135.5, S5-140.5) are quite similar (**Figures 2÷5**), following the 4 stages as revealed in **Table 1**. After the initial mass loss, which corresponds to the moisture loss, two important separate stages of mass loss were clearly observed, the first for the degradation of the cellulose fibers and the second for the decomposition of the PE from the composite plates.

DTA curves can be a viable source of information about energy absorption during the thermal conversion of different kinds of materials. The DTA curve of S1-120.5 sample can be seen in **Figure 1**. As is evident, the exothermic peaks at 250.29, 347.94 and 451.25 °C were caused by the decomposition of cellulose and PE, respectively, which were in accordance with the DTG peaks. The melting of Al would lead to the presence of a very small weight loss peak on the DTG curve, it consequently created a weak endothermic peak at 660.28 °C. It should be noticed that temperatures used for the preparation of the materials do not show major differences between the behaviors of the TG, DTG and DTA curves (**Table1**).

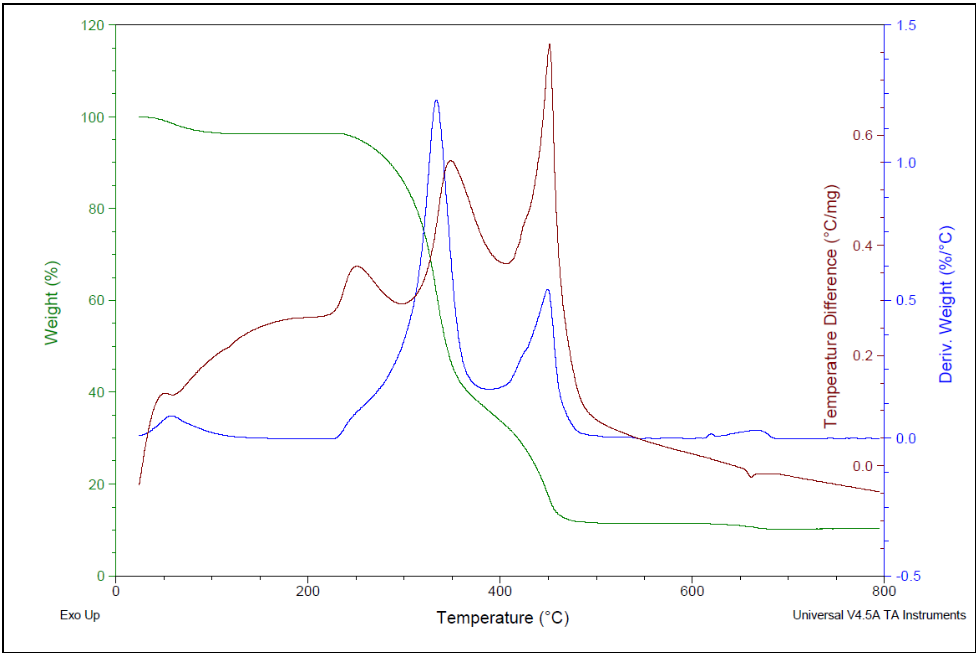


Figure 1. Thermogravimetry (TG- green), its derivative (DTG-blue) and differential thermal analysis (DTA- red) curves of sample S1-120.5

Table 1. Characteristic temperatures and mass losses for the thermal degradation reaction of the prepared composites from TP waste

Thermoforming temperature	S1-120.5 120 °C		S2-125.5 125 °C		S3-130.5 130 °C		S4-135.5 135 °C		S5-140.5 140 °C	
Thermal steps	T_i-T_f (°C)	m%	T_i-T_f(°C)	m%	T_i-T_f(°C)	m%	T_i-T_f (°C)	m%	T_i-T_f(°C)	m%
Moisture evaporation	24.21-97.75	3.39	26.27-117.12	3.42	26.72-92.10	2.52	33.99-92.10	2.89	28.34-97.75	2.96
Step I	213.16-382.65	59.95	206.70-397.98	60.52	199.44-397.98	62.63	219.62-395.56	63.53	204.28-410.09	65.8
Step II	382.65-495.64	25.34	397.98-515.82	22.10	397.98-512.59	24.83	395.56-498.87	19.70	410.09-496.45	17.78
Step III	495.64-694.99	1.40	515.82-699-83	1.95	512.59-703.06	1.76	498.87-701.45	2.06	496.45-708.71	1.95
Total mass lost		90.08		87.99		91.74		88.18		88.49

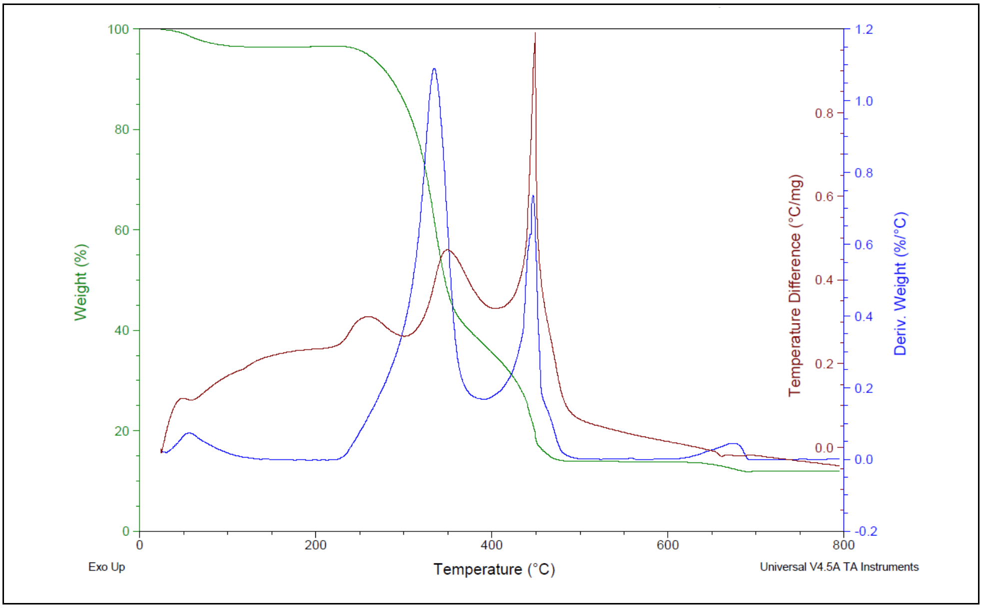


Figure 2. Thermogravimetry (TG- green), its derivative (DTG-blue) and differential thermal analysis (DTA- red) curves of S2-125.5 sample

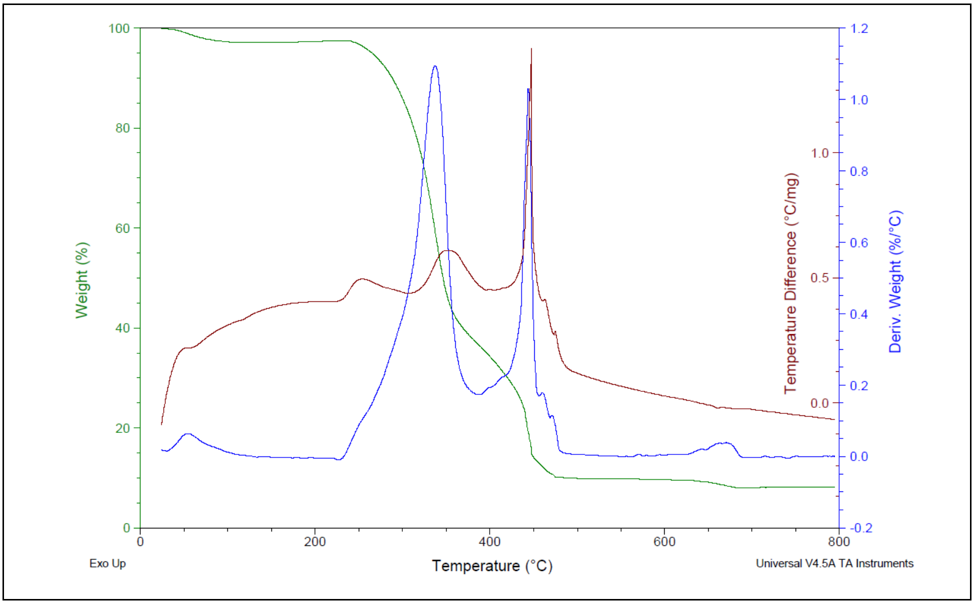


Figure 3. Thermogravimetry (TG- green), its derivative (DTG-blue) and differential thermal analysis (DTA- red) curves of S3-130.5 sample

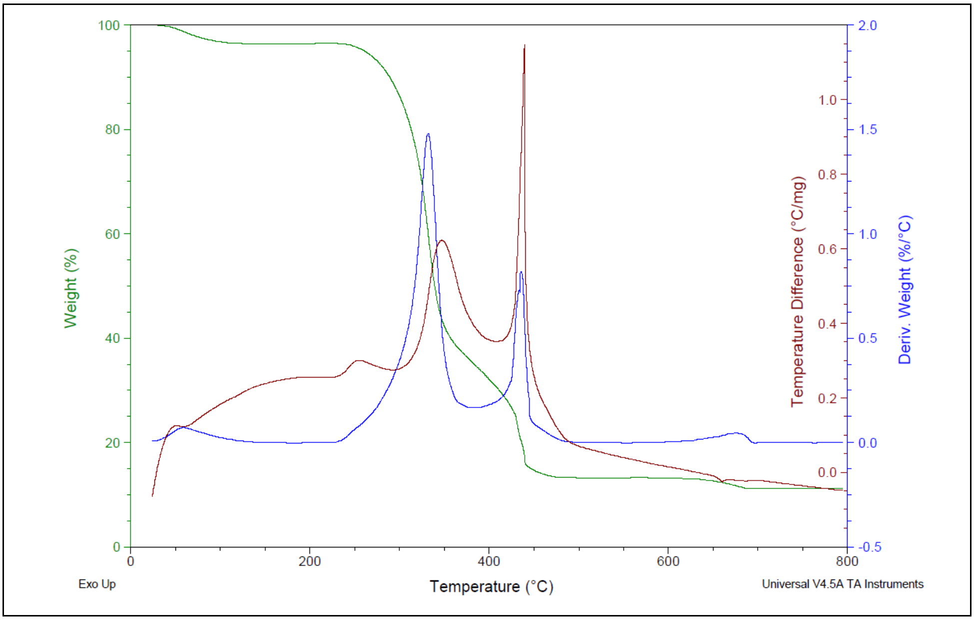


Figure 4. Thermogravimetry (TG- green), its derivative (DTG-blue) and differential thermal analysis (DTA- red) curves of S4-135.5 sample

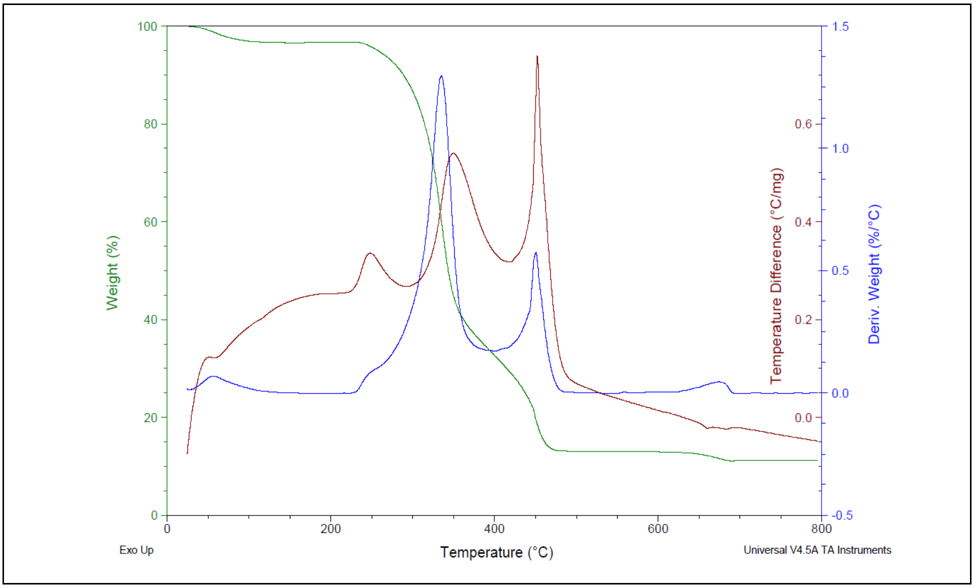


Figure 5. Thermogravimetry (TG- green), its derivative (DTG-blue) and differential thermal analysis (DTA- red) curves of S5-140.5 sample

Figure 6 shows the DSC curves of composite materials (S1-120.5, S2-125.5, S3-130.5, S4-135.5 and S5-140.5) obtained at different temperatures.

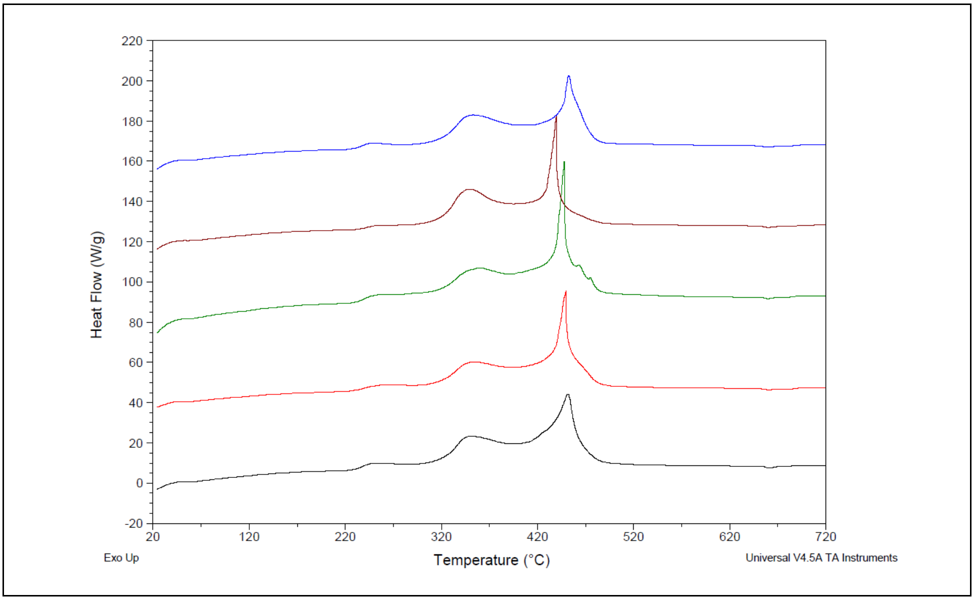


Figure 6. DSC curves of composites S1-120.5 (black), S2-125.5 (red), S3-130.5 (green), S4-135.5 (yellow), and S5-140.5 (blue), respectively.

All five DSC curves have three exothermic peaks between 220 ºC and 450 ºC. The first two exothermic peaks are attributed to the decomposition of cellulose. The third peak at ~ 480 °C showed the decomposition of low LDPE type PE polymer in the composites. Finally, the last peak ~ 660 °C was related to an endothermic process of fusion of aluminum in the samples. In general, the melting point of LDPE is lower than the linear low-density polyethylene (LLDPE).

The development of new materials with enhanced performance using various waste materials has gained significant attention in recent years, primarily due to environmental and socioeconomic benefits. In the study conducted by Yılgör et al., composite panels were manufactured from TP waste [20]. The thermal degradation properties of these panels were analyzed using TG/DTG methods. Their study demonstrated that it is possible to produce panels from TP waste without generating additional waste, in addition to utilizing them in the pulping process. The thermal degradation of the paper layer occurred at lower temperatures compared to the LDPE layer and the composite panel. Specifically, the thermal degradation of the paper layer began at temperatures ranging from 200 ºC to 350 ºC, while the LDPE layer started degrading at 432 ºC, in agreement with our results. Additionally, aluminum does not degrade during thermal treatment and melts at approximately 600 ºC, indicating that it does not influence the TG and DTG profiles of the panels. These composite panels may have limited applications because of the hydrophilic nature of cellulose in their structure. However, they can be considered for indoor applications where there is a low risk of fungal decay, and the use of additives is not required [20]. In our study, the thermal degradation of the TP composites began at temperatures beginning from ~ 200 ºC for any of the studied samples. Due to the different forming temperatures, ranging from 120 ºC to 140 ºC, which do not exceed the beginning of the degradation temperature, there is no influence on the higher critical decomposition temperatures.

In a recent study, Sergi et al. proposed the use of LDPE/Al mix obtained after the recovery of cellulose from multilayer aseptic packaging as a matrix for composite materials reinforced with short natural basalt fibers [21]. Consequently, an assessment of its thermal stability and the effects of the basalt fibers was necessary. A commercially available material, EcoAllene, which is obtained from the recycling of multilayer packaging, was reinforced with short natural basalt fibers at levels up to 30 wt.% using twin screw extrusion. The goal was to enhance the mechanical properties of this material and expand its potential applications. Thermal characterization through TG and DSC analysis revealed that the material is a complex mixture comprising LDPE, high-density polyethylene (HDPE), polypropylene (PP), and aluminum (Al). The addition of basalt fibers did not alter the melting and crystallization profiles, nor did it affect the overall degradation behavior of the composite. The EcoAllene, along with polyethylene and aluminum waste, exhibited a single-step degradation behavior with a maximum degradation temperature of approximately 485 ºC, finding which agree with existing literature [22]. This degradation step could be attributed to the presence of LDPE, while the resulting residue indicates the Al content, which is expected to show no significant weight loss.

Cravero et al. included in their study the thermograms of a pure LDPE which exhibited a maximum degradation temperature around 474 ºC [22]. In the work authored by Hidalgo-Salazar et al. [23], the authors reported a maximum degradation temperature around 470 ºC. Recently, Georgiopoulou et al. [24] reported, for a LDPE/Al by-product recovered by a selective dissolution/precipitation process, a maximum degradation temperature of 482 °C, with an onset degradation temperature of 429 °C, which are in close agreement with the values obtained by Sergi et al. In our work, the maximum degradation temperature in all the samples studied was found to be ~ 450 °C, in close agreement with the other recent studies.

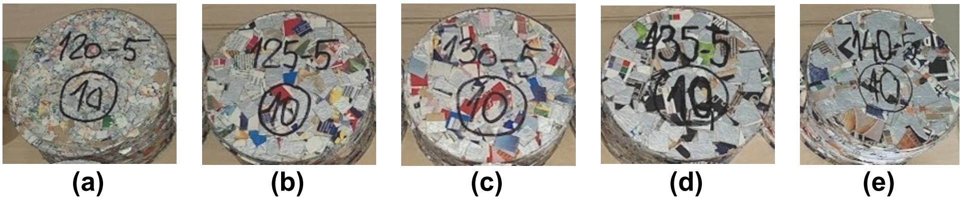
CONCLUSIONS

The recycling of waste TP packages composed of paper, LDPE, and Al is of great recycling significance. In the present work we have investigated the thermal degradation behavior of composites produced from TP waste without any significant recycling process, by thermoplastic forming. Thermal behavior of the resulting materials was studied and the results showed that thermal degradation consisted of two important distinct steps after the moisture evaporation: the first step was caused by the decomposition of cellulose fibers from the composite material and the second step corresponds to the degradation of LDPE. It’s clear that the residue consists of char and aluminum after the thermal degradation of the composites. Results from the present study show that these low-cost thermoplastic composites show potential in many applications, respecting the decomposition temperature found to begin at ~ 200 °C as proposed by the thermal analysis, which prohibits the utilization of these materials at temperatures exceeding this threshold.

EXPERIMENTAL SECTION

Composites preparation

The composite materials were obtained from multi-layer packaging wastes, containing aluminum, paper and polyethylene terephthalate (e.g. juice packs, milk packs, etc.), using the procedure previously described in literature by our group [14]. The multi-layer packages have been carefully washed and cropped out into small pieces. Afterwards, the obtained material was compressed at different temperatures, 120 ºC, 125 ºC, 130 ºC, 135 ºC and 140ºC, without any kind of bonding materials addition, with the obtaining of some plates, as illustrated in **Figure 7**. The composite material plates have been produced in a



(a) **(b)** **(c)** **(d)** **(e)**

Figure 7. Composite plates obtained at (a) 120 ºC, (b) 125 ºC, (c) 135 ºC and (d) 140 ºC, respectively.

specific mold (**Figure 8**). To ensure the heating of the material, the mold was endowed with an electrical resistance, which guarantees the required forming temperature. The samples presented in **Figure 7**, have been named considering the forming temperature, as follows: S1-120.5, S2-125.5, S3-130.5, S4-135.5 and S5-140.5, respectively (see **Table 2**).

Figure 8. The mold used for the fabrication of composite plates.

Table 2. Samples codes and process parameters

Samples Code	Process parameters
S1-120.5	T = 120 °C, P= 5MPa
S2-125.5	T = 125 °C, P= 5MPa
S3-130.5	T = 130 °C, P= 5MPa
S4-135.5	T = 135 °C, P= 5MPa
S5-140.5	T = 140 °C, P= 5MPa

Thermal analyses

The TA Instruments SDT Q600 Thermogravimetric Analyzer (TGA) & Differential Scanning Calorimeter (DSC) was used for simultaneous measurement of weight change and differential heat flow on the composite samples from ambient to 800 ºC. Data on thermogravimetry (TG), its derivative (DTG), differential thermal analysis (DTA), and differential scanning calorimetry (DSC) curves, were simultaneously acquired under the following measurement conditions: heating from laboratory temperature to 800 °C at a heating rate of 10 °C/min, under normal air atmosphere, using alumina pans.

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