Evaluation of Mechanical Properties of Composites Manufactured from Recycled Tetra Pak® Containers

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Abstract

Context: Tetra Pak® is a common material used for the production of food containers. Currently, those containers are recycled by physical separation of the cellulose fibers through a hydro pulped process, but sometimes separating the individual components is not economically viable. This work evaluates an alternative process to obtain composites from recycled Tetra Pak®.

Methodology: Tetra Pak® used containers were collected and cut into small pieces at the laboratory. Then, the containers were hot-pressed to obtain sheets in a manual hydraulic press by using different configurations. Samples were cut, and their tensile strength was evaluated (ASTM D3039). Failure analysis of samples was carried out by FE-SEM to identify issues related to processing and to understand the differences in mechanical properties.

Results: The results showed that the lowest tensile strength was 9.5 MPa (type I sample) and the highest tensile strength was 37.4 MPa (type III sample).

Conclusions: The results of mechanical tests show that this material can be used for non-structural purposes in the building industry. Failure analysis shows that fiber pull-out and delamination are the most important failure mechanisms in type I samples. For type III sample, failure was produced by a sequence of intralaminar fractures.

Keywords: composites, hot-pressing, mechanical properties, recycled, Tetra Pak®.

Resumen

Contexto: Tetra Pak® es un material común que se utiliza para recipientes de alimentos. Actualmente, esos contenedores se reciclan mediante la separación física de las fibras de celulosa a través de un proceso de hidropulpeado, pero a veces no es económicamente viable separar los componentes individuales. En este trabajo, se evaluó un proceso alternativo para obtener materiales compuestos de Tetra Pak® reciclado.

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INTRODUCTION

Current efforts in material science aim to reduce the use of primary materials as wood (Ayrilmis et al., 2013; Hidalgo et al., 2013a). Composites made from waste seem to be a good option and have gained increasing attention (Souza et al., 2018; Moreno & Saron, 2017). Tetra Pak® is a material used for food packaging and consists of three layers: carton, polymer, and aluminum. Those containers are recycled by physical separation of the cellulose fibers through three main processes: incineration, aluminum recovering by pyrolysis, and processing in an extruder (Lopes & Felisberti, 2006). However, in most of cases it is not economically viable to separate the individual components because although composites have been obtained, there is still a lack of knowledge to identify the variables required to process them and turn them into something functional.

Ali & Nystrom (1995) used Tetra Pak® in their work and performed a physical separation of polyethylene and aluminum (PEA) from the cellulose fibers through a hydro pulped process. In their work, the authors used water and shook the Tetra Pak® containers for several minutes (30-45 min). After an appropriate shaking time, the cellulose phase and PEA were partially separated from each other. The authors also added an accelerator (NaOH) to remove hydrogen bonds and recover more fibers. A similar procedure was done by Turrado et al. (2012). However, at the end of the process PEA still contained some cellulose.

Recycling of containers is possible using different processes. Hidalgo et al. (2013b) indicated several methods used to manufacture polyethylene aluminum (PEA), such as hot plates compression, extrusion, injection, and rotational molding. The authors showed several applications for the PEA such as plastic wood and automobile applications, among others. However, they did not use 100% of the containers but developed the products using the PEA in all cases. On the other hand, Paula et al. (2005) used PEA with recycled HDPE/LDPE and virgin PolyEthylene (PE) resin to get materials with improved mechanical properties, but increased the water absorption by the polyamide presence in the HDPE and LDPE recycled. However, all the mixtures had excellent chemical resistance. Salamanca & Vaca (2017) have reported the effect on mechanical properties (tensile and flexural strength) of a thermoformed composite (90 wt% Tetra Pak® and 10 wt% LDPE). Galvis & Villabona (2016) have studied the feasibility of using 100 wt% Tetra Pak® for structural applications. Bekht et al. (2016) made thermoformed panels using several types of containers.
(including Tetra Pak® and candy packing) and they found that the new product has improved mechanical properties when it was compared to virgin resin of PE.

Recent studies (Kaseem et al., 2017) indicated that small wood particles used as reinforcements (in sizes 100 μm and 600 μm) improved the adherence between the particles and the polymeric matrix. Valim et al. (2015) analyzed the possibility of using polyethylene aluminum (PEA) composite in buildings. However, the composites were affected significantly by exposure to UV light, thus degrading mechanical properties like impact resistance and strain resistance. So, the authors concluded that PEA might be used but not in structural components of civil constructions. New uses and modifications for the PEA were also reported by Caraschi & Leão (2003). In their work, the authors showed that chopped Tetra Pak® might be used as reinforcement in a polypropylene matrix since the natural fibers in the container increased the tensile strength if the content of recycled material is near to 15% wt. In another study Ebadi et al. (2016) evaluated Tetra Pak® wastes as a reinforcement in polymers. The authors found that tensile strength of composites increased 26% after including Tetra Pak® wastes. In another application of Tetra Pak®, Silva et al. (2015) fabricated several types of roofs, made from 100% PEA using zinc and PEA. The authors evaluated the tensile strength and the sheets capability for use as thermal and sound insulators. In another work, Rhamin et al. (2013) concluded that recycled panels made from Tetra Pak® might be used for internal or external applications.

In summary, several authors are trying to manufacture composites from recycled Tetra Pak®. So, this work aims to assess the mechanical properties of composite materials manufactured from 100 wt% recycled Tetra Pak® containers and to avoid the extensive use of water and resins. The premise is that the preparation of raw material must be done without extensive labor and water would not be used to separate or modify the containers to obtain a most sustainable process. Several samples were prepared by modifying the size of raw material and the manufacturing parameters. After obtaining the samples, the mechanical properties and water absorption tests of the composites were evaluated. Also, the fracture surface of the samples was analyzed by FE-SEM in order to understand the main failure mechanism. The results were compared with other materials used in non-structural applications to demonstrate the capabilities of the composites obtained in this article.

**EXPERIMENTAL METHODOLOGY**

**Materials Processing**

Commercial Tetra Pak® containers with dimensions 4.6 mm L x 3.6 mm W x 11.85 mm H were collected. The containers are composed of several layers of low-density polyethylene (LDPE) 20% wt., Aluminum (Al) 5% wt. and cellulose 75% wt., according to the manufacturer. Three different samples were prepared to develop this experiment. The samples were prepared in order to obtain composites without additives or reinforcements, and by using a scalable process to obtain sheets. Initially, the containers were disassembled and stacked manually. Fifteen as-received containers were stacked in 5 layers to obtain a final composite without cutting the containers and stacked manually. Fifteen as-received containers were stacked in 5 layers to obtain a final composite without cutting the containers and create Type I samples; Type II samples were prepared by cutting the containers into slices of 10 mm x 15 mm; and type III samples, by cutting containers into pieces of 5 mm x 5 mm. After positioning the layers in a mold of 250 mm x 120 mm x 7 mm and applying several cycles of pressure and temperature in a manual hydraulic press with hot plates, the composites were obtained. The processing parameters used in this work were selected according to the literature (Sun & Zhang, 2013), and the limitations of the equipment available in the polymers laboratory of the Metropolitan Technological Institute (ITM, in Spanish). After cutting, samples were hot-pressed without any additional binder to form the composite, so the final composite is obtained using 100% recycled materials.

The procedure C recommended in ASTM D4703-03 was used: a suitable working method
for thermoforming polyethylene sheets with a working pressure higher than 8 MPa. Figure 1 outlines the procedure, as well as the pressure, temperature, and time during the sheets manufacturing.

**Mechanical properties**

The composite sheet (200 mm x 120 mm x 7 mm) was cut manually to obtain the samples, which were prepared according to the ASTM D3039 standard (Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials) by aligning the test specimens by the sheets’ longest dimension (200 mm). The mechanical tests were done in a Shimadzu AGX-100 KN Universal testing machine available in polymers laboratory of the ITM. All samples were tested at a testing speed of 5 mm/min at 25 °C. In all cases, for every type of material, three samples of each type were tested, and the results were averaged.

**Analysis of fractured surfaces**

After performing mechanical tests, the samples were analyzed to understand the main failure mechanism and to determine how the manufacturing process could be improved. The samples were observed in a LEICA EZ4D stereomicroscope and in a FE-SEM 7100F available in the microscopy laboratory of the ITM.

![Figure 1](image-url). (a) Pressure, temperature, and time during the sheets manufacturing. (b) Details about the mold and the slices used during the manufacturing.

**Source:** Authors.
ITM. The samples were covered with a thin layer of gold to avoid surface charging.

Water absorption tests

Water absorption tests were done according to the ASTM D570-98 (2005) standard. The samples weight was recorded before and after soaking them in water for 24 hours. Both weights were compared to obtain the water absorption. The dimensions of samples were 76 mm x 24.5 mm x 7 mm (length, width, and thickness, respectively). The procedure described in numeral 7.1 of the ASTM D570 standard was used: immersing the specimen in distilled water during 24 h at 23 °C ± 1 °C.

RESULTS AND DISCUSSION

Mechanical properties

Figure 2 shows the average and standard deviation of tensile strength of the manufactured samples. The lowest resistance was found in Type I samples and the highest was found in Type III samples. Type I samples had even lower resistance than that of the LDPE matrix (13.3 – 26.4 MPa) (Barcelona, n.d.). In this case the low resistance of the composites can be explained by the lack of preparation of Type I samples since the samples were hot pressed without cutting the container.

Type II samples showed an intermediate resistance (24.1 MPa) and the standard deviation of the results shows that the manufacturing process is uniform. Conversely, Type III samples (chopped material) report the highest tensile strength, but they also have the highest standard deviation, so the processing conditions did not provide uniform materials.

This behavior can be explained by the different orientations of the chops used for the manufactured composites. Since the orientation of the chops was not controlled during the manufacture, the fibers randomly oriented can affect the tensile strength measured in just one direction. Table 1 lists different materials investigated by other authors or commercial materials and shows the maximum tensile strength possible. It is observed that the resistance of the materials investigated in this work is similar to that observed by other authors. Moreover, the resistance

![Tensile strength of composites obtained from recycled Tetra Pak®](image)

**Figure 2.** Results of tensile tests.

**Source:** Authors.
of the prepared recycled Tetra Pak® composites without any further modification is similar even though those reported in the Table 1 were modified with resins and polymers. Also, some authors have studied the mechanical properties of materials for non-structural purposes in the construction industry. The values reported in this work are comparable with those found in literature for Short Pineapple-Leaf-Fiber (PALF)-LDPE Composites (10.2 MPa–13.0 MPa, and 16.3 MPa–22.5 MP, for randomly and longitudinally oriented fiber, respectively) (George et al., 1995), and for Wood-plastic board (WPB), sugarcane bagasse particleboard (SBpb), Plywood board (PB), and Sugarcane bagasse fiberboard (SBfb), whose values are 28.20 MPa, 14.35 MPa, 15.20 MPa, and 31.45 MPa, respectively (Martínez-López et al., 2014) for non structural purposes.

Hitchon & Phillips (1979) reported that the fiber strength decreases as the length of the fiber increases; and Pamungkas et al. (2018) showed that the fiber length increases the mechanical properties of a composite of Zalacca Midrib Fiber (ZMF)/HDPE. This article does not include tests for the effect of the fiber length, but the size of the chops can be interpreted as an indirect evaluation of the fiber length. The results of this article show that the shorter the chops, the tensile strength increases. However, some authors have found ambivalent results, so there is no clear trend about the effect of fiber length on tensile strength (Amuthakkannan et al., 2013).

On the other hand, when the fibers are perpendicular to the direction of the applied stresses, they act as stress raisers, but it is worth noticing that the fibers in the fragments of the samples had a random orientation and their effect was not studied. When the fibers are cut into small pieces, there is a higher probability of finding fibers perpendicular to the load application direction, which would imply a lower resistance of the compounds in this order: Type III < Type II < Type I. However, the behavior of the tests for this research was contrary leading to the phenomenon associated with the direction of the fibers being negligible. A similar behavior has been reported in the literature by other authors (Xia et al., 2002).

### Analysis of fractured surfaces

Figure 3 shows the SEM images of fractured Type I sample. They indicate that there is a fiber pull-out after the mechanical tests. Some fibers were found to be coated with LDPE and adhesive layers, as indicated by the rows in Figures 3a and 3b.

Also, it is possible to observe that LDPE fibers stretched resulting in a ductile failure of the material. Most of the fibers did not show a separation from the matrix, which mean that the processing parameters can melt the polymer around the fibers. Also, uncoated fibers were observed and most of them were released from the matrix (Figures 3a and 3b). Figures 3c and 3d show that the delamination of some layers and the empty spaces between them (see arrows). Thus, it can be concluded from Figure 3 that pull-out and

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### Table 1. Materials reported in the literature with similar properties.

<table>
<thead>
<tr>
<th>Material</th>
<th>Tensile strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Recycled Tetra Pak®-Based Boards (RTPBB) (Quintero et al., 2017)</td>
<td>≈ 5.67 – 6.60</td>
</tr>
<tr>
<td>LDPE-AL reinforced with 2-dimensional disposable fibers (Hidalgo et al., 2012)</td>
<td>24.86</td>
</tr>
<tr>
<td>Non-reinforced plastic wood board (Martínez-López et al., 2014)</td>
<td>28.20</td>
</tr>
<tr>
<td>Plastic wood reinforced with 2.5%wt. of clay nanoparticles (Yadav &amp; Yusoh, 2015)</td>
<td>32.04</td>
</tr>
<tr>
<td>LDPE-AL reinforced with 50%wt. of fique fibers (Hidalgo et al., 2011)</td>
<td>67.26</td>
</tr>
</tbody>
</table>

**Source:** Authors.
delamination are the common failure mechanisms in Type I sample.

The surface analysis of Type III samples showed that the composite did not fracture in a transverse plane form (Figure 4). These failures are characterized by being staggered and are caused by separation of layers. Figure 4 also shows the detail of the failure observed in the SEM. Additionally, it was observed that the fracture has been produced by a sequence of intralaminar fracture in cellulose fiber layers. Greenhalgh (2009) reported that the intralaminar fracture is intrinsically linked to fiber-dominated failure. This work also identified fiber failures since there is a reduction in the cross-section area of the fibers.

**Figure 3.** SEM images of failed Type I sample.

**Source:** Authors.

**Figure 4.** SEM image of the failed Type III sample.

**Source:** Authors.
For Type II, sheets of cellulose are separated by low density polyethylene and polypropylene. Since those materials have different thermoforming temperatures, the inner layers were not melted during the process, leading to interlaminar failure around the polypropylene layers. Although there are some differences in the fusion temperature for the polymer components of the Tetra Pak® containers (polypropylene and polyethylene) and these differences can decrease the mechanical anchorage, the same differences can be found for all the configurations studied in this work.

Figure 5 shows the fractured surfaces of Types II and III samples observed through a stereomicroscope. Type II evidenced interlaminar failures (see arrows in Figure 5a) while Type III showed intralaminar failures inside the cellulose layers instead of polypropylene layers (see arrows in Figure 5b).

### Water absorption tests

Table 2 shows the results of water absorption tests for all samples. Chopped material favors water absorption. When the material is cut into small pieces the exposed area increases and so does the absorption of water (Zawadiak et al., 2017). Inks and the polymers (PP and PE) in the samples are hydrophobic and can act as water barriers. On the other hand, the fibers (cellulose) and porous clays boost the water absorption properties (Yilgor et al., 2014).

At the end of the tests (after 24 hours), the water infiltrated the entire transverse area of the samples since the polymer did not completely cover the fibers. Also, for Type III the polymer did not cover all the chopped material, so the water absorption caused by cellulose layers was higher.

![Figure 5. Fractured surfaces of Type II (a) and Type III (b) samples in stereomicroscope. Source: Authors.](image)

<table>
<thead>
<tr>
<th>Type</th>
<th>Material</th>
<th>Water Absorption (%)</th>
<th>Standard Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td></td>
<td>9.96%</td>
<td>0.01%</td>
</tr>
<tr>
<td>II</td>
<td></td>
<td>11.35%</td>
<td>0.04%</td>
</tr>
<tr>
<td>III</td>
<td></td>
<td>14.45%</td>
<td>0.03%</td>
</tr>
</tbody>
</table>

**Source:** Authors.
CONCLUSIONS

The properties of samples obtained from recycled Tetra Pak® were studied and the following results were found:

- The lowest tensile strength was found for Type I sample (9.5 MPa), and the highest was found for Type III sample (37.5 MPa). Thus, when the recycled containers are chopped into small pieces (5 mm x 5 mm) the composites obtained by hot-pressing showed the best mechanical performance.

- Type III sample showed the highest water absorption (around 15% wt.). The water absorption is caused by the hydrophilic condition of cellulose fibers and clay on the containers.

- Two different failure mechanisms with the highest mechanical strength were identified in the samples. Type II showed interlaminar failure between cellulose and polyethylene layers, while Type III samples showed intralaminar failures in cellulose layers.

- Samples obtained using recycled Tetra Pak® containers may be applied for non-structural purposes in the building industry.

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