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Multilayer beverage containers as mechanical reinforcement of polyester resin
Envases de bebidas multicapa como refuerzo mecánico de resina poliéster

C. U. Escobar-Campos [a,*], G. Martínez-Barrera [b], H. A. Barrera-Rodríguez [a]

[a] Posgrado en Ciencia de Materiales, Facultad de Química, Universidad Autónoma del Estado de México, Toluca de Lerdo, CP 50180. México.
[b] Laboratorio de Investigación y Desarrollo de Materiales Avanzados (LIDMA), Facultad de Química, Universidad Autónoma del Estado de México, Km.12 de la carretera Toluca-Atlacomulco, San Cayetano, CP 50200, México.

Abstract

Tetra Pak multilayer containers are used to preserve food for long periods of time and at low cost. Unfortunately, only 30% of the packaging used is recycled, and is not reused in any application, making it highly polluting materials. For these reasons, in this work 1x1 mm particles were obtained from Tetra Pak containers, which were mixed in amounts of 1, 2, 3 and 10% by weight with unsaturated polyester resin (UPR). These composite materials were subjected to tensile, Charpy impact and water absorption tests. The results show that with the addition of Tetra Pak particles, improvements of 50% in deformation, 47% in elastic modulus, 44% in tensile strength and 36% in impact resistance were obtained. The maximum water absorption was 0.17%.

Keywords: Multilayer packaging, Tetra Pak, recycling, waste management, unsaturated polyester resin, mechanical properties.

Resumen

Los envases multicapa Tetra Pak son utilizados para conservar alimentos por largos periodos de tiempo y a bajos costos. Lamentablemente, solo se recicla el 30% de los envases utilizados, además estos no son reutilizados en alguna aplicación, lo que los convierte en materiales altamente contaminantes. Por estas razones, en este trabajo se obtuvieron partículas de 1x1 mm, de envases Tetra Pak, las cuales se mezclaron en cantidades de 1, 2 ,3 y 10% en peso con resina poliéster insaturada (UPR). Estos materiales compuestos fueron sometidos a pruebas de tensión, impacto tipo Charpy y absorción de agua. Los resultados muestran que, con la adición de partículas de Tetra Pak, se obtuvieron mejoras del 50% en la deformación, 47% en el módulo de elasticidad, 44% en la resistencia a la tensión, y 36% en la resistencia al impacto. Mientras que la máxima absorción de agua fue de 0.17%.

Palabras clave: Envases multicapa, Tetra Pak, reciclamiento, manejo de residuos, resina poliéster insaturada, propiedades mecánicas.

1. Introduction

Multilayer packages are in great demand worldwide, due to their effectiveness in food preservation and conservation, with Tetra Pak packages being the most widely used for this purpose (Recupido et al., 2023). Unfortunately, only a small percentage of them are recycled or reused (Muñoz-Batista et al., 2022; Ncube et al., 2021). In Mexico, 46 billion Tetra Pak packages are recycled each year, representing only 30% of consumption. This low recycling rate makes Tetra Pak packages a highly polluting material. Therefore, its carbon footprint must be addressed without delay to incorporate it into a circular economy model (Sleiniute et al., 2024; Guerra-Garcés et al., 2022; Batista et al., 2021; Platnieks et al., 2020).

1.1. *Tetra Pak as a food package*

Tetra Pak packages were developed in 1940 by Swedish engineer Ruben Rausing as a material to solve the food storage problems (Robertson, 2021). It is made from 75% cellulose, 20% low-density polyethylene (LDPE) and 5% aluminium (Bonocore & De Luca, 2022).

The containers have four layers of low density polyethylene (LDPE), as shown in Figure 1, which guarantee total protection of the food and prevent contact between the aluminium and the food. They also contribute to the bonding between the aluminium and the cellulose. The aluminium layer blocks oxygen and light, while the cellulose layer (75% of the package) provides strength and stability. The outer LDPE layer protects the cellulose from external moisture (Georgiopoulou et al., 2021).

*Autor para la correspondencia: cescobarc240@alumno.uaemex.mx
Correo electrónico: cescobarc240@alumno.uaemex.mx (Carlos Uriel Escobar-Campos), gonzomartinez02@yahoo.com.mx (Gonzalo Martínez-Barrera), hectorbarreraiq@gmail.com (Héctor Adrián Barrera-Rodríguez)

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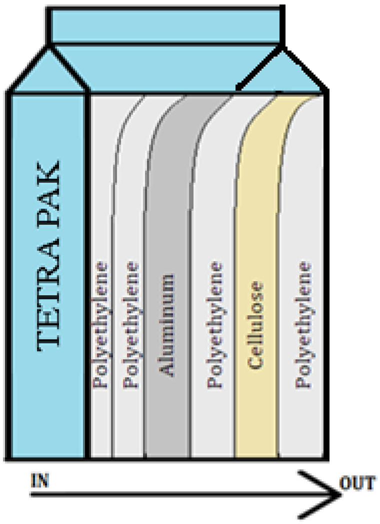

Figure 1. Layers of Tetra Pak packages (Authors´ elaboration).

1.2. Recycling methods for Tetra Pak packages

The recycling of Tetra Pak packages is quite difficult, due to the separation of cellulose, polyethylene and aluminium. Hydropulping is the most commonly used industrial method. However, due to high operational costs, low recycling rates are achieved (Maduwantha & Jayasinghe, 2023; Sahin & Karaboyacı, 2021; Budaycı et al., 2023).

The pyrolysis method is the second most widely used method, which involves thermal degradation of organic matter at high temperatures in the absence of oxygen. It is considered by the recycling industry as an emerging technology for recycling Tetra Pak packages (Yan et al., 2024; Jasim et al., 2020; Cravero & Frache, 2020).

In recent years, the scientific community has been engaged in research on innovative approaches to the use of Tetra Pak packages. Some alternatives to traditional ones have been proposed. For example, recovery cellulose from Tetra Pak packages have been used to produce biofuels, having excellent results (Dave & Reddy, 2023). Also, components of Tetra Pak containers can remove toxic metals in water such as lead, nickel and copper, as these behave as biosorbent materials (Muhammadi et al., 2021). Aluminium nanoparticles (nAl) from Tetra Pak packaging can be used as a low-cost biodiesel additive to improve the performance of internal combustion engines (Marousek, 2022). In studies related to flame retardation, the Tetra Pak performs well, as 95% of it consists of carbonaceous compounds (Matta et al., 2022; Cravero & Frache, 2020).

1.3. Components of Tetra Pak containers used to produce composite materials

Composite materials are produced with a matrix and aggregates. The matrix mainly provides the mechanical properties and serves as a binder for the aggregates (Kangishwar et al., 2023). Composite materials, produced with a thermoset polymer matrix, are becoming exceptional due to their properties, simple processing and diverse applications such as aerospace, automotive, infrastructure and construction (Sharma et al., 2022; Alhijazi et al., 2020; Rajak et al., 2022).

Polymer composites can be reinforced with synthetic fibres for industrial applications (Qiao et al., 2023). These include glass, carbon, silica carbide and aramid fibres. However, natural fibres, such as jute, wool, sisal, silk, flax, hemp and loofah, can also be used as reinforcing materials (Seydibeyoglu et al., 2023; Elfaleh et al., 2023).

A third type of reinforcement belongs to waste materials that are becoming increasingly important in composite materials, e.g., Tetra Pak waste packages (Pawluczuk et al., 2022; Li et al., 2022; Shiferaw et al., 2023).

For the manufacture of sheets of 250x120x7 mm, Tetra Pak waste containers have been used. Three Tetra Pak particle sizes were used, 4.6x11.85 mm, 10x15 mm and 5x5 mm. The results show that the highest tensile strength values (37.4 MPa) were obtained with 5x5 mm Tetra Pak particles, followed by those sheets produced with 10x15 mm particles (Macías-Gallego et al., 2020).

In another study, residual Tetra Pak particles were incorporated into the core layer of a board as a partial wood substitute. The results show a value of 15.67 MPa for flexural strength and 3.2 GPa for elastic modulus, for composites with 25% Tetra Pak particles (Auriga et al., 2021).

In accordance with the aforementioned findings, this study aims to investigate the effects of Tetra Pak packaging waste as filler in unsaturated polyester resin, through measurements on tensile and impact strength, as well as contribute to reduce its environmental impact, and facilitate its integration into a circular economy model for materials science.

1.3.1. Materials and methods

1.3.2. Materials

The unsaturated polyester resin (UPR) was supplied by Polynt Composites Mexico S.A. de C.V. (Atlacomulco México) and marketed under the name Polylite® 32335-10. Methyl ethyl ketone peroxide (MEKP) was used to polymerization of resin, which was added at a rate of 2g per 100g of resin, according to the resin manufacturer.

The Tetra Pak beverage cartons were subjected to a thorough water cleaning process, including the removal of any residual impurities. The cleaned cartons were then dried for 24 hours at room temperature. Finally, they were cut in an average size of 1x1x0.6 mm using scissors (Figure 2) which is a method that has proven to be more efficient than milling, as it does not produce plastic deformations.

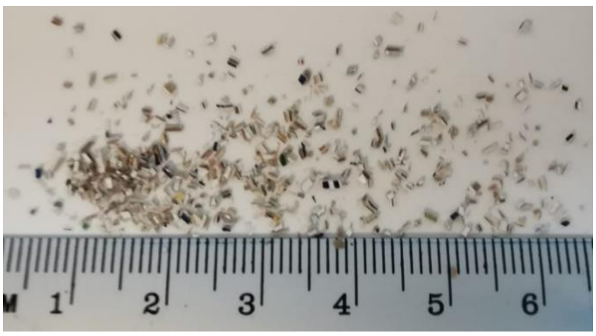

Figure 2. Tetra Pak particles cut with scissors (Authors´ elaboration).

1.3.3. Production of the resin/Tetra Pak composites

The composites were produced with unsaturated polyester resin (UPR) as matrix and Tetra Pak particles as filler. In Table 1 the composition and the density of each specimen is shown. Six specimens for each formulation were produced.

Table 1: Formulations of the UPR/Tetra Pak composites and their densities.

Specimen (code)	UPR	Tetra Pak	Density
	% wt		kg/m^3
R	100	0	1052.4
R/TP-1	99	1	1086.0
R/TP-2	98	2	1119.1
R/TP-3	97	3	1198.7
R/TP-10	90	10	1225.3

The dimensions of the specimens for tensile tests were 165×20×4 mm, as illustrated in figure 3 whilst Figure 4 shows three specimens for each formulation.

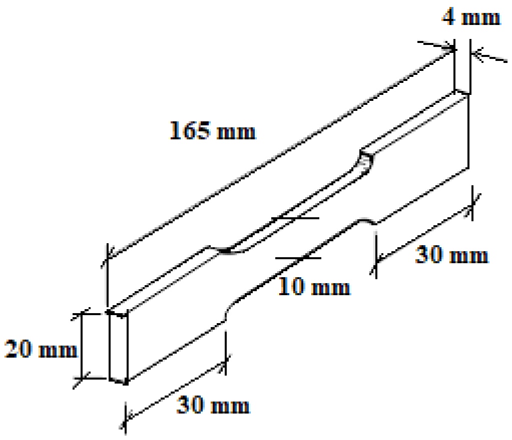

Figure 3. Dimensions of the specimens for tensile test. (Authors´ elaboration).

2. Experimental Procedures

2.1. Morphology of Tetra Pak particles

The surfaces of the specimens tested were observed using a JEOL model NeoScope JCM-6000 scanning electron microscope (SEM) equipped with a tungsten filament, 1.0 µm resolution and operated at 5 kV acceleration. Images were taken in the secondary electron mode.

2.2. Mechanical tests

Tensile tests were performed in an Instron universal testing machine model 3382 equipped with a 100 kN load cell at a rate of 0.1 mm/min between clamps in accordance with ASTM D638.

Charpy impact testing was performed on an XJ-50Z series testing machine; that use a standardized test piece subjected to the action of a 1 Joule pendulum hammer placed at 158°, according to the specifications of the ISO 179 standard.

Both mechanical tests were conducted in room temperature, about 22 ± 3 °C.

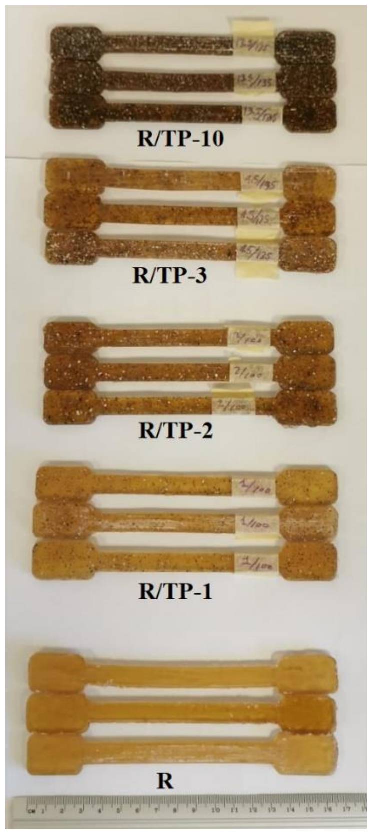

Figure 4. Specimens for tensile tests. (Authors´ elaboration).

2.2.1. Production of composites for impact test

Same formulations from table 1 were used for impact test, nothing changed but specimens´ dimensions 80x10x4 mm, which are illustrated in figure 5. Six specimens for each formulation were produced.

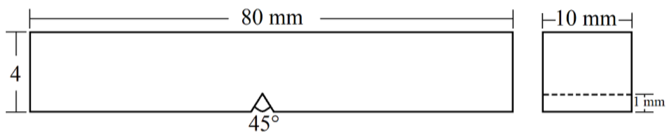

Figure 5. Dimensions of the specimens for impact test. (Authors´ elaboration).

3.3 Water absorption

The water absorption test was performed to evaluate the ability of the UPR/Tetra Pak composite to absorb water at a given time. As is known, Tetra Pak particles are composed of 75% cellulose, a hydrophilic material. Initially, the composites were exposed in an oven at 50°C for 24 h. They were then weighed and immersed in distilled water at 25°C for 24 h. Finally, the specimens were removed from the water and weighed.

The percentage of water absorbed was calculated using equation (1).

$$\%\, Water\ absorption = \frac{wet\ weight - dry\ weight}{dry\ weight} \text{ x } 100 \qquad (1)$$

3.4 Density

Composites' densities were calculated using ISO 1183-1 standard following the equation (2). Results in kg/m^3 units are presented in table 1.

$$Composite\acute{}s\ density = \frac{weight}{displaced\ volume}\ [=]\ \frac{kg}{m^3} \quad (2)$$

3. Results and discussion

4.1 Surface morphology of Tetra Pak particles

Figure 6 shows SEM images of Tetra Pak particles seen from the inner side (Figure 6a) where is continuous and smooth, or seen from the outer side (Figure 6b), where a roughness surface is observed.

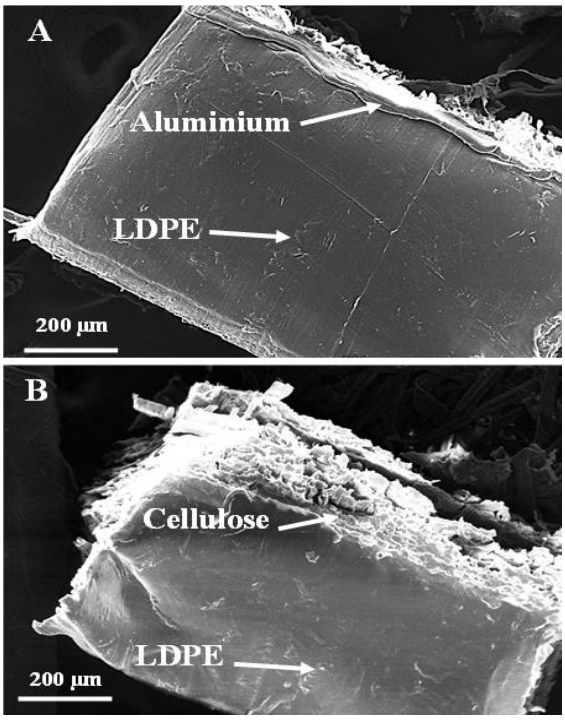

Figure 6. SEM images of the Tetra Pak particles: from the inner side (a) and from the outer side (b). (Authors´ elaboration).

4.2 Tensile strength

As illustrated in Figure 7 for the results in tensile strength, a remarkable improvement is obtained for composites with 1% Tetra Pak. They have a value of 2.30 MPa, which means an improvement of 42% with respect to the value for pure resin, R. This improvement can be attributed to the optimal stresses transfer between the Tetra Pak particles and resin. For composites with 2% Tetra Pak particles, the values are 5% higher. While, for higher concentrations of Tetra Pak particles the values are similar to those for pure resin. Therefore, higher concentrations of Tetra Pak particles produce agglomeration in the composites and the stresses transfer is lower, so the strength decreases as R/TP-3 and R/TP-10 results show, 0.80 and 0.88 MPa, respectively.

4.3 Strain at yield point

The results of the strain at yield point are shown in Figure 8. The Tetra Pak particles successfully improved the deformation of the composite materials. The addition of 1% Tetra Pak produces an improvement of 50%, showing a value of 0.009 mm/mm. Likewise, 2% and 3% Tetra Pak produce 16% of improvement. Thus, the components of Tetra Pak, i.e. cellulose, LDPE and aluminum, act as elastic materials during the application of tensile stresses. It is only necessary to add 1% Tetra Pak particles to produce the greatest deformation. However, at higher concentrations of Tetra Pak particles, agglomeration occurs, allowing stress dissipation and thus early fracture.

4.4 Modulus of elasticity

Figure 9 illustrates the results for modulus of elasticity. It can be observed the highest values for composites with 1% Tetra Pak particles, namely 57.43 MPa, which is 60 % higher than that for pure resin. Thus, the addition of 1% Tetra Pak particles produces the most optimal stress transfer with the resin. Composites with 2% and 3% Tetra Pak particles showed low values with respect to pure resin. Although, the addition of 10% Tetra Pak produced and improvement of 10%.

4.5 Impact strength

Figure 10 shows the result of the impact tests. For composites with 1% and 2% Tetra Pak particles the values decrease up to 41% with respect to the value of 1.7 kJ/m^2 for pure resin. However, with the addition of 3% Tetra Pak the value is 36% higher than that for pure resin. For higher concentrations of Tetra Pak particles, the values decrease. Thus, 3% Tetra Pak was optimal for absorbing the higher impact stresses. However, a higher concentration of Tetra Pak particles can promote crack formation and, consequently, decrease impacts.

4.6 Water absorption

Figure 11 illustrates the results of the water absorption experiment. The resin presents a value of 0.09%, which is lower than that reported by the manufacturer, namely 0.14% (Reichhold México, 2020). As is known, the values are low due to the hydrophobic nature of the polyester resin. In the experiment, the values increase as the concentration of Tetra Pak increases. The highest value is 0.17% when 10% Tetra Pak particles is added, which means an increase of 88%. These increases are due to the presence of the 75% cellulose contained in the Tetra Pak containers.

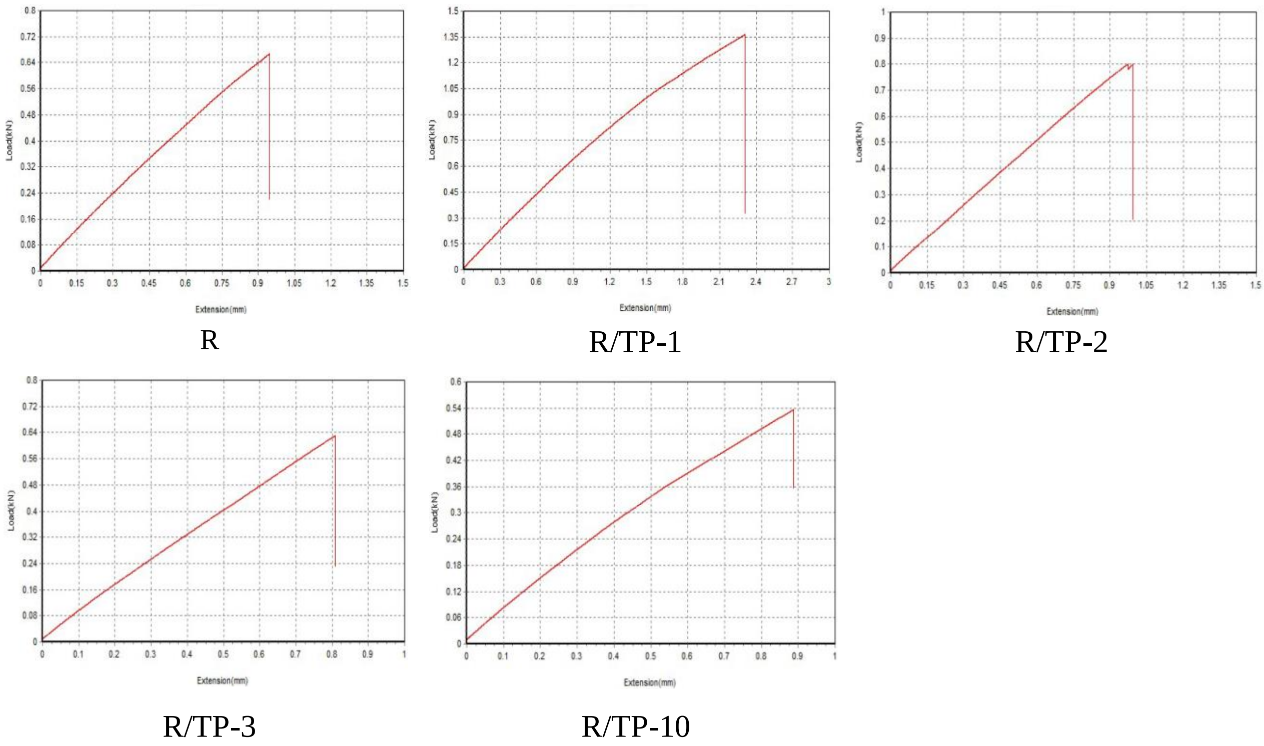

Figure 7. Stress vs strain curves of the UPR/Tetra Pak composites.

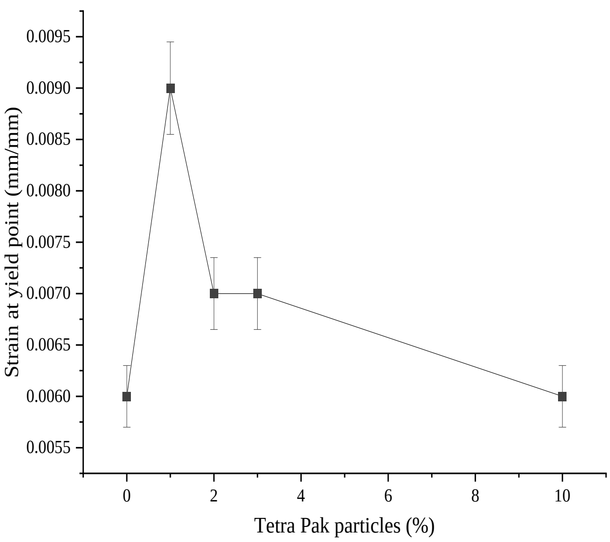

Figure 8. Strain at yield point of the UPR/Tetra Pak composites.

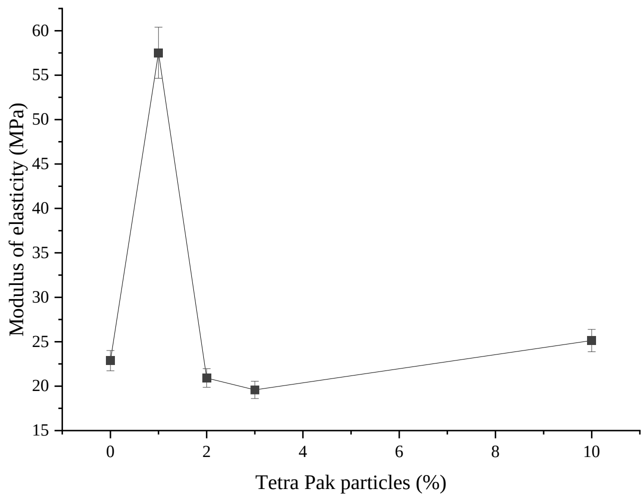

Figure 9. Modulus of elasticity of the UPR/Tetra Pak composites.

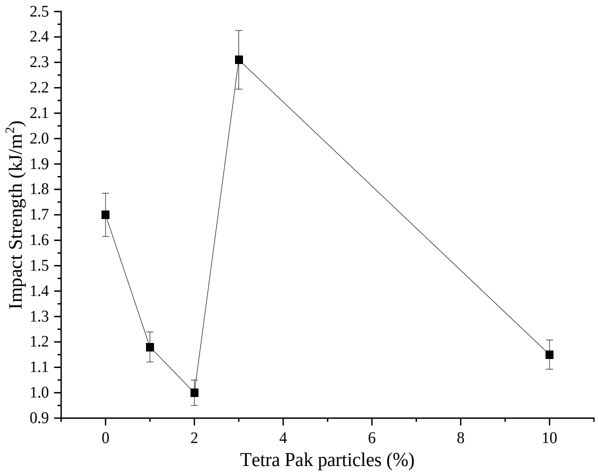

Figure 10. Impact strength of the UPR/Tetra Pak composites.

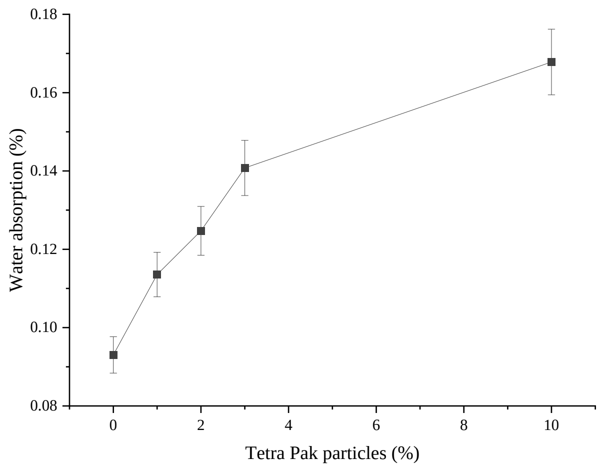

Figure 11. Water absorption of the UPR/Tetra Pak composites.

5 Conclusions

The results of the tensile tests shown that the addition of 1% Tetra Pak particles is optimal for the stresses transfer between polyester resin and Tetra Pak particles, which can ensure the maximal improvements, namely 42% in resistance, 50% in deformation and 60% in the modulus of elasticity. However, higher concentrations of Tetra Pak particles produce lees stress transfer resulting in a decrease in strength and ductility. Impact tests revealed that the highest values were obtained with the addition of 3% Tetra Pak particles, resulting in a 36% improvement. Thus, this concentration is enough to ensure high impact resistance.

Water absorption increases by 88% with the addition of Tetra Pak particles, compared to pure resin value, likely due to the high cellulose concentration in Tetra Pak containers (75%). Moreover, this suggests potential applications for the composites in humid environments.

This paper has demonstrated that adding a small concentration of Tetra Pak particles can achieve valuable improvements in mechanical properties and produce more hydrophilic composites.

Abbreviations

LDPE: Low Density Polyethylene
MEKP: Methyl Ethyl Ketone Peroxide
UPR: Unsaturated Polyester Resin

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