ARTICLE IN PRESS

Fuel xxx (2014) xxx-xxx



Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel



Development of a process using waste vegetable oil for separation of aluminum and polyethylene from Tetra Pak

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HIGHLIGHTS

- The polyethylene and the aluminum were separated from Tetra Pak.
- Waste vegetable oil was used in the separation process.
- Different solvents were used to wash the polyethylene.

ARTICLE INFO

Article history: Received 15 March 2014 Received in revised form 5 September 2014 Accepted 10 September 2014 Available online xxxx

Keywords: Tetra Pak Waste vegetable oil Aluminum Polyethylene Separation process

ABSTRACT

Tetra Paks are multi-layer polycoated paperboards, which are widely used as aseptic packages for beverages. This packaging system allows distribution of products without refrigeration for long periods of time (Haydary et al., 2013). Owing to their widespread use, Tetra Paks contribute to significant part of municipal solid waste (MWS), and it is estimated that in Mexico, each person generates Tetra Pak waste of about 800 g/day (SEMARNAT, 2010). Thus, development of a separation or recycling process is currently important, especially for Tetra Pak packaging, owing to the enormous amount of Tetra Pak waste produced every day. In the present study, a process for the separation of aluminum and polyethylene from Tetra Pak was developed. The method was realized with used waste vegetable oil, which was previously utilized for cooking. The main purpose of this process was to generate products at low cost by consuming less energy and source materials. The separation process comprised eight stages. The polyethylene was washed with different solvents such as ethanol, chloroform, and isopropyl alcohol, while the aluminum was washed with ethanol. The obtained low-density polyethylene was characterized by IF-IR, thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC). The percentage of low-density polyethylene in the final sample obtained after washing with isopropyl alcohol, chloroform, and ethanol was 73%, 68%, and 52%, respectively, and that of aluminum in the final sample obtained after washing with ethanol was 85%. Lastly, the results were compared in terms of the percentage of polymer content in the mixture used to produce pellets of poly-aluminum via extrusion in the industry. The findings revealed that the separation process proposed in this study could be an effective technology to recover aluminum and polyethylene from Tetra Pak.

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1. Introduction

Organic waste production has continuously grown in the last decade. Nowadays, one of the major challenges faced by the modern world is effective utilization of organic waste, including plastics, tires, and municipal solid wastes [1]. There is a necessity not only to recycle wastes, but also to reduce their volume [3]. Municipal solid waste (MSW) is composed of different materials such as organic

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http://dx.doi.org/10.1016/j.fuel.2014.09.032 0016-2361/© 2014 Elsevier Ltd. All rights reserved. matter, paper and cardboard, plastic, glass, and aluminum and other metals [2].

Aseptic packaging, produced by Tetra Pak and used for milk packing after ultrapasteurization, is composed of three materials organized into six layers, namely, paper (75 wt.%), low-density polyethylene (LDPE, 20 wt.%), and aluminum (5 wt.%) [4]. Recycling is one of the alternatives for recovering this packaging. The recycling process starts in the paper industry where recyclable cellulose can be recovered by the repulping process. The separation of cellulose takes place inside a washing device by means of a perforated sieve that has two compartments. In the inner compartment, polyethylene with aluminum is retained, and in the outer compartment, the paper pulp is retained. In the hydropulping process, thin layers of plastic and aluminum are separated from cellulosic fibers by centrifugal forces in a hydropulper and high-quality fibers are used in the production of paper products such as tissues, paper towels, and fine writing paper [4b,5], while the agglutinate of both the materials (LDPE and aluminum) are extruded to obtain pellets [4b]. Alternatively, polyethylene and aluminum can be separated in three different ways with energy generation from paraffinic oil: recovery of aluminum in pyrolysis ovens, recovery of polyethylene and aluminum by plasma technology, and processing of the mixtures of polyethylene and aluminum to obtain high-end plastic lumber products [6].

Pyrolysis of Tetra Pak is performed in an inert atmosphere in ovens at temperatures ranging from 350 °C [7] to 500 °C, and the products obtained are aluminum, gas, oil, and char. Both gas and oil can be used as fuels and the carbonaceous wastes can be used as activated carbon in some cases [7]. Plasma technology is employed for the separation of polyethylene and aluminum with a plasma jet at 15,000 °C for heating the layers of polyethylene and aluminum together. The plasma is generated by electric current, which produces paraffin from polyethylene and high-purity aluminum (Alcoa Company).

However, according to Tetra Pak International, in many countries, there is still a lack of infrastructure for managing the household wastes generated, which are disposed of through landfill. The global recycling of used Tetra Pak cartons has increased by 10% in 2012, from 528 to 581 kilo tonnes, which is 22.9% of the total [3]. It is important to boost the production of recyclable materials for use in new production processes or for the generation of new products along with the creation of economic activities, new companies, income, and formal employments, which Mexico needs [2,8]. However, inadequate management of residues can pose significant risks to public health and ecosystem [9]. The objective of the present study was to recover aluminum and polyethylene from Tetra Pak packaging using a new separation process, which can also be considered as sustainable with a low cost of operation, using the least possible amount of raw materials and energy.

2. Experiment

2.1. Materials

The Tetra Pak waste sample without cellulose (TP) was obtained from a pretreatment, and its theoretical composition is shown in Table 1. The sample was ground to an indefinite particle size (see Fig. 1). Different samples of polyethylene, namely, the commercial-grade LDPE in Pellet (LDPE commercial degree; average molecular weight (Mw) = 2.0×10^5 , density = 0.92 g/cm³, and melting point = 110-115 °C; sample provided by Industria Cangrejera de Mexico, Coatzacoalcos Veracruz), industrial poly-aluminum (Pellet); provided by the Transforma Company dedicated to the collection of Tetra Pak and separation of cellulose from these containers), and films of polyethylene (Layer TP; obtained from Tetra Pak packages) were examined. The samples were characterized by methods such as Fourier transform infrared (FTIR) analysis.

Table 1 Compositions of different samples (wt.%).

Samples/materials	Cellulose	Polyethylene	Aluminum
Tetra Pak ^a	75	20	5
Tetra Pak w/o cellulose	0	83	17
Pellet ^b	0	95°	5

- ^a Tetra Pak Company.
- ^b Transforma Company.
- ^c Mixture of LDPE, HDPE, and polypropylene (see Fig. 3; Pellet).

The waste vegetable oil of canola (Oil; density = 0.92 g/ml; viscosity = 78 cst; smoke point = $285 \,^{\circ}\text{C}$) was collected from establishments where fried foods such as potatoes and fast food are cooked. Different solvents such as ethyl alcohol, chloroform, and isopropyl alcohol were used to wash the polyethylene and aluminum obtained.

2.2. Separation process

In a 250-ml beaker, 100 g of waste vegetable oil (Oil) and 10 g of ground TP were added at $140\,^{\circ}\text{C}$ for 1 h with stirring for 50 min. Owing to the difference in the density, the solution was separated into two parts; the supernatant of the solution was oil with polyethylene and aluminum precipitated with less oil and polyethylene at the bottom of the vessel. The solution of oil with polyethylene was filtered and the oil was reused for second and third heat treatments.

The aluminum precipitated with less oil and polyethylene remained in the vessel under the same conditions (140 °C for 1 h with stirring for 50 min), and after polyethylene and oil were separated and filtered, the polyethylene was washed with three different solvents, namely, 50 ml of ethyl alcohol (99.8% Fermont), 50 ml of chloroform (99.8% Fermont), and 50 ml of isopropyl alcohol, respectively, and boiled for 1 h. The obtained aluminum was washed with 50 ml of ethyl alcohol and 50 ml of isopropyl alcohol and boiled for 1 h. After washing the products, the aluminum and polyethylene were filtered. The products obtained were analyzed by FTIR, thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC), and the general process is shown in Fig. 2.

2.3. Methods of analysis

TGA and DSC were conducted with Thermal analyzer Netzsch STA 449 F3 Jupiter and Microbalance Mettler Toledo XP6, respectively, and Savitzky-Golay smoothing algorithm was employed for TGA curves. With a heating ramp of 10 °C/min, the LDPE, Oil, LDPE-CHCl₃, LDPE-IPOH, and LDPE-EtOH samples were heated from room temperature until 560 °C; the TP, Pellet, and aluminum samples were heated to 800 °C in a nitrogen atmosphere at a flow rate of 20 ml/min; aluminum crucibles of 5-mm diameter were heated to 560 °C; and alumina crucibles of 6-mm diameter were heated to 800 °C. The FTIR spectra of the products were obtained using a spectrophotometer (Tensor 27, Bruker with accessories Platinum ATR) to qualitatively identify the chemical functionality of the products.

3. Results

3.1. Interactions between LDPE and Oil

Waste vegetable oil is used as a non-polar solvent [10], which penetrates the polyethylene molecules that cause inflammation and begins to form a gel. After the formation of a solution, the macromolecules of the polymer chains begin to separate and solubilize in the solvent. In the solution, certain interactions [11] (dispersion forces, electrostatic forces, and similar functional groups) that are equal to or greater than the interactions between the molecules in each component occur. The crystallinity of the polymers causes a decrease in the free energy required to overcome the dissolution process. Most of these processes are endothermic; therefore, increased temperature promotes the solubility of linear polymers. The interaction between oil and polyethylene increases when the temperature increases. As a result, the molecules of the same compound will tend to aggregate or agglomerate, thus indicating a high tendency for the separation of LDPE at higher temperature and in the presence of solvent (Oil).

J.E. Rodríguez-Gómez et al./Fuel xxx (2014) xxx-xxx



Fig. 1. (a) TP, (b) Layer TP, (c) LDPE, and (d) Pellet.

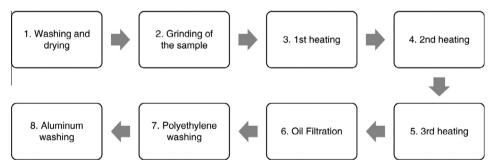


Fig. 2. Steps of the separation process.

3.2. TGA and DSC analysis of TP and Pellet

The DSC and TGA were performed to determine the thermal stability and polyethylene content of the different products obtained and to establish a comparison with a reference product and commercial product. The LDPE and Oil contents were analyzed by TGA, while the heat energy involved in the chemical and physical processes was evaluated by DSC. As shown in Fig. 3, three main endothermic processes were noted in two samples, namely, TP and Pellet. These two samples showed an endothermic process at 106 °C (100-110 °C) corresponding to fusion of LDPE. The Pellet sample presented a second peak at 110 °C, which indicated another endothermic process caused by industrial treatment when the Tetra Pak packaging is mixed with pieces of polypropylene during the industrial separation process. The third peak at 480 °C showed the decomposition of LDPE in the TP. Finally, the last peak at 655 °C was related to an endothermic process of fusion of aluminum in the Pellet sample.

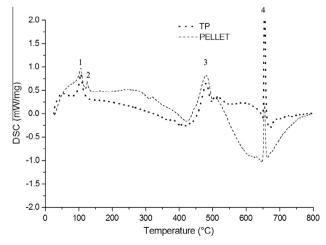


Fig. 3. Results of DSC analysis for TP and Pellet.

Please cite this article in press as: Rodríguez-Gómez JE et al. Development of a process using waste vegetable oil for separation of aluminum and polyethylene from Tetra Pak. Fuel (2014), http://dx.doi.org/10.1016/j.fuel.2014.09.032

J.E. Rodríguez-Gómez et al./Fuel xxx (2014) xxx-xxx

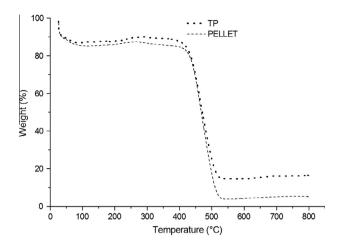


Fig. 4. TGA results for TP and Pellet.

Fig. 4 shows the TGA results for TP and Pellet samples. The Pellet sample showed a higher percentage of polyethylene, when compared with the TP sample. The percentage of polyethylene in the Pellet and TP samples was 94% and 82%, respectively, which was confirmed by a higher polyethylene decomposition heat in the DSC analysis (Table S1; SI). The initial mass loss was caused by the loss of moisture in the TP and Pellet samples, because the samples were not previously dried. This moisture represented 11% and 15% of the total mass of the TP and Pellet samples, respectively. Thus, following the loss of moisture, the percentage of polyethylene in the TP and Pellet samples was 82% and 94%, respectively. These findings indicated that the experimental results

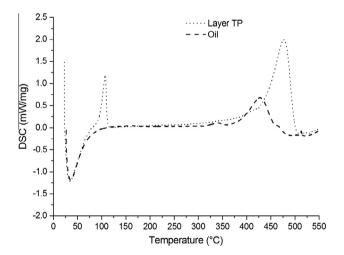


Fig. 5. DSC curves for Layer TP and Oil.

were in agreement with the hypothetical calculation, which suggested 80% polyethylene content in the TP sample.

3.3. DSC analysis of Layer TP and Oil

DSC analysis was performed at 25–560 °C for Layer TP and Oil, and the results obtained are presented in Fig. 5. Three peaks, including two endothermic peaks for Layer TP and one for Oil, were noted. As discussed earlier, the first peak for Layer TP indicated the fusion of LDPE (107 °C), the second peak for Layer TP suggested the decomposition of LDPE (475 °C), and the last peak for Oil denoted the decomposition of Oil at a temperature lower than that for Layer TP.

3.4. DSC analysis of LDPE washed with different solvents

DSC analysis was performed at 25–560 °C for LDPE obtained by the separation process. The sample was washed with ethanol (LDPE-EtOH) (Fig. 6a), chloroform (LDPE-CHCl₃) (Fig. 6b), and isopropyl alcohol (LDPE-IPOH) (Fig. 6c), respectively, and the results obtained are presented in Fig. 7. The three curves showed three main peaks: peaks 1 and 2 were related to LDPE (105, 481 °C for LDPE-IPOH; 107, 482 °C for LDPE-EtOH; and 102, 477 °C for LDPE-CHCl₃) and peak 3 was correlated to the decomposition of Oil. TGA revealed that the percentage of LDPE in the LDPE-IPOH, LDPE-CHCl₃, and LDPE-EtOH samples was approximately 73%, 68%, and 52%, respectively, with the Oil forming the remaining part of the samples. The decomposition of vegetable oil decreased with the decreasing percentage of the Oil in the samples (Table S2; SI).

3.5. DSC and TGA of aluminum obtained from the separation process

Fig. 8 shows the loss of mass and consumption of energy (mW/mg), with an endothermic peak at 656 °C indicating a fusion process. The loss of mass (15%) was owing to the fact that aluminum was not dry, and the loss of mass at 25–80 °C corresponded to the loss of moisture and residual solvent. The aluminum obtained after washing with ethanol is presented in Fig. 9.

3.6. Overall yield of the separation process

Based on the previous results, the overall yield of the separation process was determined. The calculations were performed based on the initial material used and the final products obtained, and the amount of LDPE and aluminum was quantized. The overall yield was ascertained based on the amount of LDPE obtained from the process of washing with isopropyl alcohol owing to the high percentage of recovery.

After grinding and washing, 10 g of the TP sample (83% polyethylene and 17% aluminum) were subjected to three heat treatments, filtration, and finally, washing. At the end of the process, two products were obtained, namely, 7.75 g of polyethylene

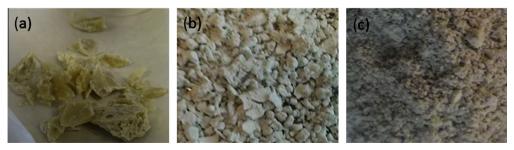


Fig. 6. LDPE washed with (a) ethanol, (b) chloroform, and (c) isopropyl alcohol.

J.E. Rodríguez-Gómez et al./Fuel xxx (2014) xxx-xxx

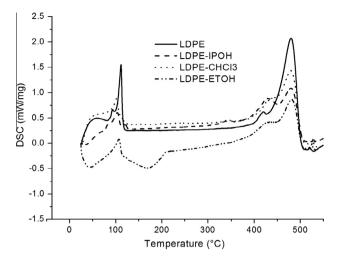


Fig. 7. DSC curves for LDPE samples washed with different solvents.

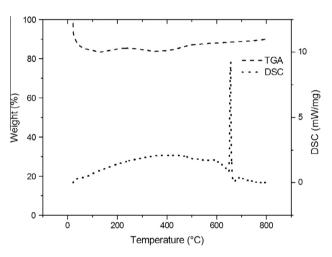


Fig. 8. DSC and TGA curves for the aluminum separation process.



Fig. 9. Aluminum obtained from the separation process.

(91% polyethylene and 9% impurities) and 1.51 g of aluminum (90% aluminum and 10% solvent). The compositions of the products were determined by TGA and DSC analysis. From the results obtained regarding the composition, the recovery rate of polyethylene and aluminum was 85% and 80%, respectively, and some losses resulting from washing with different solvents and filtration were observed.

3.7. FTIR analysis

The sample obtained after washing was subjected to FTIR analysis to confirm the presence of functional groups in LDPE and Oil. The signals for PE-EtOH and PE-CHCl₃ at 2920 cm⁻¹ (CH₂ υ_{as}), 2850 cm⁻¹ (CH₂ υ_{s}), 1470 cm⁻¹ (CH₂ scissor), and 720 cm⁻¹ ([CH₂]_n rock) confirmed the presence of polyethylene (Figs. S1, S2, S3; SI).

4. Conclusions

In the present study, a separation process using two common residues (TP and Oil) was developed to separate and recover polyethylene and aluminum. The percentage of LDPE and aluminum separated from the TP sample was 85% and 80%, respectively. Isopropyl alcohol was observed to be the best solvent to remove LDPE, when compared with other solvents. The developed process could be further improved to obtain products with higher purity by removing the oil, which could be achieved by increasing the washing time and amount of solvent used. Nevertheless, in future, the quality of the products obtained needs to be determined for any possible applications, such as foil for food industry or tools with aluminum; containers, bags, toys, and furniture with polymer; or mixture with commercial polyethylene. Lastly, the products obtained will be subjected to mechanical tests in further studies.

Acknowledgments

The authors wish to acknowledge the support given by the Centro Conjunto de Investigación en Química Sustentable, UAEM-UNAM, financial support from UAEM project 3411/2013M, and help from the undergraduate student, Martin Cabral.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.fuel.2014.09.032.

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