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BUT
Research Note

Recycling of Multilayer Packaging Foils by Using Diferent Organic Solvent

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P A P E R I N F O

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A B S T R A C T

Multilayer Packaging wastes are one of the major problem in the world. About 6 billion tons of packages are generated per year in the world (in EU is about 82 million tons per year). Multilayer packaging foils are approximately 17% of all produced packaging films. Most produced multilayer film is based on different polymers, such as: polyethylene terephthalate (PET), polypropylene (PP), and polyethylene (PE) as main components, and an aluminum layer. However, because of their poor recyclability, most multi layers are usually incinerated or landfill, this is counteracting the efforts towards a circular economy. There are different recycling methods for this type of waste, but the main problem is that there is less company which recycling multilayer packaging foils. Also, it is very important that, mostly they recovering only one or two components and others are burned or landfilled. The aim of this research was to find the most suitable solvent for recycling multilayer packaging foils and recover two common components: polymer and Al foil. For the first experiment, there was chosen multilayer packaging foil's different samples, such as: packaging for chocolate, chips, medicines and coffee. There was used several chemicals: acetic acid, acetone, dimethyl formamide, ethanol, ethyl acetate and toluene. These solvents are on the list of green chemicals and they were selected, due to their less impact on environment and human health. The recycling process and result was different for each samples and solvents, because of their individual characteristics.

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INTRODUCTION

Packaging wastes are significant part of municipal solid waste which has caused increasing environmental concerns, resulting in strengthening of EU Regulations in order to reduce amounts of packaging wastes [1,2]. Accounting for the fastest growing segments of the packaging industry, flexible packaging provides an economical method to package, preserve and distribute food, beverages, other consumables, pharmaceuticals and other products that need extended shelf life. These type of packaging are made up of multiple layers of plastic made from different monomers along with a metallic layer which is commercially known as Tetra Pak. They are typically included one or more adhesive layers and printing layers. Compared to multi-materials using different types of plastic only, the metallic layer offers additional protection against moisture, air, odors, and UV light [3,4]. There is no proper closed-loop system put in place to handle the recycling of flexible packaging, especially multi-layer high-barrier materials, specifically structures composed of aluminum foil within a polyethylene (PE) laminate. Mono-layered packaging is more easily recyclable because it doesn't contain the aluminum foil center. But the process to recycle multi-layered flexible PE packaging involves more steps than the traditional plastic recycling regimen by including separation of each layer. Each layer is then analyzed, identified and recycled individually. Because of that these types of packaging are used for food, another hurdle for recycling is food contamination. Due to food contamination in most flexible packaging going through the current recycling system, a very small percentage is actually reusable [5]. Because of this type of packaging waste is typical and at the same time problematic, various companies and researchers are working on resolving this problem; For example: Enval have developed a process where mixing shredded waste is mixed with carbon, a highly microwave-absorbent material. The energy from the microwaves is transferred to the waste by thermal conduction from the carbon, providing both a very efficient energy transfer mechanism and a highly reducing chemical environment. The process recovers 100% of the aluminum present in the laminate clean and ready to recycle, and produces oils and gases suitable for fuel for steam/electricity generation or for use as chemical feedstock in other processes [6]. Beigbeder et al. [7] have developed the "Fine Sort" platform which targets

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streams where the flakes are between 4-20mm in size. Once the layers of different materials are separated from each other, a method is required to sort the different material flakes e.g. PE, PET, aluminum foil etc. from each other. To be valuable as a secondary material, the resulting sorted streams needs to be as "pure" as possible and, ideally, sorted into fractions with similar properties [7]. Urban Mining Corp's new magnetic density separation technologies [8] create an apparent density range inside a ferromagnetic fluid in order to create multiple density fractions in a single process. The process fluid consists of a dilute mixture of water and ferrous oxide and is brought in a magnetic field. When such liquid is placed above a magnet, it is affected by the magnetic field and by the gravitational field [8]. Rodríguez-Gómez et al. [9] used waste vegetable oil for separation of aluminum and polyethylene from Tetra Pak. The main purpose of this process was to generate products at low cost by consuming less energy and source materials [9]. Kulkarni et al. [10] have used an innovative and environment-friendly sub- and super-critical water for successful recovery of aluminum from composite laminated wastes. Favaron et al. [11] used supercritical ethanol for PET and aluminum recycling from multilayer food packaging. There is a brief study about the separation process by dissolving PE–aluminum (PE-Al) composites into a series of organic solvents with a combination of time and temperature. Cervantes-Reyes et al. [12] have introduced non-polar solvents as more efficient in the recovery process of PE films from PE-Al composites using a polymer dissolving as key step in the recovery strategy. This efficiency can reach to 56% under xylene reflux conditions. The recovered PE films presented good thermal properties which indicate high purity [12].

EXPERIMENTAL WORK

Wastewater was taken from paper mill producing several types of paper. The characteristics of the wastewater are presented in Table 1. 10% NaOH solution and 0.1% cationic polyelectrolyte (PE) solution as flocculants were used to increase wastewater pH and to agglomerate fine flocks in flocculation processes, respectively. All chemicals used in this study were technical reagent grade.

Equipment and chemicals

Magnetic stirring and distillatory bench scale were used as experimental set up. All chemicals such as; acetic acid, acetone, dimethyl formamide, ethanol, ethyl acetate and toluene were analytical grade supplied by local companies.

Sample preparation

Five types of multilayer packaging foils were selected for experimental research-packaging for chips and chocolate, coffee packages, pharmaceuticals blister's primary and secondary packaging. According to scientific literature sources, aluminum content in the pharmaceutical blister package is from 15 to 20% of the weight [13-17]. Here are examples for Aluminum and polymer content in several multilayer packages foil (see Figure 1).

According to the literature, for aluminum recovering from aluminum alloy processing, wet process separation method has been selected, for which, we used six different solvents: acetic acid, acetone, dimethyl formamide, ethanol, ethyl acetate and toluene. In fact, solvents were selected, based on less adverse effects on environmental and human health. Before any experiment, the samples were washed in distilled water in order to remove the contaminated particles and cut into small pieces [13]. We determined, the differences of sample mass before and after treatment. Here is example, for pharmaceutical blisters samples mass before and after treatment by ethyl acetate (see Table 1).

Also, it was determined volume of solvent used; here is example, volume of ethyl acetate, before and after experiments and after distillation (see Table 2).

After experimentation, in order to reduce waste and reduce uses of a new material, the used solvent was distillated and recovered by simply distillation method [14]. We collected together and distillated all used ethyl acetate, in order to reduce time and energy. This was simply and favorable method for laboratory experimentation. (see Figure 2).

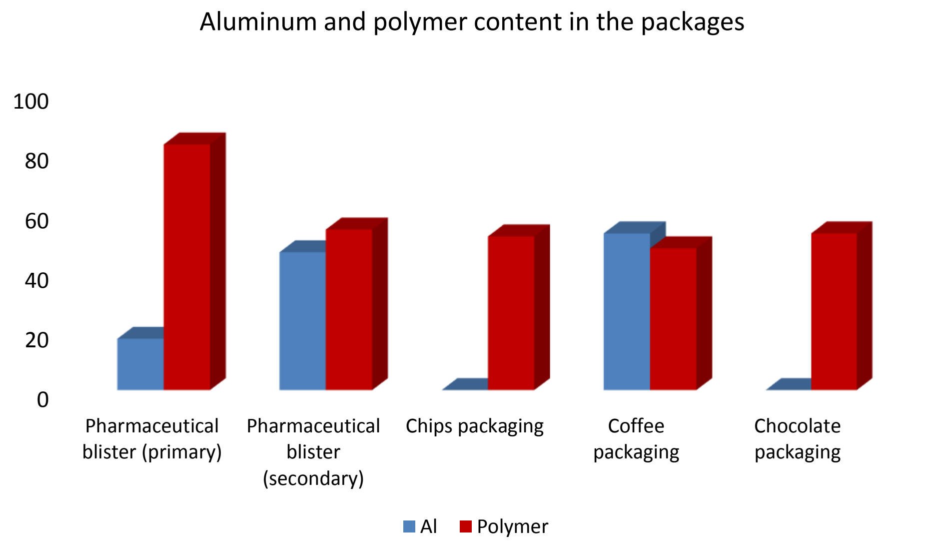

Figure 1. Aluminum and polymer content in the packages

TABLE 1. Pharmaceutical blister (primary) packaging sample mass before and after by treatment by Ethyl acetate

Samples	Sample weigh before experiment (g)	Sample weigh after experiment (g)	
		Al	Polymer
Pharmaceutical blister (primary)	10	1.72	8.28
Pharmaceutical blister (secondary)	10	4.62	5.38
Chips packaging	2	-	≈1.03
Coffee packaging	2	1.05	0.95
Chocolate packaging	2	-	≈1.05

TABLE 2. Ethyl acetate volume before, after experiment and after distillation

Samples	Solvents volume before experiment (ml)	Solvents volume after experiment (ml)	Solvent volume after distillation (ml)
Pharmaceutical blister (primary)	100	≈85	≈80
Pharmaceutical blister (secondary)	100	≈90	≈80-85
Chips packaging			
Coffee packaging	50	≈45	≈40-42
Chocolate packaging			

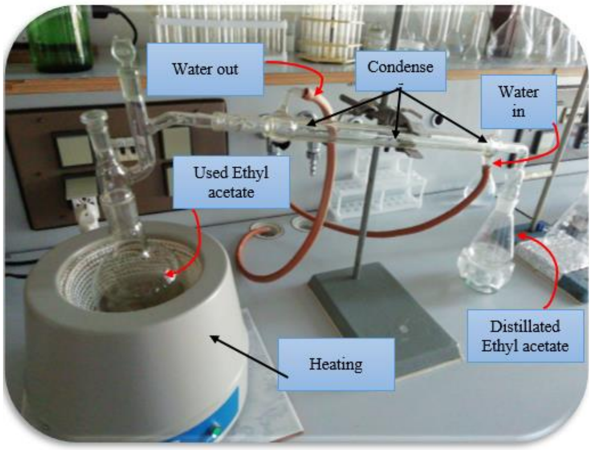

Figure 2. Distillation process used for ethyl acetate recovery

RESULTS OF ALUMINUM RECOVERING FROM MULTILAYER PACKAGING FOILS

The remaining samples must be catted and well dried for moisture removal purposes before analysis. The prepared Al-PE layer is catted into small portion and each samples weighed was different. For each experiment there was used: pharmaceutical blister (primary) packaging 10 g; pharmaceutical blister (secondary) packaging 10 g; packaging for chocolate, chips and coffee about 2 g the volume of reagent range was 50-100 ml. The extraction time was in the range of 5-10 minutes, the temperature range was 60-90 ºC and mechanical stirring ≈300 rpm; the volume of reagents was depended on the samples weight and size (samples must be covered by reagent). Separation by ethyl acetate and ethanol was not totally successful for all samples, after first step of separation, for some samples removed printed ink without using heating and stirring, so after heating (60-90 ºC), during 5-10 minutes, for all samples remove first layer of polymer, after that, it was possible to find that there was minimum four layers: two outer polymer layers: clear thin polymer layer and polymer layer with an unremoved paint; and polymer and Al layers together.

For the separation by acetic acid, dimethyl formamide and toluene, the first 5 minutes' separation result was the same, they easily removed the first layer of polymer, but after the experiment extension, the aluminum layer was starting to dissolve into the solvents. But in this case it is possible to separate, for example, by using filtration [15].

The result of separation of the pharmaceutical blister, packages for chocolate, coffee and chips

When pharmaceutical blisters were separated by acetic acid, acetone, dimethyl formamide, ethyl acetate, it was observed that separation time could be shortened. The process recovery can be seen in Figure 3. Here is shown example of pharmaceutical blister separation by ethyl acetate. We took about 10 g blister sample, add 100 ml solvent and used mechanical steering (≈300 rpm) and heating (90^0C) for duration of 8 minutes. After separation process, there are two separated layers Al and polymer layers. The two layers are Al layers with ink and polymer layers.

Separation process of packages for chocolate, coffee and chips, by six different reagents was not successful; because each sample has different aluminum and polymer layers. Here is example of separated packaging foils.

During Acetic acid and toluene treatment it was observed that packages, which are used for coffee, chocolate and chips are separated as three layers. The results are shown in Figure 4. The extraction time was 5-8 minutes, the temperature rate was 80-90 ^{0}C, also, mechanical stirring was used (≈300 rpm) in order to accelerate the process. After separation, here we can see one polymer layer with ink, also another polymer thin aluminum layer and Al layer.

After continuing separation process by using acetic acid and toluene, samples starting to dissolve in solvent, this process is shown in Figure 5.

Figure 3. Separation of pharmaceutical blisters

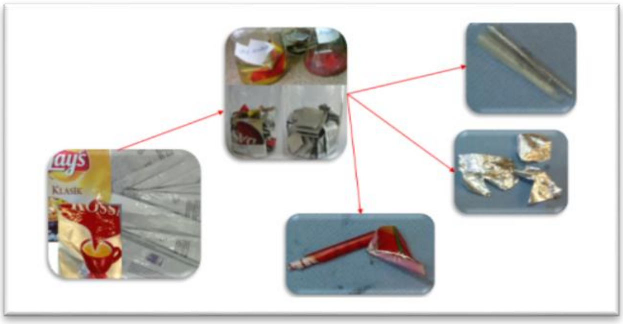

Figure 4. Separation packaging of coffee, chocolate, chips and secondary packaging for pharmaceutical blisters by using acetic acid, acetone, dimethyl formamide, ethanol, ethyl acetate, toluene (first step separation)

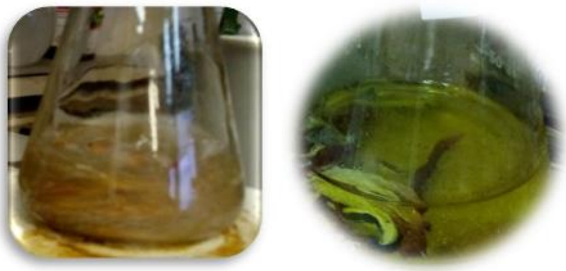

Figure 5. Separation packaging by acetic acid and toluene

By using acetone, the process need less time, and also it is possible process goes without heating, this is more safety and environmentally friendly solvent for separation.

As we can see, the separation result for all samples and reagents are not similar. It is necessary to find best available technique for separation multilayer packaging.

CONCLUSIONS

This research have shown that, on the first step of separation, there were separated minimum three layers. The study showed that, after separation Aluminum and polymer content in the packages was different for each samples: Pharmaceutical blister (primary) packaging ≈ 17.2% of Al and 82.2% of polymer; Pharmaceutical blister (secondary) packaging ≈ 46.2% of Al and 53.8% of polymer; Chips packaging≈ 51.5% of polymer (partially recovered); Coffee packaging ≈ 52.5% of Al and 47.5% of polymer; Chocolate packaging ≈52.5% of polymer (partially recovered). The experiment was successful for Pharmaceutical blister (primary) packaging, and less successful for other samples. As we noticed the main component was Al and polymer. After separation, also it was possible to regenerate used solvent, by simply distillation method, with minimum losses of solvents.

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Persian Abstract

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چکیده

زباله‌های بسته‌بندی چندلایه یکی از مشکلات عمده در جهان است. این نوع زباله حدود ۶ میلیارد تن بسته در هر سال در جهان تولید می‌شود (در اتحادیه اروپا حدود ۸۲ میلیون تن در سال تولید می‌شود). فیلم بسته‌بندی چندلایه تقریبا ۱۷٪ از تمام فیلم‌های بسته‌بندی تولیدشده است. اکثر فیلم‌های چندلایه تولیدشده بر پایه پلیمرهای مختلف نظیر: پلی اتیلن ترفتالات (PET)، پلی پروپیلن (PP) و پلی اتیلن (PE) به عنوان اجزای اصلی و لایه آلومینیوم است. با این حال، به دلیل کسر کوچک قابل بازیافت آن‌ها، اکثر لایه‌های چندگانه معمولاً سوزانده و یا دفن می‌شوند، این تلاش‌ها را در برابر یک اقتصاد دایره‌ای مقابله می‌کند. روش‌های مختلف بازیافت برای این نوع ضایعات وجود دارد، اما مشکل اصلی این است که کمپانی‌های کمتری وجود دارد که فیلم بسته‌بندی چندلایه را بازیافت می‌کنند. همچنین بسیار مهم است که اکثر آن‌ها تنها یک یا دو جزء را بازیافت می‌کنند و بقیه سوزانده یا تخلیه می‌شوند. هدف از این تحقیق، یافتن بهترین حلال برای بازیافت فیلم بسته‌بندی چندلایه و بازیافت دو عنصر رایج: پلیمر و آلومینیوم است. برای آزمایش اول، نمونه‌های مختلف فویل بسته‌بندی چندلایه انتخاب شدند، مانند: بسته‌بندی برای شکلات، چیپس، دارو و قهوه. از مواد شیمیایی متعددی استفاده گردید: اسید استیک، استون، دی متیل فرممید، اتانول، اتیل استات و تولوئن. این حلال‌ها در لیست مواد شیمیایی سبز قرار دارند و به دلیل تاثیرات کمتر آن‌ها بر محیط زیست و سلامت انسان، انتخاب شده‌اند. فرآیند بازیافت و نتیجه برای هر یک از نمونه‌ها و حلال‌ها به دلیل ویژگی‌های منحصر به فرد آن‌ها متفاوت می‌باشند.