Delamination and Separation of Aluminum-Polyethylene-Paper Packing Material

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Abstract: Delamination and separation of laminated aluminum- polyethylene-paper packaging material were conducted for recycling packaging materials. Delamination was carried out using glacial acetic acid (GAA) solution. L_9 (3^4) orthogonal experiments demonstrate that the most significant factor is GAA concentration followed by temperature and liquid/solid ratio. The delamination time decreased sharply with increasing temperature and the GAA concentration. The packaging material was delaminated under conditions of 60 ˚C, 70 v% GAA solution, liquid/solid ratio 20:1 and delamination time 60 min, and separation of polyethylene, paper and aluminum foil was conducted through sink-float method and air separation. Polyethylene, paper and aluminum foil were separated efficiently. The recovery and purity of aluminum foil was 90.81% and 100%, respectively; the purity of polyethylene was 100%; the recovery and purity of paper was 100% and 96.03%, respectively. This study offers some technical insights for recycling of aluminum-plastic packaging.

Keywords: Delamination, separation, aluminum, polyethylene, packaging material.

1. INTRODUCTION

Aluminum-plastic-paper material is widely used as packaging material owing to its light resistance, hygiene and low-cost [1], replacing many other materials, e.g. glass as containers for liquid foods. Paper gives strength to the packages, and the plastics ensure a tight seal. Polyethylene (PE) is the most common polymer used in food packaging and has excellent sealing properties and relatively low cost [2]. Aluminum foil is employed due to the poor barrier properties of PE for oxygen.

Wastes of aluminum-plastic-paper materials are constantly increasing due to the mass production and consumption [3]. The 2011 sustainable report of Tetra Pak shows that the the number of Tetra Pak packages is up to 158 billions while the recovery rate is only 20.1% in 2010. Since waste packages cause environmental pollution and waste of resources, and the growth of wastes has a great impact on their management.

Recycling allows the materials containing aluminum, high quality fibers and plastics to be reused, reducing disposal problem and increasing economic and social benefits [1]. Recycling of aluminum-plastic packaging materials receives increasing attention [3-6], and numerous techniques were developed such as separation with solvent [7-9], electrical separation [10] and argon electrolysis [11]. Separation of packaging materials can be achieved through soaking in acid or base solutions [3, 8]. Strong acid or strong base solutions can react with aluminum in the materials, and thus plastics are separated; weak acid solutions react with aluminum oxide that is between polyethylene and aluminum foil, and thus polyethylene and aluminum foil are delaminated [12]. In our study, delamination aluminum-polyethylene-paper packaging materials was studied, and separation of the delaminated products was further conducted through air separation and sink-float method. The separated PE was identified by Fourier transform infrared spectrum.

2. MATERIALS AND METHODS

2.1. Materials

The samples of packaging material were from Tetra Pak, which was a laminate of low-density polyethylene (LDPE), paper and aluminum foil. The intersecting surface of packaging material can be shown as Figure **1**, and the content of paper, LDPE and aluminum foil in the material was 75%, 20% and 5%, respectively [13]. Glacial acetic acid (GAA), analytically pure, was used to delaminate the laminated packaging material.

2.2. Delamination and Separation of the Packaging Material

The packaging material was cut into pieces of 1cm×1cm, and it was delaminated in 100 mL conical flask with GAA solution. GAA solutions with different concentration were prepared by mixing 100 mL GAA with certain volume of distilled water. The delamination

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experiments were conducted in a thermostat-controlled (DF-101S, Shanghai Bilon Instruments Co., LTD, China) water bath, and, after a given time, the conical flask was taken out. In orthogonal experiments, liquid/solid ratio was also studied, which was conducted by adding a certain amount of packaging material into a certain volume of GAA solution. The average value of delamination time was obtained from three tests.

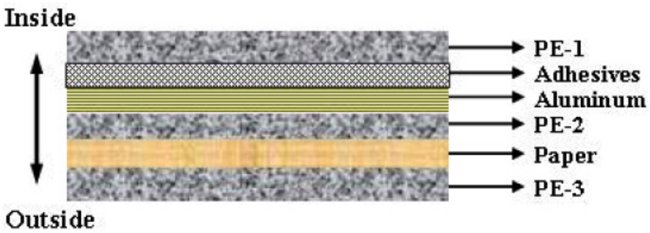

Figure 1: Schematic diagram of aluminum-polyethylene-paper packaging material.

After delamination of the packaging material with GAA, separation of the delaminated materials was further conducted. The delaminated PE was first separated by sink-float method, which was carried out by mixing the delaminated materials with tap water; after draining off, air separation was conducted to separate aluminum foil using the air separation column; the left materials were further soaked in GAA solution, and PE was separated from paper through sink-float method.

3. RESULTS AND DISCUSSION

3.1. L_9 (3^4) Orthogonal Experiments

The factors of temperature (A), GAA concentrations (B) and liquid/solid ratio (C) were investigated by orthogonal experiment of three factors and three levels, and the tests were performed without agitation. The variable assignment and the level settings are shown in Table **1**. The results of L_9 (3^4) orthogonal experiments are presented in Table **2**. K_1, K_2 and K_3 represent the sum of leaching rate of Al of level 1, level 2 and level 3 of a factor, respectively. $K_1/3$, $K_2/3$ and $K_3/3$ represent the average of K_1, K_2 and K_3, respectively. R denotes the maximum difference value among K_1, K_2 and K_3.

Table 1: Experimental Factors and Levels

Level	Factor		
	A (°C)	B (v%)	C (mL/g)
1	30	20	15
2	40	40	25
3	50	60	35

Table 2: The Results of L_9 (3^4) Orthogonal Experiments

	Factor			
	A (°C)	B (v%)	C (mL/g)	Delamination time (min)
1	1	1	1	4.9
2	1	2	2	2.5
3	1	3	3	1.8
4	2	1	2	3.6
5	2	2	3	1.25
6	2	3	1	0.95
7	3	1	3	3.2
8	3	2	1	1.1
9	3	3	2	0.25
K_1	9.2	11.7	6.95	
K_2	5.8	4.85	6.35	
K_3	4.55	3	6.25	
$K_1/3$	3.07	3.90	2.32	
$K_2/3$	1.93	1.62	2.12	
$K_3/3$	1.52	1.00	2.08	
R	1.55	2.90	0.23	

As demonstrated in Table **2**, the most significant factor is GAA concentration followed by temperature and liquid/solid ratio. The optimal scheme is $A_3B_3C_3$; the experimental conditions are 50 °C, 60 v% and liquid/solid ratio 35:1. In order to prove the optimized scheme, leaching tests were conducted under condition of temperature (50 °C), GAA concentration (60 v%) and liquid/solid ratio (35:1), and delamination time was 10 min, which is in agreement with the result from orthogonal experiments.

The GAA solution reacts with Al_2O_3 located on the surface of aluminum foil. The GAA ionizes first, and then the aluminium oxide is dissolved. The action mechanism of GAA can be explained by the Equation (1) [12]. The GAA concentration dominates mainly the reaction rate, and temperature has significant effect on the reaction.

$$\begin{aligned} &\mathrm{CH_3}COOH \rightarrow \mathrm{H}^+ + CH_3COO^- \\ &6H^+ + Al_2O_3 \rightarrow 2Al^{3+} + 3H_2O \end{aligned} \quad (1)$$

3.2. Effect of GAA Concentration on Delamination Time

The samples of the packaging material were delaminated in GAA solution at 60 ˚C to investigate the effect of concentration of GAA on delamination time, and the result is shown in Figure **2**.

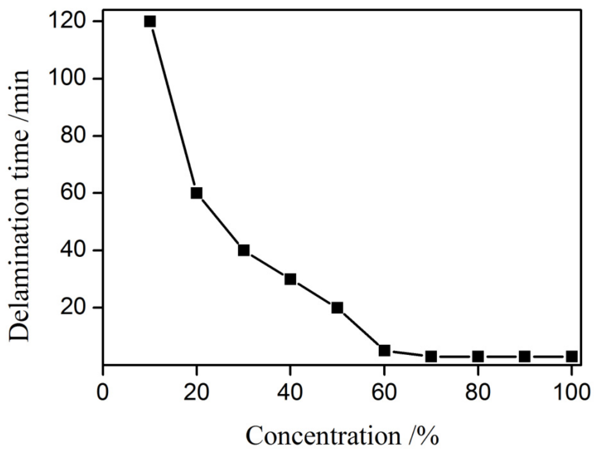

Figure 2: Delamination time as a function of GAA concentration (60 ˚C, liquid/solid ratio 20:1).

As shown in Figure **2**, the delamination time was 120 min when the concentration of GAA was 10 v%, and the delamination time decreased sharply with increasing the concentration of GAA. The delamination time was reduced to 5 min when the concentration of GAA was 60 v%, and the delamination time dropped very slightly with further increasing the concentration of GAA. Considering the high concentration of GAA was easily volatile, the proper concentration of GAA was 70 v%.

The delamination is the result of reactions between GAA and aluminum oxide. At lower concentration, the reaction rate rests on the ionization of GAA, which provides enough H^+ for delamination reaction. When the concentration is above 60 v%, the reaction rate is dominated by other factors such as temperature and the liquid/solid ratio.

3.3. Effect of Temperature on Delamination Time

Delamination of the samples of the packaging material was conducted in GAA solution at different temperature to investigate the effect of temperature on delamination time, and the result is shown in Figure **3**. Temperature has significant impact on delamination time; the delamination time reduces remarkably with increasing temperature. Increasing temperature results in a decrease of reaction rate, and temperature is the governing factor of delamination reaction at temperature above 60 ˚C. When the concentration of GAA is 60 v%, 70 v% and 80 v%, the delamination time at 60 ˚C is 5min, 3min and 3min, respectively. When the temperature is higher than 60 ˚C, the delamination time remains steady. The higher temperature promotes the volatilization of GAA, and thus the applicable temperature is 60 ˚C.

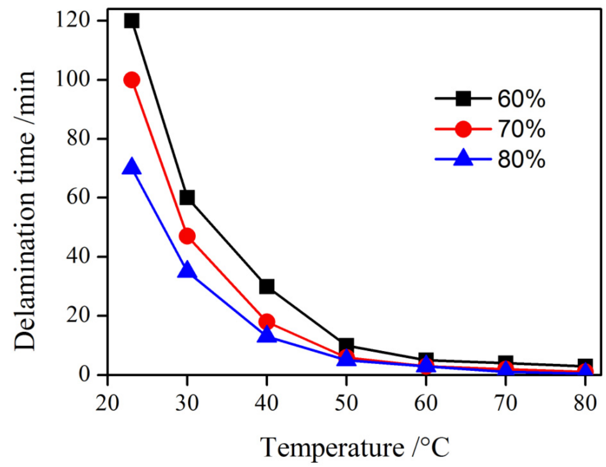

Figure 3: Delamination time as a function of temperature (liquid/solid ratio 20:1).

3.4. Effect of Liquid/Solid Ratio on Delamination Time

Delamination time as a function of liquid/solid ratio is shown in Figure **4**. It can be seen that delamination time decreases with an increase of liquid/solid ratio, the time reduces remarkably from 41 min to 27 min when the ratio increases from 10 to 25, and it drops slightly with further increasing of the ratio. From Figure **4**, it can be concluded that liquid/solid ratio of above 20 is practicable for delamination of the packaging material.

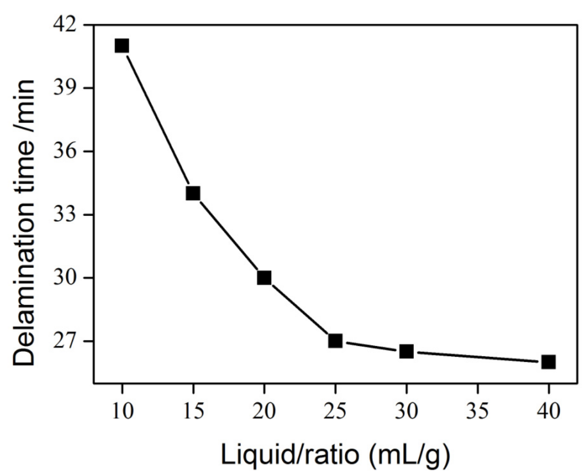

Figure 4: Delamination time as a function of liquid/ratio (60 ˚C, 40 v%).

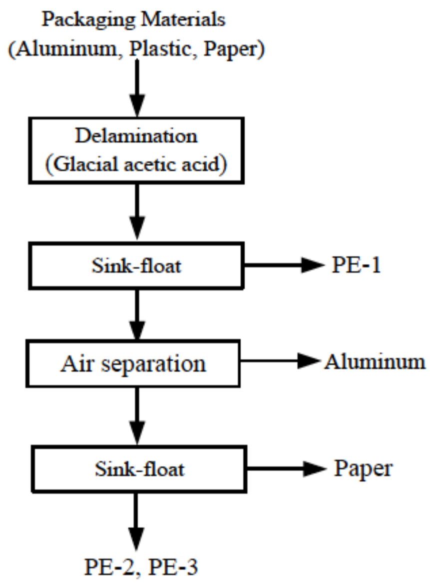

Figure 5: The flow sheet of separation of the packaging materials.

3.5. Separation of the Delaminated Materials

The packaging material was delaminated under conditions of 60 ˚C, 70 v% GAA solution, liquid/solid ratio 20:1 and delamination time 60 min. After filtration, the sink-float was conducted with water as medium, and the PE-1 was obtained. The left materials were drained off and separated using air separation, and the aluminum foil was separated. The left paper and PE were further soaked in 60% GAA solution for 30min with stirring rate of 20 rpm. After filtration, the sink-float was conducted with water as medium to separate paper and PE. The flow sheet of separation of the packaging materials is shown as Figure **5**, and the result is displayed in Table **3**.

Table 3: Separation Result of the Packaging Materials

Materials	Recovery (%)	Purity (%)
Aluminum foil	90.81	100
PE-1	95.45	
PE-2	100	100
PE-3	100	
Paper	100	96.03

As demonstrated in Table **3**, the recovery and purity of aluminum foil is 90.81% and 100%. The purity of PE is 100%, and a small quantity of PE-1 was left in paper. The recovery and purity of paper is 100% and 96.03%, and the paper was contaminated with a small amount of PE and aluminum foil.

(a) Aluminum foil

(b) PE-2, PE-3

(c) PaPer

(d) PE-1

Figure 6: The pictures of separated products.

The separated products are shown in Figure **6**. It can be seen that aluminum is separated from paper and PE plastic, and pure aluminum is obtained. PE-1 located inside of the packaging material is separated partly with little contamination, while PE-3 located outside of the material is contaminated with a little paper and printing ink. In separation process, paper pulp is achieved and can be used for paper recycling directly, and Figure **6c** shows the dried paper. The separated aluminum, PE-1 and paper are of high purity and can be utilized to produce high quality products.

The separated PE-1 was identified by Fourier transform infrared (Figure **7**). As shown in Figure **7**, spectrum of PE shows typical molecular vibrations at 2916 cm^{-1}, 2850 cm^{-1}, 1470 cm^{-1} and 723 cm^{-1}. Thereinto, molecular vibrations at 2916 cm^{-1} and 2850 cm^{-1} result from the asymmetric and symmetric stretching vibrations of methylene (-CH_2); the absorption peak at 1470 cm^{-1} is attributed to –C-C- bonds; molecular vibration at 723 cm^{-1} is rotational absorption peak of methylene (-CH_2) [14]. The strong absorption intensity is owing to that the tested PE film is thick. Fourier transform infrared spectrum of the separated PE verifies that purified PE is recovered effectively.

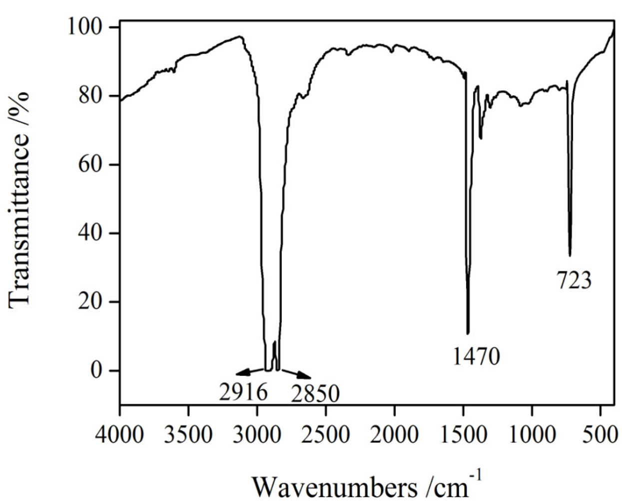

Figure 7: Fourier transform infrared spectrum of PE.

This experimental finding leads to an environmentally friendly method for the effective material cycling of large quantity of aluminum-plastic packaging. The GAA solution is cheap, and its recycling is available. The process is simple, and the delaminated products are recovered with high purity. Applications of this technology in the industry could have great potential economically and environmentally.

4. CONCLUSIONS

The orthogonal design was employed to investigate the effects of temperature, GAA concentration and liquid/solid ratio on delamination time. The L_9 (3^4) orthogonal experiments indicate the most significant factor is GAA concentration followed by temperature and liquid/solid ratio. Under the optimal experimental conditions of temperature (50 °C), GAA concentration (60 v%) and liquid/solid ratio (35:1), and delamination time was 10 min.

The delamination time decreased sharply with increasing of GAA concentration, temperature and liquid/solid ratio. The proper GAA concentration of was 70 v%, the optimal temperature was 60 ˚C and the practicable liquid/solid ratio is above 20.

The packaging material was delaminated under conditions of 60 ˚C, 70 v% GAA solution, liquid/solid ratio 20:1 and delamination time 60 min, and separation of polyethylene, paper and aluminum foil was conducted through sink-float method and air separation. Polyethylene, paper and aluminum foil were separated efficiently. The recovery and purity of aluminum foil was 90.81% and 100%, respectively; the purity of PE was 100%; the recovery and purity of paper was 100% and 96.03%, respectively. This study offers some technical insights for recycling of aluminum-plastic packaging.

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