

Exploration of Weak Acid Solvent Usage in the Separation of Aluminum (Al) from Multilayer Plastic Waste

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Abstract

Multilayer plastic waste is difficult to biodegrade, causing environmental problems. An alternative to processing multilayer plastic waste is to separate aluminum from the polymer using a delamination process with acid solvents. In this study, a combination of chemical (acid solvents), thermal (90°C), and mechanical (stirring at 300 rpm) methods was employed to evaluate the efficiency of delamination of multilayer plastic packaging. Several weak acid solvents, including acetic acid, citric acid, and formic acid, were tested at a concentration of 30% and a reaction time of 10 minutes. The method involved immersing cut multilayer plastic samples into acid solutions under controlled conditions, followed by filtration and drying to recover the aluminum layer. The best result was obtained using 30% formic acid, which successfully separated and recovered 0.940 g or 87% of aluminum from a 1.08 g multilayer sample. This process demonstrates an effective, relatively low-cost approach to enhance the recyclability of multilayer plastic waste, contributing to a more sustainable plastic waste management strategy.

Keywords

Multilayer Plastic, Delamination Process, Weak Acid Solvent, Aluminum

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1. INTRODUCTION

Aluminum (Al) separation from multilayer plastics is an important challenge in plastic waste management (Permana et al., 2023). Multilayer plastics, which are often used in the packaging of products such as snacks, coffee, and other packaged products, are difficult to recycle because the layers made of different materials, such as plastic and aluminum, require a complicated and expensive separation process (Cabrera et al., 2022; Soares et al., 2022; Tambunan et al., 2024). If multilayer plastic waste is not managed properly, it can cause negative impacts on the environment. Various separation methods for multilayer plastics have been developed, such as solvent extraction, thermal separation, and density-based separation. One effective method is the use of weak acid compounds, which can reduce the bond between plastic and aluminum without dissolving aluminum, so that aluminum can be reused (Lahtela et al., 2020; Ügdüler et al., 2022). Successful separation will result in separate aluminum and plastic, making both easier to recycle. Aluminum, which has high thermal conductivity (273 W/mk), can be used for a variety of applications, including as a conductor material in the pyrolysis process, which helps to

create an even temperature and produce a more consistent product (Li et al., 2020; Nugraha et al., 2022).

Separation of aluminum from multilayer plastics using acid solvents is usually done by the chemical delamination method. This process utilizes the properties of acids that are able to dissolve or weaken the bond between layers so that aluminum can be separated from plastic. Separating aluminum from multilayer plastics using weak acid solvents that are more environmentally friendly and safer than strong acids (Capkin and Gokelma, 2023; Diop et al., 2017). Separation of Aluminum from multilayer plastics using strong acids i.e. hydrochloric acid (HCl) can lead to the formation of aluminum chloride, which dissolves Aluminum and reduces the effectiveness of separation (Yan et al., 2015). In addition, research by Loukodimou et al. (2024) used a mixture of carboxylic acids and bases to weaken the adhesion bond between plastic and Aluminum, allowing for more effective separation.

This research aims to develop a method of separating Aluminum foil from multilayer plastic by using a variety of solvent types in the delamination process. This process aims to remove the bond between plastic and Aluminum, resulting in reusable Aluminum foil, particularly as a heat

conductor.

2. EXPERIMENTAL SECTION

2.1 Materials

The raw material was multilayer plastic waste collected from the neighborhoods around Palembang and Indralaya. Solvent materials with various acid variations consisting of weak acids namely formic acid (HCOOH) 98% from Merck, acetic acid (CH₃COOH) 99.8% from Merck, and lactic acid (C₃H₆O₃) 90% from Merck, as well as deionized water were purchased at the Palembang Chemical Store.

2.2 Methods

Multilayer plastic waste raw materials were first manually cleaned to remove any residual contaminants such as food or adhesive. The cleaned materials were then cut into pieces of approximately 3 cm × 3 cm to ensure uniform exposure during the delamination process. A mass of ±1 g of the prepared multilayer plastic was immersed in a weak acid solution (acetic acid, citric acid, or formic acid) with varying concentrations of 10%, 20%, 30%, and 40% (v/v) in a closed 100 mL borosilicate glass beaker. The acid-plastic mixture was subjected to a thermal and mechanical delamination treatment by heating at 90°C with constant magnetic stirring at 300 rpm for 10 minutes. These operating parameters were selected based on previous studies that showed the synergy of thermal and mechanical agitation accelerates acid penetration and interlayer separation. After treatment, the mixture was allowed to cool to room temperature. Following the delamination reaction, the mixture was filtered using vacuum filtration to recover the separated aluminum foil layer. The recovered aluminum was washed with deionized water to remove residual acid and other impurities, then dried in an oven at 80°C for 5 hours. After drying, the aluminum and the remaining polymer layer were weighed using an analytical balance with ±0.001 g precision. This allowed the calculation of delamination efficiency and aluminum recovery yield. This study employed a quantitative experimental method, focusing on the measurable outcome of aluminum recovery as a function of acid type and concentration. Data collection involved direct mass measurements (pre- and post-delamination), while the data analysis was conducted using descriptive statistics and comparative analysis to identify the optimum delamination condition. The underlying research perspective is rooted in environmental engineering and circular economy principles, aiming to develop scalable and eco-friendly solutions for multilayer plastic waste treatment (Creswell and Creswell, 2018; Singh et al., 2017).

3. RESULTS AND DISCUSSION

3.1 Separation Mechanism of Aluminum from Multilayer Plastic

The separation of aluminum from multilayer plastics is based on the principle of “like dissolves like”, i.e. the solvent is

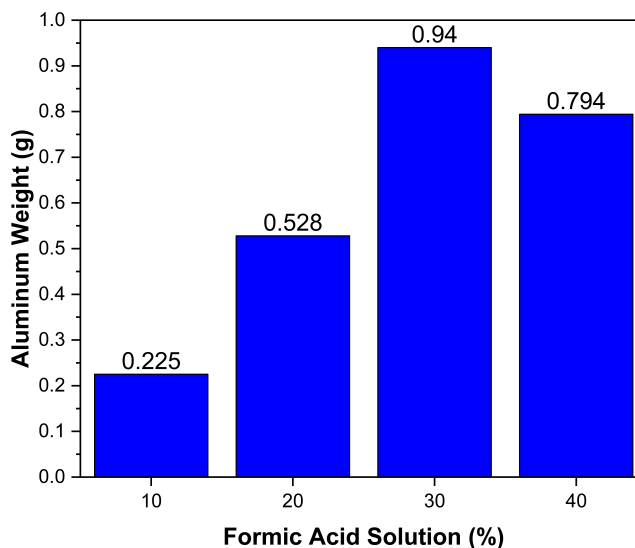


Figure 1. Aluminum Weight Using Formic Acid

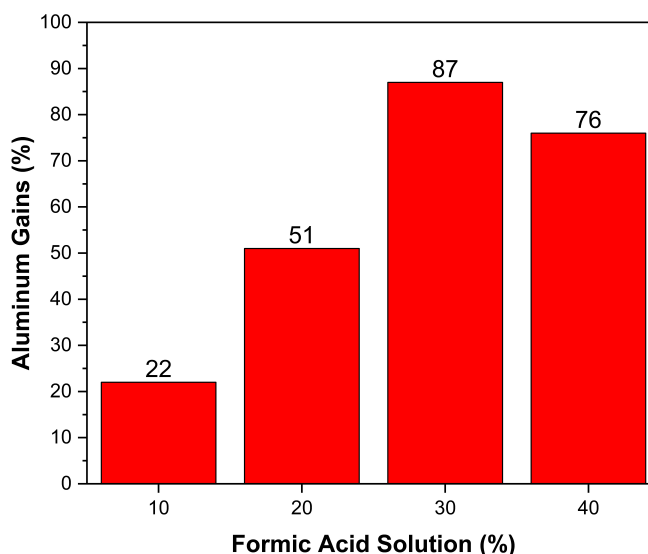


Figure 2. Aluminum Recovery Percentage Using Formic Acid

selected based on its suitability to the type of adhesive to be dissolved. The adhesive in multilayer plastic is polar, so a polar solvent is needed to dissolve it. The polar solvents used were formic acid, acetic acid, and lactic acid, with concentrations of 10%, 20%, 30%, and 40% in a mixture with 100 mL of distilled water. The separation process was carried out at 90°C for 10 minutes to ensure complete separation of plastic and aluminum.

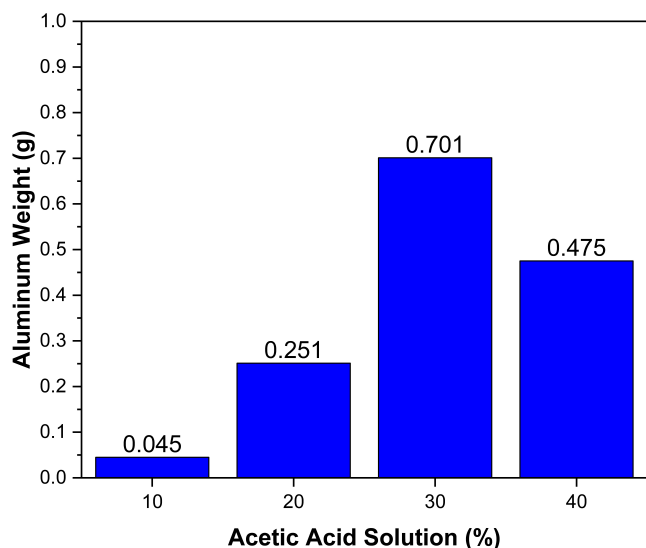
When the solvent comes in contact with the polyurethane adhesive, the solvent molecules diffuse into the polymer matrix due to the difference in concentration. This diffusion causes the transfer of solvent molecules from the

Table 1. Separation of Aluminum with Formic Acid Solution

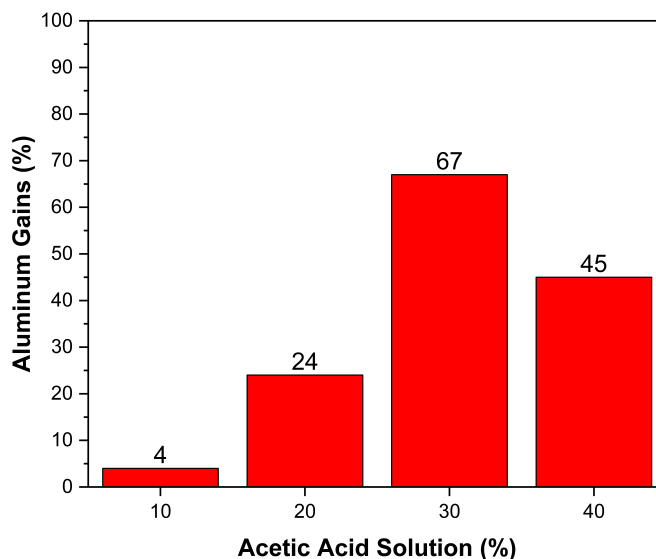
Formic Acid Concentration (% v/v)	10	20	30	40
Initial weight of multilayer plastic (g)	1.037	1.029	1.076	1.046
Al weight (g)	0.225	0.528	0.940	0.794
Percentage of Al gains (%)	22	51	87	76

high-concentration area (solvent) to the low-concentration area (adhesive) until equilibrium is reached. During this process, solvent molecules penetrate the polyurethane, which results in swelling of the polymer chains. The swelling occurs because the solvent disrupts the intermolecular forces that bind the polymer chains, thereby enlarging the distance between the chains.

As diffusion continues, the adhesive begins to detach from the substrate due to the weakening of the polyurethane network structure. Delamination occurs because the solvent reduces the cohesive strength of the adhesive, making it unable to maintain a bond with the surface. Prolonged exposure to solvents can break chemical bonds in the polyurethane structure through hydrolysis, particularly in the urethane bond.

**Figure 3.** Weight of Aluminum Using Acetic Acid

Adhesive decay is caused by the disruption of several types of bonds, such as hydrogen bonds and urethane bonds ($-\text{NH}-\text{CO}-\text{O}$). Hydrogen bonds, which are abundant in the soft segments of polyurethane, are disrupted when polar solvents form new hydrogen bonds with the polymer chains. This weakens the structure of the adhesive. In addition, urethane bonds are prone to hydrolysis in the presence of water vapor or polar solvents. This hydrolysis

**Figure 4.** Aluminum Recovery Percentage Using Acetic Acid

facilitates the degradation of the polymer network, leading to swelling, plasticization, and eventual breakdown of the adhesive, resulting in a loss of bond strength and integrity (Sánchez-Rivera et al., 2023).

3.2 Separation of Aluminum with Formic Acid Solvent

Separation of Aluminum with Formic Acid Solvent Formic acid (HCOOH) plays an important role in the separation of aluminum from multilayer plastics, especially in dissolving the adhesive layer and weakening the bond between layers without damaging the aluminum. Formic acid is a polar solvent that can dissolve and break down the polyurethane adhesive on multilayer plastics, thus separating the aluminum foil from the plastic layer (Ügdüler et al., 2022). This delamination process is influenced by formic acid concentration and reaction time, with an increase in formic acid concentration accelerating the separation. In Figures 1 and 2 and Table 1 for formic acid concentrations of 10% to 30%, the weight of aluminum produced continues to increase and the peak at 30% concentration is obtained as much as 0.940 g (87%) of aluminum, that is, almost all aluminum is successfully separated in 10 minutes, there is little possibility of acid residue being absorbed into aluminum. However, at 40% formic acid concentration, the weight of aluminum obtained decreased to 0.794 g (76%). The results show that the concentration of formic acid affects the effectiveness of Aluminum separation from multilayer plastic. Formic acid (HCOOH) plays a role in the separation of Aluminum from multilayer plastics, especially in dissolving the adhesive layer and weakening the bond between layers without damaging Aluminum. At concentrations of 10-30%, Aluminum

remains clean and does not undergo significant chemical reactions. At 40%, there is a possibility of reaction with the Aluminum oxide layer, causing discoloration or slight corrosion causing the Aluminum yield to decrease. reaction with the aluminum oxide layer (Al_2O_3), causing discoloration or slight corrosion.

Table 2. Separation of Aluminum with Acetic Acid Solution

Acetic Acid Concentration (% v/v)	10	20	30	40
Initial weight of multilayer plastic (g)	1.088	1.028	1.042	1.050
Al weight (g)	0.045	0.251	0.701	0.475
Percentage of Al gains (%)	4	24	67	45

3.3 Aluminum Separation with Acetic Acid Solvent

Acetic acid is a polar solvent with a high dielectric constant that effectively dissolves polyurethane adhesives on multilayer plastics, so it has the ability to separate aluminum foil from multilayer plastics (Haque et al., 2023). This separation process depends on the solvent concentration and reaction time, with 30% acetic acid solvent being able to delaminate almost 70% or 0.701 g of aluminum in 10 minutes (Table 2 and Figures 3 and 4). In contrast, 20% solvent is less effective, while 40% solvent can damage aluminum if used for 10 minutes, so a shorter reaction time is required. The results show that the acid concentration affects the speed and effectiveness of aluminum separation.

3.4 Aluminum Separation with Lactic Acid Solvent

Lactic acid is a polar solvent capable of dissolving polyurethane adhesives and breaking the bond between aluminum and plastic layers through water-facilitated hydrolysis of urethane. However, the Aluminum separation process using lactic acid is less effective than formic acid and acetic acid. The percentage of Aluminum separation from multilayer plastic only reached 23% or 0.239 g with 30% lactic acid solvent in 10 minutes can be seen in Figures 5 and 6, for 40% solvent the Aluminum recovery decreased to 13% which can damage aluminum if higher concentration solvents are used (Table 3).

Table 3. Separation of Aluminum with Lactic Acid Solution

Lactic Acid Concentration (% v/v)	10	20	30	40
Initial weight of multilayer plastic (g)	1.088	1.028	1.042	1.050
Al weight (g)	0.139	0.144	0.239	0.132
Percentage of Al gains (%)	13	14	23	13

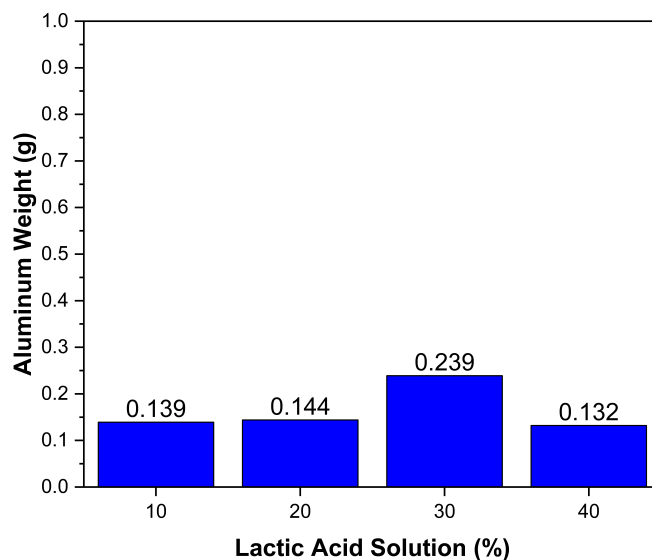


Figure 5. Weight of Aluminum Using Lactic Acid

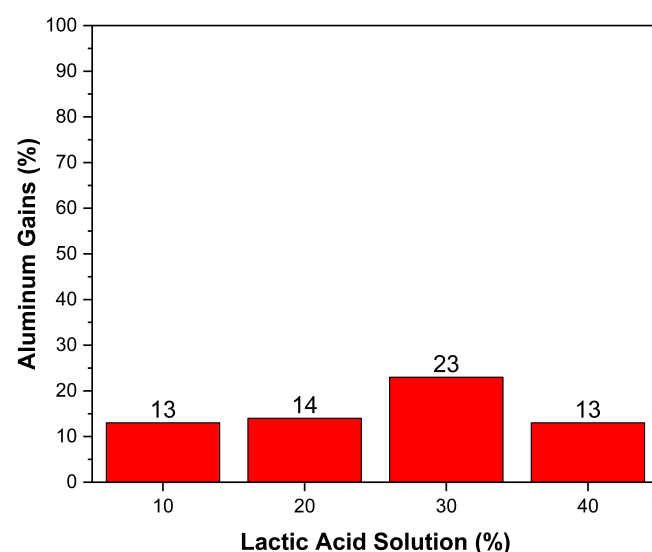


Figure 6. Aluminum Recovery Percentage Using Lactic Acid

3.5 Effect of Weak Acid Solvent on Aluminum Separation from Multilayer Plastics

Formic acid with a pKa of 3.75 was a more effective solvent in dissolving the adhesive than acetic acid and lactic acid. Acetic acid with pKa 4.76 is less powerful in dissolving adhesives due to its weaker nature. Requires a higher concentration (>30%) to get the same separation results as formic acid. While lactic acid has a pKa value of 3.86 similar to formic acid but weaker. Can dissolve adhesives but the reaction is slower than formic acid. It is more viscous due to its high viscosity, so diffusion into multilayer plastics is also slower, more environmentally friendly, but less efficient for



Figure 7. Determination Results of Multilayer Plastic (a) Formic Acid, (b) Acetic Acid, and (c) Lactic Acid

rapid separation. In terms of dipole moment, Formic Acid (HCOOH) has a dipole moment of 1.41 D. It is effective in dissolving adhesives because it is quite polar and has a small molecular size. Quickly penetrates the adhesive layer, Acetic Acid (CH_3COOH) with a dipole moment of 1.74 D, more polar than formic acid, can penetrate adhesives faster than formic acid, but requires higher concentrations (>30%) for optimal efficiency.

Less reactive than formic acid due to its larger size, so it is slower in penetrating the adhesive structure. Lactic Acid ($\text{C}_3\text{H}_6\text{O}_3$) with a dipole moment of 2.69 D. is most effective in attracting adhesives due to its high polarity and ability to form many hydrogen bonds. It is better at dissolving protein-based or polyurethane-based adhesives with strong hydrogen bonds. However, the large molecular size causes slower diffusion than formic acid and acetic acid. It is more viscous, making it less effective for applications that require rapid separation (Usino et al., 2020).

Polyurethane or epoxy-based adhesives, lactic acid is better because it has the highest dipole moment, but it is slower in diffusion. For quick and efficient methods, formic acid remains the best choice despite its lower dipole moment, but its molecular size is smaller and penetrates the adhesive more easily.

Acetic acid is somewhere in the middle, better than formic acid at dissolving adhesives but not as fast as formic acid and not as effective as lactic acid in more complex adhesives. Formic acid (HCOOH) is most balanced in efficiency and speed of aluminum separation from multilayer plastics.

In Figures 7 and 8, aluminum separation using formic acid is more effective than acetic acid and lactic acid in 10 minutes at 90°C , because formic acid has the shortest molecular chain (one carbon atom), which allows faster diffusion into the adhesive. In contrast, acetic acid and lactic acid have longer chains, which reduce polarity and slow down the separation process, requiring higher solvent

concentrations and longer times for the same results.

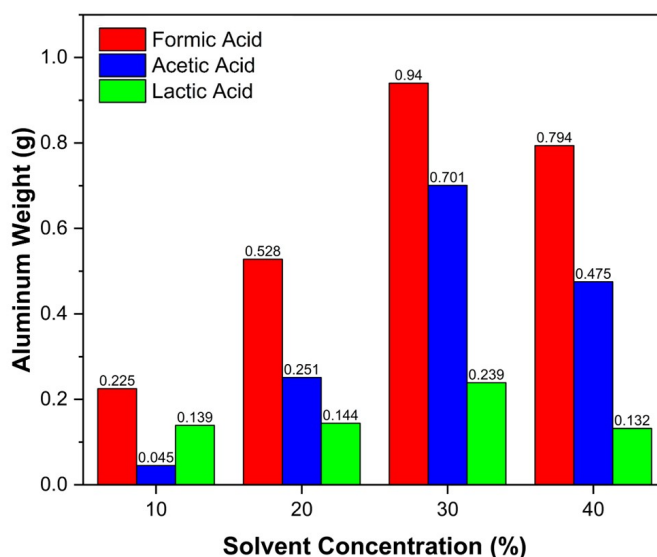


Figure 8. Effect of Weak Acid Solvents on Aluminum Separation

Dielectric constant (ϵ) is a measure of the ability of a solvent to weaken the attractive force between charged ions or molecules. In the context of separating aluminum from multilayer plastics, the dielectric constant affects the ability of the acid to dissolve the adhesive that binds the aluminum to the plastic. Formic acid has the highest dielectric constant (58), followed by lactic acid (19), and acetic acid (6.2). Formic acid is most effective in weakening polar adhesives such as polyurethane due to its high dielectric

constant, as it easily penetrates the adhesive structure and separates the aluminum layer from the plastic. However, if the concentration is too high, it can cause mild corrosion of aluminum.

Acetic Acid in terms of dielectric constant is less effective in weakening polar adhesives due to its low dielectric constant (Bibi et al., 2023; Wang et al., 2024, 2021), better in dissolving non-polar adhesives, but less optimal for polyurethane-based adhesives. It required more time in the process of separating aluminum from multilayer plastics. The dielectric constant of a solvent determines its ability to weaken the attractive force between the adhesive molecules that bind aluminum and plastic (Raos and Zappone, 2021; Sanghvi et al., 2022; Shanmugasundram et al., 2022). Adhesives on multilayer plastics are generally polyurethane or epoxy based, which have strong polar and hydrogen bonds. Therefore, solvents with a high dielectric constant are more effective in dissolving such adhesives (Bourgi et al., 2024; Liu et al., 2021; Rimšaitė et al., 2019; Zhang et al., 2021a,b).

4. CONCLUSIONS

The effectiveness of Aluminum separation from multilayer plastics using a variety of weak acids is highly dependent on the molecular size of the solvent. Formic acid has a smaller molecular size than acetic acid and lactic acid so that the ability to diffuse with the adhesive is better so that with a concentration of 30% it can separate the Aluminum layer as much as 87% within 10 minutes.

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