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A novel aqueous route for separating aluminum and plastic recovery in multilayer packaging waste for sustainable recycling

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Abstract. Multilayer packaging, such as blister packs and milk cartons, poses significant recycling challenges due to the strong interlayer adhesion. This study investigates the chemical separation of aluminum and plastic by using aqueous ammonia-based solutions. For pharmaceutical blister packs, an $NH_3/NH_4Cl/H_2O$ solution was applied. Aluminum recovery was determined through titration of dissolved Al^{3+} and gravimetric analysis of residual foil; PVC recovery was measured with gravimetric methods. Under optimal conditions (5 M NH_3, 2.5 M NH_4Cl, 1:25 g/mL ratio, 60-minute immersion at 70 °C), the process achieved complete separation, yielding 10.7% aluminum and 88.8% plastic by mass. For milk cartons, separation was carried out using only an NH_4OH/H_2O solution under the same temperature and ratio, but with 5 M NH_4OH and 30-minute immersion. The recovered fractions included 7.8% plastic, 63.7% paper, and 26.3% aluminum-plastic composite, while no separated aluminum foil was observed. A maximum recovery efficiency of 97.82% was achieved, indicating substantial, though incomplete, material separation. These results demonstrate that while ammonia-based chemical treatments enable nearly complete recovery from blister packs, separation from milk cartons is less effective because of stronger bonding between aluminum and plastic. Nonetheless, the method contributes to resource recovery and waste reduction, supporting circular-economy goals in multilayer packaging management.

Keywords: aluminum recovery, multilayer packaging, ammonia-based solutions

1 Introduction

Multilayer packaging, particularly that combines aluminum and plastic, is an essential and growing component of municipal waste, vital for preserving and distributing food, beverages, pharmaceuticals, and other consumer products [1, 2]. This type of packaging is extensively used in industries like food and beverage, pharmaceuticals, and consumer goods because of its ability to provide superior protection, durability, and convenience. Common examples include milk cartons (Tetra Paks), blister packs for pharmaceuticals, and flexible pouches for snacks and liquids [1, 3–6].

These packages often feature multiple layers of materials such as polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polyvinyl chloride (PVC), paper and thin aluminum foil [1, 3, 5]. For instance, food and beverage cartons typically use a thin aluminum foil as a barrier, laminated to plastics like low-density polyethylene (LDPE) or linear low-density polyethylene (LLDPE), or cardboard [5]. Tetra Paks typically consist of 74% paper, 22% polyethylene, and 4% aluminum.

Pharmaceutical blister packages commonly consist of plastic films (85%) (e.g., PVC, PP, PET) and aluminum layers (15–17%), with some variations including aluminum in both forming

and lidding films (cold forming packages) [6–8]. The synergy between aluminum and plastic in multilayer structures results in packaging that is lightweight, cost-effective, and highly effective in preserving product integrity [9, 10]. Aluminum's ability to block external elements that could cause spoilage or degradation makes it indispensable in packaging solutions like Tetra Paks, blister packs, and flexible pouches, while plastics add flexibility, toughness, and printability. This combination makes multilayer packaging a preferred choice for industries requiring high-performance solutions that meet stringent requirements for product preservation and consumer convenience [3].

However, this complex, multi-material structure poses a significant challenge for existing recycling systems [1, 3]. Because of the difficulty in separating these tightly bonded layers, multilayer packaging waste is frequently disposed of in landfills or incinerated alongside other municipal solid waste. This approach leads to substantial resource waste and environmental pollution. Incineration of plastics can release hazardous gases such as dioxins, hydrogen chloride, and nitrous oxide, leading to environmental problems and preventing aluminum from being recycled. Landfilling, on the other hand, can cause soil acidification because of the presence of aluminum and plastic [3–5, 11].

Recycling aluminum is crucial, as producing it from bauxite is highly energy-intensive [12]. In contrast, transforming aluminum scrap into recycled aluminum requires only 5% of the energy used for virgin production and generates just 5% of the greenhouse gas emissions [6, 12, 13]. Therefore, recovering both the polymeric and metallic layers as separate entities has gained increasing attention [11, 14]. The primary goal of these recycling efforts is to recover both aluminum and polymers with high purity and quality to ensure their economic value and prevent "down-cycling" [1, 3, 11].

Several methods have been developed to separate aluminum and plastic layers in multilayer packaging waste, particularly Tetra Paks and pharmaceutical blister packs (Table 1). These include thermal, mechanical, chemical (hydrometallurgical), and solvent-based approaches, each with distinct principles and limitations. Thermal processing, such as pyrolysis, vapourises non-metallic components, allowing aluminum recovery; however, high temperatures may oxidise aluminum, reducing its value, and produce toxic emissions. Mechanical methods, like cryo-comminution with electrostatic separation or electrohydraulic fragmentation [15], physically delaminate materials with relatively high recovery efficiency but are often energy-intensive and prone to cross-contamination. Hydrometallurgical methods involve dissolving aluminum with acids or bases (e.g., HCl, NaOH, and methanoic acid), achieving high recovery but generating corrosive wastewater and potential material loss [7, 8, 15]. Solvent-based methods, including selective dissolution-precipitation (SDP) [1, 16] and switchable hydrophilicity solvents [14], target the polymer matrix or adhesive. Samy Yousef et al. (2018) [17] used switchable hydrophilicity solvent (N-dimethylcyclohexylamine, DMCHA), combining with ultrasonic treatment to recover metallic and nonmetallic components from waste pharmaceutical blisters. Switchable hydrophilicity solvents systems like DMCHA offer high recovery rates and energy efficiency though they may slightly discolour plastics or cause minor aluminum oxidation. Challenges across all methods include high energy demands, secondary pollution, solvent recyclability, and the need for material purity in downstream recycling applications.

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The reviewed literature highlights the urgent need for the development of efficient and environmentally friendly technologies to process complex multilayer waste such as pharmaceutical blisters and aseptic cartons. Meeting the goals of a circular and carbon-neutral economy requires solutions capable of recovering both aluminum and polymers with high quality and purity. The growing market for recycled plastics and the global shift toward circularity further emphasise this demand [1, 4, 15]. In response, this study investigates the use of an $NH_3/NH_4Cl/H_2O$ solution to separate aluminum foil and plastic from pharmaceutical blister packs and milk cartons. The method is based on weakening the interfacial adhesion between the layers, facilitating their gradual delamination.

Table 1. Methods for separating aluminum and plastic in multilayer packaging waste

No.	Method	Description	Recycling Rate and Purity Achieved	Applications	Challenges
1	Selective Dissolution-Precipitation (SDP) [1]	Uses sustainable solvents (e.g., biodiesel, 2-MeTHF, and CPME) to dissolve LDPE, leaving aluminum intact.	~100% Al >98%, LDPE comparable to virgin	Tetra Paks, pouches	Solvent recovery, cost
2	Switchable Hydrophilicity Solvents [14]	Uses CO_2-switchable solvents to break bonds and separate layers via CO_2 saturation under cooling.	>99%, High for both Al and plastics	Blister packs, pouches	Scalability, CO_2 handling
3	Alkali Dissolution [7,8,15]	Dissolves aluminum with NaOH or KOH, forming so dium aluminate, which can be precipitated as aluminum hydroxide.	High, Al as hydroxide, plastics intact	Tetra Paks, blister packs	Hydrogen gas management, safety
4	Nitric Acid Delamination [15]	Uses 50–70% nitric acid to dissolve adhesive binder, separating aluminum and plastic in 4–7 hours.	Moderate, Good for plastics, Al intact	Blister packs, films	Long processing time, safety risks
5	Organic Acids with Surfactants [15]	Uses mixtures of 40–70% organic acids (e.g., acetic and formic), 10–20% surfactants, and 20–40% solvents to reduce adhesion.	Moderate, Variable	Blister packs, pouches	Energy-intensive, solvent disposal
6	Fatty Acids Delamination [15]	Fatty acids diffuse through LDPE to the aluminum interface, reducing adhesion via ionic, hydrogen, and Van der Waals bonds.	Low to Moderate, Variable	Tetra Paks, pouches	Long reaction times, low efficiency
7	Density Separation [18]	Separates plastics by specific gravity (e.g., PP/PE float and PET sinks) after aluminum removal.	High for plastics, Depends on prior steps	Tetra Paks, post-delamination	Requires pre-treatment
8	Extrusion [18]	Extrudes low specific gravity plastics (PP/PE) at ≤220 °C with a 20–200 mesh screen to filter unmelted PET.	Moderate, Depends on prior steps	Tetra Paks, post-delamination	Limited to specific polymers
9	Electrostatic Separation [13,15]	Separates aluminum from polymers based on electrical conductivity, often post-delamination.	Moderate to High, Depends on prior steps	Tetra Paks, blister packs	Requires pre-treatment, variable purity

No.	Method	Description	Recycling Rate and Purity Achieved	Applications	Challenges
10	Pyrolysis/Gasification [19,20]	Heats waste to break down plastics into hydrocarbons (fuels, waxes), recovering aluminum as a solid residue.	Variable, Al may oxidise	Tetra Paks, mixed waste, blister packs	Energy-intensive, potential contamination

2 Materials and methods

2.1 Materials

This study used multilayer packaging waste materials, including pharmaceutical blisters (PVC plastic film) and milk cartons. The collected blister packs were cut into small samples with ranges of 1 × 1 cm, 2 × 2 cm, and 3 × 3 cm (Fig. 1a). These samples were sourced from medical facilities located in Go Vap District, Ho Chi Minh City. The milk cartons were collected and cleaned thoroughly with tap water to remove residual milk and surface contaminants. After air-drying, the cartons were cut into samples of three dimensions: 1 × 1 cm, 1 × 2 cm, and 1 × 3 cm, for subsequent experiments (Fig. 1b).

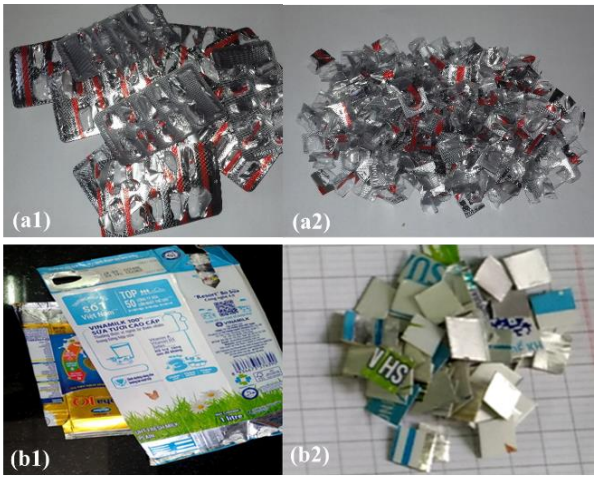

Fig. 1. Pharmaceutical blister samples before cutting (a1) and after being cut into 1 × 1 cm pieces (a2); Tetra Pak milk carton samples before cutting (b1) and after being cut into 1 × 1 cm pieces (b2)

For the separation experiments, a solution of ammonia (NH_3) and ammonium chloride (NH_4Cl), both supplied by Xilong Chemical Co., China, was used as the chemical agent. This solution was selected for its potential to weaken the adhesion between aluminum and polymer layers, facilitating effective delamination.

2.2 Separation of aluminum and plastic components from multilayer packaging waste using aqueous ammonia-based solutions

The separation of aluminum and plastic layers was carried out with aqueous ammonia-based solutions, specifically a mixture of ammonia (NH_3, 1–6 M) and ammonium chloride (NH_4Cl, 0.5–3 M). A two-gram sample of chopped pharmaceutical blister packs was introduced into an Erlenmeyer flask containing the solution, with a solid-to-liquid ratio ranging from 1:5 to 1:30 (g/mL). The suspension was heated and stirred with a magnetic hotplate (Stuart CB162, 230 V, 50–60 Hz) at controlled temperatures, and the soaking time varied across 15, 30, 60, and 90 minutes. After treatment, the aluminum foil and plastic were separated by filtration. The dissolved aluminum content in the solution was quantified with the complexometric titration method.

Following the soaking step, insoluble fractions were collected via filtration and subjected to froth flotation to further separate the aluminum-plastic composite and plastic components. Froth flotation was conducted by injecting air (2 L/min) for 10 minutes in the presence of 0.1% methyl isobutyl carbinol (MIBC) as a frothing agent. During this process, PVC

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particles adhered to the air bubbles and floated to the surface, while denser aluminum foil settled at the bottom. Air was then stopped to allow phase separation, and both fractions were collected, naturally dried at ambient temperature, and weighed to determine recovery efficiency.

A schematic diagram of the experimental workflow is presented in Fig. 2, illustrating the chemical delamination process followed by flotation-based physical separation.

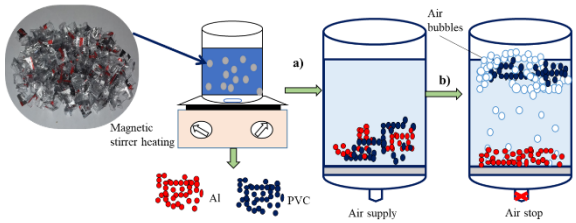

Fig. 2. Schematic illustration of the separation process of aluminum and plastic components from multilayer pharmaceutical blister packaging using ammonia-based chemical treatment followed by froth flotation

For Tetra Pak milk carton samples, the separation process was carried out with aqueous ammonium hydroxide (NH_4OH, 1, 3 and 5 M) as the sole delaminating agent, based on its moderate reactivity and superior preservation of the paper component. Preliminary tests with a mixture of NH_4OH and NH_4Cl—found effective in blister pack separation—led to severe degradation and fragmentation of the paper layer. Therefore, all subsequent trials employed NH_4OH alone to minimise damage and improve selectivity. Approximately 2 g of manually cut milk carton pieces were immersed in an NH_4OH solution with a solid-to-liquid ratio ranging from 1:5 to 1:30 (g/mL). The suspension was heated to 30–70 °C and stirred with a magnetic hotplate for 15, 30, 45, or 60 minutes. After treatment, the mixture was filtered to collect insoluble materials, while the filtrate was retained for aluminum quantification. The solid residues were then subjected to froth flotation under the same conditions as applied for blister pack samples—air injection at 2 L/min for 10 minutes in the presence of 0.1% MIBC. During this process, the hydrophobic aluminum-plastic fraction floated to the surface, while the more hydrophilic paper fibres remained dispersed or settled. Both fractions were collected, washed, air-dried, and weighed to evaluate recovery efficiency.

2.3 Quantification of aluminum and the separated fractions

The concentration of Al^{3+} ions in aqueous ammonia and ammonia-mmonium chloride solutions was quantified by using a two-step acid-base titration with 1 M HCl and 10% potassium fluoride (KF), employing phenolphthalein as an indicator. A 5 mL sample aliquot was diluted with 50 mL distilled water, and 3–8 drops of phenolphthalein turned the solution pink. Titration with 1 M HCl continued until the pink colour disappeared, followed by the addition of a known volume of 10% KF solution to restore a light pink colour. A second titration with 1 M HCl to complete decoulorisation, confirmed by an equal KF volume, quantified the Al^{3+} content. This method is consistent with the principle described in MS 699:2008 [21], which utilises KF to render aluminum neutral to phenolphthalein, enabling indirect quantification via titration. Comparable findings by Moon and Yang (2017) [22] confirmed the selective reactivity of KF with Al^{3+} in acidic media, validating its applicability in determining aluminum content through either gravimetric or titrimetric means. This approach offers a practical and reliable means to assess aluminum dissolution in complex aqueous systems.

The separated fractions via froth flotation were collected, washed with distilled water, naturally dried at ambient temperature (25–30 °C), and weighed to determine recovery efficiency. The recovery yield of each component was calculated as a percentage of the original dry mass of the sample according to the following formula.

$$E = \left(\frac{\sum m_{separated\ components}}{m_{initial\ sample}}\right) \times 100\%$$

where $\sum m_{\text{separated components}}$ is the total mass of separated fractions after the separation process, and $m_{\text{initial sample}}$ is the initial waste mass (g), providing a metric to assess the effectiveness of the separation process.

3 Results and discussion

3.1 Recovery of aluminum and plastic from pharmaceutical blisters

As shown in Fig. 3a, the recovery efficiency of aluminum and plastic was significantly influenced by the solid-to-liquid (S/L) ratio. At low ratios of 1:5 and 1:10, the recovery was negligible (1.1 and 5.3%, respectively). A substantial increase in recovery was observed at a ratio of 1:15 (75.4%), reaching 85.3 and 94.0% at 1:20 and 1:25, respectively. However, further increasing the ratio to 1:30 slightly reduced the recovery to 90.7%. The optimal S/L ratio was thus determined to be 1:25, where the effective separation yielded approximately 11% aluminum and 83% PVC by mass.

Fig. 3b illustrates the effect of NH_3/NH_4Cl concentration on the recovery process. At low concentrations (1-1 M), the recovery was only 44.7%. Higher concentrations improved performance: 73.2% at (5-5) M, 89.8% at (3-1.5) M, and a maximum of 94.0% at (5-2.5) M. Slight reduction occurred at (6-3) M (92.2%), suggesting that excess NH_4Cl may not further enhance separation. Therefore, the optimal concentration was 5 M NH_3 and 2.5 M NH_4Cl.

As depicted in Fig. 3c, a 1:1 volume ratio of 5 M NH_3 to 2.5 M NH_4Cl resulted in a 62.7% recovery. Increasing the proportion of NH_4Cl reduced the recovery to only 12.0%. In contrast, a 2:1 ratio yielded the highest recovery of 99.5%. NH_4Cl plays a role in dissolving the aluminum oxide formed during the reaction with NH_3, helping to clean the plastic surface and improve plastic recovery. The following reaction scheme is proposed as a tentative mechanism that may occur under the elevated temperature (70 °C), high pH, and chloride-rich conditions of the NH_3/NH_4Cl system:

$$Al_2O_3\ (\text{surface}) + 6NH_4Cl \rightleftharpoons 2Al^{3+}\ (\text{complex}) + 6NH_3 + 3H_2O \quad (1)$$

$$Al^{3+} + 6Cl^- \rightleftharpoons [AlCl_6]^{3-}\ \text{or}\ AlCl_3\ (\text{soluble}) \quad (2)$$

$$Al^{3+} + 3NH_3 + 3H_2O \rightleftharpoons Al(OH)_3\downarrow + 3NH_4^+ \quad (3)$$

These reactions are not direct bulk reactions but rather represent possible surface complexation and ligand exchange processes that facilitate the partial release of aluminum from aluminum oxide under mildly hydrothermal, alkaline, and ammoniacal conditions. Similar reaction behaviour has been reported in aluminum leaching studies with phosphoric acid or chloride-ammonia solutions [9, 23].

It should be noted that the proposed reaction mechanism is hypothetical and supported by indirect experimental evidence, such as the detection of dissolved aluminum and the improved separation performance with NH_4Cl. While these findings suggest a plausible surface interaction between Al_2O_3 and chloride ions, confirmation of the exact pathway would require further investigation using *in-situ* spectroscopic or kinetic analysis techniques.

The recovery slightly declined at higher NH_3/NH_4Cl ratios of 3:1 (94.0%) and 4:1 (93.6%). Therefore, the optimal ratio was 2:1 (5 M NH_3/2.5 M NH_4Cl), yielding 0.22 g aluminum (11%) and 1.75 g PVC (87.5%). The amount of dissolved aluminum was 0.03 g, accounting for 13.6% of the total recovered aluminum and 1.5% of the initial blister weight.

As illustrated in Fig. 3d, soaking time significantly affected separation efficiency. When

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the soaking time was 15 and 30 minutes, recoveries attained 5.7 and 41.5%, respectively. The maximum recovery of 99.5% was obtained after 60 minutes. A slight decline to 96.2% when soaking for 90 minutes may result from the fact that dissolved aluminum adhered to the plastic surface. Thus, 60 minutes was found to be the optimal soaking time.

Although the proposed mechanism requires further validation, the experimental results lend indirect support to its plausibility. Under the optimal NH_3/NH_4Cl conditions, both aluminum (10.7%) and PVC (88.8%) were effectively recovered, with clear physical separation between the components. The role of NH_4Cl appears critical in modifying the aluminum oxide interface, likely through surface complexation or ligand exchange reactions that weaken adhesion to the polymer. By contrast, treatments using NH_4OH alone yielded significantly lower separation efficiency, suggesting that chloride-mediated reactions play a key role in delamination. Additional evidence from material characterisation (discussed in Section 3.3) reinforces this interpretation, indicating that the aluminum surface was free of organic residues and that polymer identity was preserved post-separation.

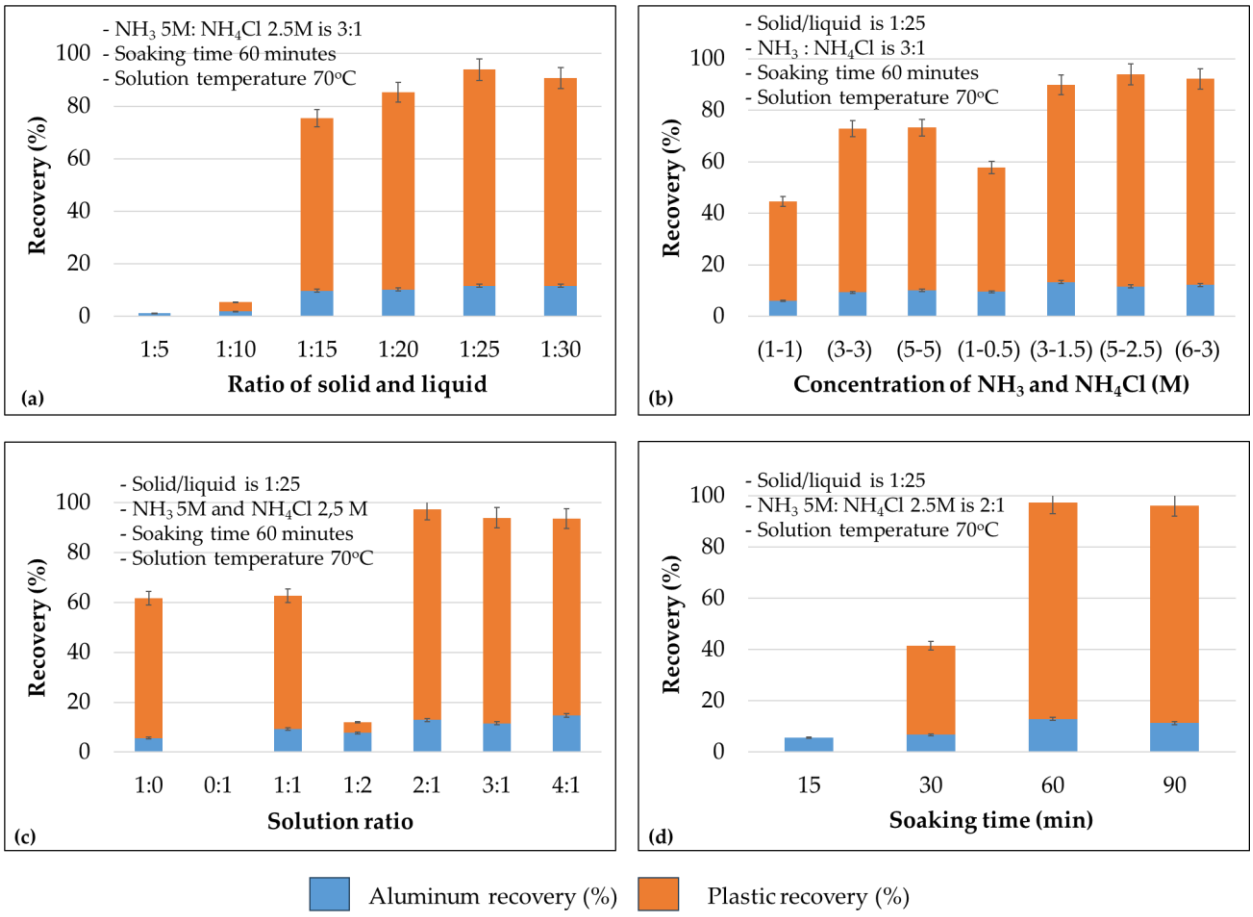

Fig. 3. Recovery of aluminum and plastic separation at different solid-liquid ratios (a); solution concentrations (b); solution ratios (c); soaking times (d)

These findings demonstrate that the $NH_3/NH_4Cl/H_2O$ system offers an effective chemical route for delaminating pharmaceutical blister packs under relatively mild conditions. Compared with other studies, such as Wang et al. (2015) [9] using HCl and Shukla et al. (2024) using H_3PO_4 [23], which also achieved complete aluminum separation, our method avoids the need for acid recovery steps or strong oxidants. Additionally, unlike organic solvent-based systems (e.g., DMCHA [24] and acetone-isopropanol [6]) that require more complex solvent handling, our approach uses aqueous reagents with moderate environmental impacts and no VOCs. While the aluminum recovery yield (11%) is slightly lower than that in other methods, the overall process maintains a high plastic recovery (87.5%) and avoids degradation, making it a promising alternative for scalable and environmentally friendly blister pack recycling.

3.2 Separation efficiency of components from milk cartons under soaking conditions

While various alkaline solutions have been explored for delaminating multilayer packaging materials, ammonium hydroxide (NH_4OH) was selected as the sole treatment agent for milk cartons because of its moderate reactivity and better preservation of the paper component. Preliminary trials using a combination of NH_4OH and NH_4Cl—effective in blister packs separation—resulted in poor recovery efficiency and excessive fragmentation of the paper layer, rendering it largely unrecoverable. Consequently, subsequent experiments employed only NH_4OH to enhance selectivity and minimise damage to the recovered fractions.

The separation efficiency of components from milk cartons—namely paper, plastic, and aluminum-plastic fractions—was systematically evaluated under varying treatment conditions, including NH_4OH concentration, reaction time, temperature, and solid-to-liquid ratio. Experimental results show that aluminum remained largely undissolved in the solution, while the paper layer was partially released during soaking and could be effectively separated through flotation. Minimal plastic detachment was observed, and the aluminum-plastic laminate exhibited strong interfacial adhesion, resisting further chemical delamination. For recovery assessment, the separated materials were categorised into two distinct fractions:

(1) Paper recovered, collected from the lower flotation phase, washed, dried, and weighed

(2) Aluminum-plastic composite recovered, collected from the froth layer, dried, and weighed.

The recovery efficiency of each component was determined gravimetrically and expressed as a percentage of the initial dry mass of the milk carton sample.

Initially, the effect of NH_4OH concentration was examined at 70 °C with stirring and a fixed ratio of 1:25 (2 g sample in 50 mL solution) (Fig. 4a). At low concentrations (1 and 3 M), the recovery of aluminum-plastic fraction was limited to 30.4 and 21.6%, respectively, while the paper recovery was 34.3 and 48.8%. Increasing the NH_4OH concentration to 5 M significantly improved the separation efficiency to 97.8%, with aluminum-plastic fraction representing 34.1% (0.68 g) and paper 63.7% (1.3 g) of the total mass. Minimal aluminum dissolution into the solution was detected, whereas plastic remained insoluble. The composition of separated components closely matched Tetra Pak data, establishing 5 M NH_4OH as the optimal concentration.

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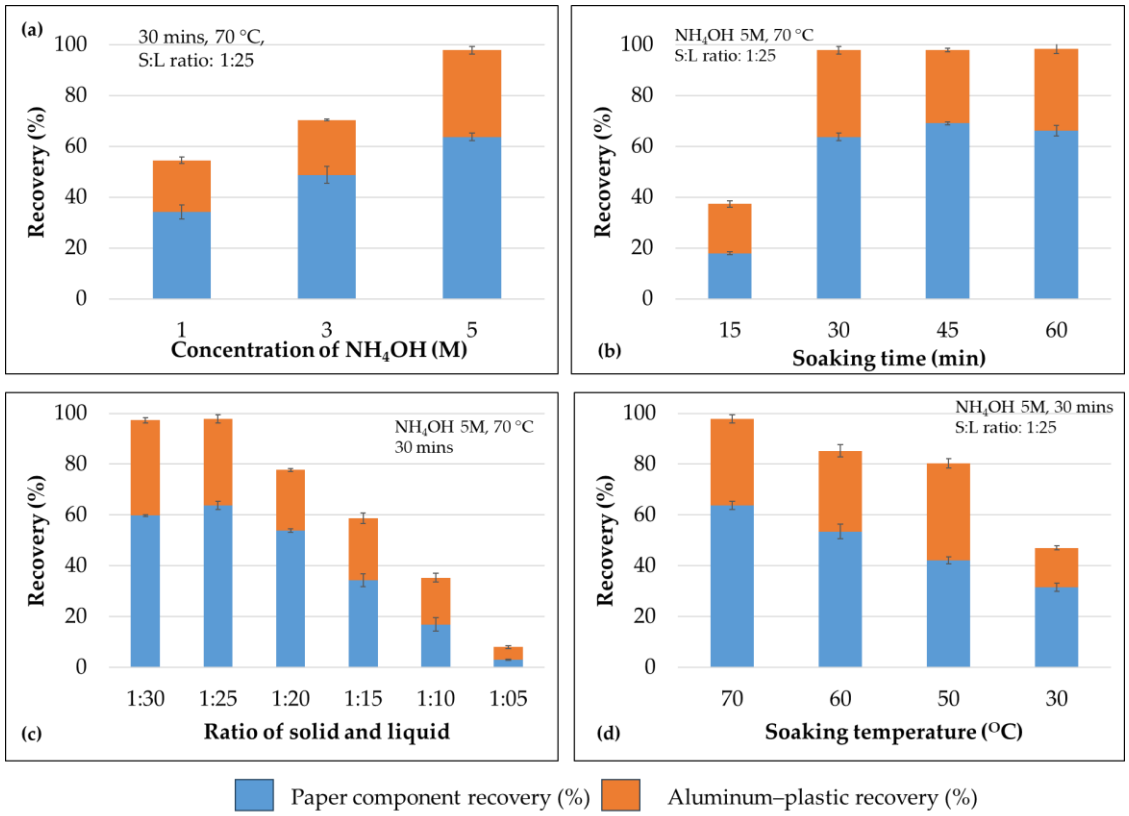

Fig. 4. Recovery efficiency of paper and aluminum-plastic components from Tetra Pak cartons under various conditions using NH_4OH.: (a) Effect of NH_4OH concentration; (b) Effect of soaking time; (c) Effect of solid-to-liquid (S/L) ratio; (d) Effect of soaking temperature

Next, soaking time was varied from 15 to 60 minutes under the optimised conditions (5 M NH_4OH, 70 °C, 1:25 ratio) as shown in Fig. 4b. The separation efficiency increased from 37.4% at 15 minutes to 97.8% at 30 minutes, with only marginal improvement beyond this point (98% at 45 minutes and 98.4% at 60 minutes). Thus, 30 minutes was determined as the optimal reaction time. The influence of the sample-to-solution ratio was evaluated with values ranging from 1:5 to 1:30 (g/mL), maintaining other optimal conditions. The efficiency rose from 8.06% at 1:5 to a peak of 97.8% at 1:25, and beyond, the gains were negligible (97.3% at 1:30). Therefore, 1:25 was selected as the optimal ratio (Fig. 4c).

Temperature effects were studied between 30 and 70 °C, showing separation efficiencies of 47, 80.3, 85.2, and 97.8%, respectively, confirming 70 °C as the optimal temperature (Fig. 4d).

Overall, the optimal conditions for separating milk carton components were identified as 5 M NH_4OH concentration, 70 °C temperature, 30 minutes reaction time, and 1:25 sample-to-solution ratio. To assess separation efficiency, the solid residues after NH_4OH soaking were subjected to froth flotation. Separated fractions were naturally dried and weighed. Under these optimised conditions, approximately 97.8% of the initial mass was recovered, consisting of 63.7% paper, 24.1% bonded aluminum-plastic laminate, and only 7.8% free plastic film. No fully detached aluminum foil was observed, and HDPE layers remained mostly bonded.

The primary limitation arises from the interfacial adhesion between aluminum and polyethylene, formed through high-temperature extrusion lamination in Tetra Pak manufacturing

[25]. This adhesion is structurally different from the binding mode in pharmaceutical blister packs, which feature an intermediate oxide layer that can undergo ligand exchange with ammoniacal complexes. The absence of such a reactive interlayer in Tetra Pak significantly limits chemical separation under mild alkaline conditions. The FTIR analysis of the aluminum-plastic residue (discussed in Section 3.3) confirmed the presence of both polyethylene and aluminum oxide functional groups, indicating that the bonded interface persisted after treatment.

Thus, the use of NH_4OH alone did not yield complete delamination of the aluminum-plastic laminate, and the overall recovery performance reflects this inherent structural constraint.

Alternative approaches for separating the aluminum-polyethylene layers have been reported with promising outcomes. For instance, Yan et al. (2015) employed formic acid (HCOOH) to achieve complete delamination in less than 30 minutes, utilising the acid's capacity to induce nanoscale cracking on the polyethylene surface and dissolve aluminum oxides, resulting in an aluminum recovery purity of 87.2% [26]. Similarly, Zhang et al. (2014) demonstrated that a mixed organic solvent system of benzene-ethanol-water could separate aluminum-plastic laminates in just 6 minutes, facilitated by the solvents' complementary solubility parameters [27]. Recent innovations, such as switchable hydrophilicity solvents like DMCHA, have demonstrated aluminum recovery rates exceeding 99% recovery of aluminum and >80% recovery of polyethylene under mild conditions (<100 °C), while maintaining material integrity and minimising environmental impact [28]. Though bio-based solvents like CPME and 2-MeTHF offer greener alternatives [1], some (e.g., biodiesel) were limited because of high viscosity and contamination of recovered materials. The present study emphasises the need for interface-specific separation strategies when dealing with complex multi-layer packaging and suggests that ammonium hydroxide, though selective and mild, may require a combination with other treatment steps or alternative reagents for complete material recovery from Tetra Pak waste.

These findings underscore the importance of tailoring separation strategies to the interfacial structure of each multilayer packaging type and suggest that future work should explore hybrid or sequential treatments that combine mild alkaline conditions with targeted interfacial modifiers.

3.3 Effects of sample sizes and component properties on separation of multilayer packaging waste

Under the optimised separation conditions (2:1 NH_3 5 M/NH_4Cl 2.5 M ratio, 70 °C temperature, 1:25 (g/mL) solid-to-liquid ratio, and 60 minutes reaction time, the effect of sample size was evaluated for pharmaceutical blisters (Table 2). The highest total recovery (99.6%) was achieved with the 1 × 1 cm samples, corresponding to 10.9 ± 0.4% aluminum and 88.7 ± 2.7% plastic recovery. As particle size increased to 2 × 2 cm, the efficiency dropped markedly to 79.2%, with the plastic recovery decreasing to 69.7 ± 2.2%. At 3 × 3 cm, the separation efficiency plummeted to just 4.7%, with negligible plastic recovery. This indicates that a smaller sample size facilitates more effective delamination and separation of aluminum and PVC layers, likely because of enhanced chemical accessibility and mechanical exposure.

A similar trend was observed for Tetra Pak cartons under optimised conditions (5 M NH_4OH, 70 °C, 1:25 solid-to-liquid ratio, and 30 minutes). The 1 × 1 cm samples yielded a total recovery of 97.8%, including 34.1 ± 1.5% aluminum-plastic

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fraction and 63.7 ± 1.6% paper. Larger sample sizes (2 × 2 cm and 3 × 3 cm) resulted in lower total recoveries of 94.9% and 87.5%, respectively, primarily because of reduced recovery of the paper component. Notably, aluminum-plastic recovery remained relatively stable across sizes, suggesting the paper layer is more susceptible to the effects of reduced chemical accessibility in larger samples. These findings clearly establish the 1 × 1 cm sample as the optimal particle size for achieving high separation efficiency under the specified chemical and thermal conditions. The enhanced efficiency at this size supports its use for maximising material recovery in practical recycling applications involving multilayer packaging waste.

Table 2. Effect of sample size on separation efficiency of multilayer packaging materials

Pharmaceutical blisters

Sample sizes (cm)	Aluminum recovery (%)	Plastic recovery (%)	Recovery efficiency (%)
1 × 1	10.9 ± 0.4	88.7 ± 2.7	99.6
2 × 2	9.5 ± 1.6	69.7 ± 2.2	79.2
3 × 3	4.7 ± 0.3	0.0	4.7

Tetra Pak milk carton

Sample sizes (cm)	Aluminum-plastic recovery (%)	Paper component recovery (%)	Recovery efficiency (%)
1 × 1	34.1 ± 1.5	63.7 ± 1.6	97.8
2 × 2	35.9 ± 1.1	59.0 ± 1.3	94.9
3 × 3	34.6 ± 0.6	52.9 ± 1.8	87.5

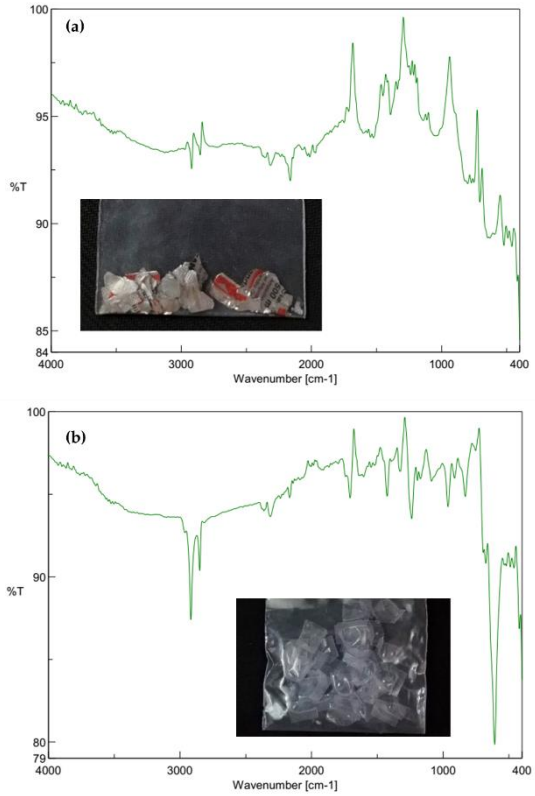

Fig. 5. FTIR and accompanying images of aluminum layer (a) and plastic (b) component from separation of pharmaceutical blisters under optimum conditions

The FTIR analysis provided further evidence supporting the effectiveness of the NH_3/NH_4Cl system in delaminating aluminum-plastic laminates. As shown in Fig. 5a, the spectrum of the recovered aluminum foil displayed strong absorptions in the 400–800 cm^{-1} region, characteristic of Al–O lattice vibrations in aluminum oxide (Al_2O_3) [29]. Notably, the absence of peaks in the 2800–3000 cm^{-1} range—where C–H stretching of residual organic materials typically appears—indicates that the aluminum surface was effectively stripped of polymer layers during treatment. Conversely, the recovered plastic layer (Fig. 5b) exhibited distinct bands corresponding to polyvinyl chloride (PVC), including C–Cl stretching at 616 cm^{-1}, CH_2 bending at 1427 cm^{-1}, and CH_2 rocking at 966 cm^{-1} [6, 10, 30], confirming successful material separation.

These spectroscopic results not only validate the effectiveness of the separation process but also offer indirect support for the proposed chemical interaction between Al_2O_3 and NH_4Cl discussed in Section 3.1. The clean Al–O bands and absence of C–H stretching in the aluminum spectrum suggest that surface-bound polymers were effectively removed, likely facilitated by chloride-mediated surface reactions. This observation aligns with the hypothesised mechanism wherein NH_4Cl, in the presence of NH_3, promotes surface complexation and ligand exchange with aluminum oxide, thereby weakening the adhesion between aluminum and PVC layers and enhancing delamination. The contrast between the clean aluminum surface and the intact PVC signature in the plastic spectrum further substantiates the selectivity of the process.

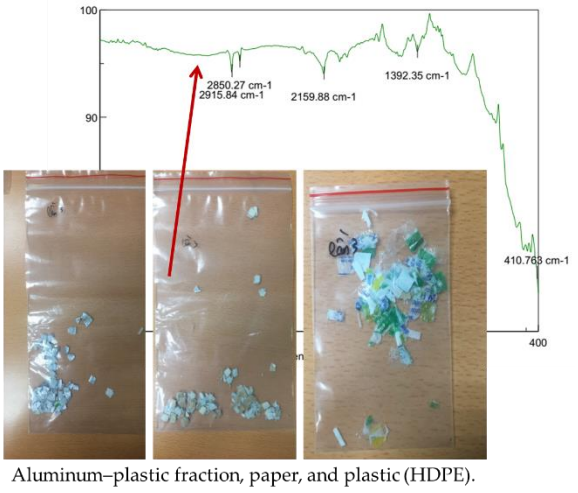

Fig. 6. From left to right: aluminum-plastic, paper, and HDPE plastic layers from Tetra Pak milk cartons and FTIR of aluminum-plastic fraction

For the separated components of milk cartons the FTIR spectrum of the aluminum-plastic component (Fig. 6) indicates characteristic absorption bands observed at 2915 and 2850 cm^{-1}, corresponding to C–H stretching vibrations, while the band at 1392 cm^{-1} is attributed to CH_3 bending [16, 25]. These peaks are consistent with the vibrational modes of polyethylene. Additionally, a band at 410 cm^{-1} may be associated with inorganic lattice vibrations from the aluminum-oxide substrate [29]. The presence of both organic (PE) and inorganic (aluminum oxide) features in the spectrum suggests that the interface between the polymer layer and the aluminum foil remained intact after the separation attempt, indicating incomplete delamination at this interface. This result underscores a well-documented challenge in recycling composite packaging, particularly Tetra Pak, where strong interfacial bonding between aluminum and polyethylene, formed via high-temperature extrusion, prevents effective delamination under mild chemical conditions, such as NH_4OH treatment.

This study reinforces the importance of matching reagent systems to material interface characteristics and highlights that a "one-size-fits-all" approach may be inadequate for multi-layered packaging waste. Tailored separation protocols that consider both technical efficiency and sustainability metrics will be a key to advancing circular solutions in post-consumer packaging recovery.

In particular, the aqueous NH_3/NH_4Cl system demonstrated not only effective delamination for blister packs but also potential for reagent reuse and low environmental impact, given its mild conditions and absence of volatile organic solvents. The recovered materials (aluminum, PVC, paper) preserved their chemical integrity and physical form, supporting their recyclability in conventional recycling streams.

Future research should continue to focus on developing cost-effective, environmentally benign, and industrially scalable processes that can address the complex adhesive chemistries of multilayer packaging, ensuring higher material recovery yields and fostering a more circular economy for these challenging waste streams.

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4 Conclusions

This study demonstrates the efficacy of ammonia-based chemical treatments for the delamination of multilayer materials from pharmaceutical blister packs and milk cartons—two widely used packaging—that pose significant recycling challenges because of strong interfacial adhesion. The $NH_3/NH_4Cl/H_2O$ system enabled complete separation of aluminum and PVC from blister packs, achieving recovery yields of 10.7% aluminum and 88.8% plastic by mass, confirming both the feasibility and selectivity of the proposed method. In contrast, the application of NH_4OH to milk cartons resulted in partial separation, with an overall recovery efficiency of 97.8%. While the paper core was successfully isolated, HDPE films were recovered only in negligible amounts, and the aluminum-plastic laminate remained largely inseparable because of robust extrusion bonding between aluminum and polyethylene.

In addition to separation efficiency, this study considered the economic and environmental implications of the process. The aqueous NH_3/NH_4Cl system operates under mild conditions and does not involve volatile organic solvents, offering potential for closed-loop reagent reuse and reduced environmental impact. Furthermore, the recovered aluminum, plastic, and paper fractions retained their chemical identity and physical integrity, indicating compatibility with conventional recycling streams.

These findings highlight the potential of ammonia-based processes to enhance material recovery from multilayer packaging waste in alignment with circular economy principles. Future work should focus on improving the delamination of high-adhesion interfaces such as aluminum-PE in milk cartons and on integrating hybrid or sequential treatment strategies that ensure both technical performance and long-term sustainability.

Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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