



Recycling of multilayer packaging waste with switchable anionic surfactants

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ABSTRACT

Switchable Anionic Surfactants (SAS) were used for delaminating flexible packaging waste composed of various plastic layers and aluminium, thereby promoting the recycling of such waste streams from a circular economy perspective. The delamination protocol was optimized on de-pulped food and beverage cartons containing low-density polyethylene (LDPE) and aluminium, varying the carboxylic acid and its counterion constituting the SAS (C₈–C₁₈ carboxylic acids as the anionic part; inorganic bases and primary, secondary and tertiary amines as the cationic one) their molar ratio (carboxylic acid: base molar ratio from 1:1 to 1:3), SAS concentration (0.15, 0.3 and 0.5 wt%), time (0.5–3 h) and material weight in input (1–10 wt%). High-quality LDPE and aluminium were separated and recovered by using a diluted solution of a surfactant based on lauric acid and triethanolamine (C₁₂-TEA), with performances not achievable with other anionic or cationic surfactants available on the market. The C₁₂-TEA solution was then applied to a large variety of multilayer waste materials composed of polypropylene and aluminium, polyolefins/polyethylene terephthalate/aluminium, giving a material separation dependant on the structure and composition of the material in input. At the end of the process, lauric acid was recovered from the aqueous solution used for washing the separated materials by tuning its water solubility with CO₂.

1. Introduction

Amongst plastic packaging waste to be managed at the end of life, multilayer materials are particularly widely diffused (about 2 Mt/y in the EU, Kaiser et al., 2018) but challenging. Even if the combination of several polymeric layers is essential to attain the technical performances of the packaging, the uncertainty about the variable number of layers, their thickness and composition, the presence of glues or additives, and the presence of aluminium, in turn, variable in the mode of deposition and amount, decrease the rate of success of layers separation and recycling. To date, this complex kind of waste is still mainly incinerated or landfilled since mechanical recycling is not possible (Vollmer et al., 2020), and such approaches leave undeniable room for improvement directed to increase the sustainability of the End of Life (EoL)

management (De Mello Soares et al., 2022; Zawadiak et al., 2017). The recycling/upcycling of multilayer packaging materials is crucial from a circular economy perspective but challenging, above all because the quality of the recycled polymers and aluminium should meet the market needs (e.g. high molecular weight, transparent colour and good mechanical properties for polymers, avoided oxidation of aluminium) (Barkane et al., 2023). To achieve this goal, two approaches can be adopted: i) solubilizing the polymeric layers with organic solvents (Georgiopoulou et al., 2021; Samori et al., 2023; Vollmer et al., 2020), the so-called Selective Dissolution-Precipitation (SDP) process; ii) delaminating the multilayer material using solvents that can remove the adhesive/tie layer between the polymeric layers, or with aqueous solutions of suitable additives, like surfactants, that can swell the layers or solubilize the adhesive as well (Ügdüler et al., 2021, 2022).

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Herein we investigated the delamination ability of long-chain carboxylates that can act as surfactants in neutral/alkaline aqueous environments in the presence of organic/inorganic counterions and become water-insoluble upon the addition of CO₂ and the consequent pH decrease (Ceschia et al., 2014; Chen et al., 2017; Samori et al., 2015; Xu et al., 2015). These surfactants are known as Switchable Anionic Surfactants (SAS), and, differently from the other anionic surfactants, have the undeniable advantage to be recoverable from the aqueous solution by exploiting their pH-dependant solubility behaviour. The use of CO₂ as a trigger to reversibly switch the properties of chemicals like solvents, surfactants, and additives, is an elegant approach developed in the last years to bypass some energy-intensive steps that characterize many syntheses and processes (Jessop et al., 2010, 2011, 2012; Phan and Jessop, 2009; Vanderveen et al., 2014). Switchable solvents like *N,N*-dimethylcyclohexylamine (DMCHA)/*N,N*-dimethylcyclohexylammonium hydrogen carbonate, and 3-cyclopentylpropionic acid/sodium 3-cyclopentylpropanoate have been used for polymer recycling in SDP processes exploiting their capability of solubilizing some of the polymers present in mono-material or multilayer packaging waste like polystyrene (Cunha et al., 2022), polyethylene (Mumladze et al., 2018; Samori et al., 2017), or poly(butyl methacrylate) (Su et al., 2017) when they are in the hydrophobic neutral form, and to separate when they are in the hydrophilic ionic form after CO₂ addition. Switchable solvents have also been used in the delamination of complex multilayer materials like pharmaceutical blisters and food and beverage cartons exploiting the solubilization of the adhesives present between the polymeric layers and thus promoting the separation of aluminium, paints and ink from the polymeric layers (Mumladze et al., 2018; Yousef et al., 2018). To the best of our knowledge, these two approaches described so far in the literature (SDP process and delamination) foresee the use of pure solvents that could have concerning safety issues and create risks at the industrial scale (Yousef et al., 2018). On the contrary, the diluted aqueous solutions of SAS that were here used contain only inherently safe compounds. Aqueous solutions of short and long-chain carboxylic acids (e.g. formic and acetic acid, and hexanoic and decanoic acids) have been already industrially used in combination with inorganic acids and/or swelling agents for the delamination of a broad range of multilayer structures containing aluminium (Lovis et Schulze, 2022; Ügdüler et al., 2021). Other studies on laminated packaging materials containing polyethylene and aluminium suggest that delamination occurs when long-chain carboxylic acids penetrate through the polymer layer and accumulated in the polymer-aluminium interface, creating a weak boundary layer by acid-base interactions between the carboxylic acid and the aluminium oxide present on the surface of the aluminium foil (Olafsson and Hildingsson, 1995). Such a weak boundary layer decreases the acid-base and van der Waals adhesive forces that occur between aluminium oxide and oxidized groups on the polymer surface (aldehydes, ketones, and carboxylic acids), formed when the polymer is extruded at elevated temperatures.

SAS solutions were here obtained from the reaction of mono-carboxylic acids ranging from C₈ to C₁₈ (caprylic, capric, lauric, myristic, and oleic), with alkyl amines (monoethanolamine MEA, diethanolamine DEA, *N*-ethylbutylamine EBA, and triethanolamine TEA) or inorganic bases (NaOH, KOH, and NH₄OH). The obtained aqueous surfactant solutions have been applied to a variety of multilayer packaging waste containing aluminium (metalized packaging) widely diffused in the food and beverage industry, firstly focusing on de-pulped food and beverage cartons containing low-density polyethylene (LDPE) and aluminium (i.e. LDPE/aluminium), and then extending the optimized delamination protocol to coffee packaging bags composed of PE, aluminium and polyethylene terephthalate (PET) (i.e. PE/aluminium/PET), to other packaging waste containing PE or polypropylene (PP) (i.e. polyolefins, PO), aluminium and PET (i.e. PO/aluminium/PET) and to PP and aluminium multilayer materials (i.e. PP/aluminium).

2. Materials and methods

2.1. Surfactant preparation and switching procedure

Surfactant preparation (Table S1 in ESI) was performed by mixing carboxylic acid (1 eq.) and base (amine or hydroxide, 1.5 eq.) at 60 °C in water for 1 h. The amounts of carboxylic acid and base in the mixture necessary to reach a concentration of surfactant (RCO₂⁻base⁺) of 0.5 wt % are reported in Table S2 in ESI. After that, CO₂ was bubbled into the system at ambient pressure for 1 h by keeping the flask in an ice bath to enhance CO₂ absorption, and the carboxylic acid was filtered off from the aqueous solution, dried overnight under vacuum and weighed. The recovery of the carboxylic acid (%) was reported in Table S2 in ESI. Other carboxylic acid: base molar ratios (1:1.2 for K- and Na-surfactants; 1:1, 1:1.1, 1:2, 1:2.5 and 1:3 for C₁₂-TEA) were also tested. The pH of the aqueous solutions before and after CO₂ addition was measured for each combination of carboxylic acids and bases.

2.2. Delamination of LDPE/aluminium waste with SAS aqueous solutions

The waste derived from multilayer cartons consisting of paper, LDPE and aluminium, widely used as aseptic packages in the food and beverage industry (Tetra Pak®), was kindly provided by Lucart S.p.A., a paper mill located in Italy. Full characterization of this material was reported in our previous work (Samori et al., 2023) and it is summarized here: i) the material did not contain the paper layer, removed through hydropulping by Lucart for recycling the cellulose fibers; ii) on average the amounts of LDPE (estimated by elemental analysis) and aluminium (estimated by calcination) were 70.8 ± 7.2% and 26.9 ± 4.1%, respectively; iii) no distinct layers of adhesive or glue were identified between LDPE and aluminium layers by ATR analyses and UV-Vis microscopy of the samples embedded in KBr and cross-sectioned; iv) the material did not apparently contain any organic or inorganic ink since, typically, the original post-consumer food and beverage cartons have the ink on the paper layer, removed by the paper mill.

LDPE/aluminium waste (1 g) was charged in a round bottom flask equipped with a condenser with 100 mL of the aqueous solutions containing capric (C₁₀) or lauric (C₁₂)-based SAS (0.5 wt%, 1.5 eq. of base) or the chosen commercial surfactants (sodium dodecyl sulfate, cetyltrimethylammonium bromide, Tween® 20 and Triton™ X-100, representative of the classes of anionic, cationic and non-ionic surfactants, respectively). The system was heated at 90 °C for 2 h; in the cases of C₁₂-NH₄, C₁₂-MEA, C₁₂-DEA, and C₁₂-EBA, ultrasounds were furtherly applied for 1 h to accelerate and improve the separation of the samples that were not delaminated after 2 h at 90 °C. In the optimization phase with C₁₂-TEA, the effect of time, amount of TEA (1, 1.1, 2, 2.5 and 3 eq.) and surfactant concentration (0.15, and 0.3 wt%) was investigated (Table 1), while the ratio waste/aq. solution was kept constant (1 wt%).

At the end of the process, delaminated LDPE layers floated and they were manually recovered from the solution, while the precipitated aluminium was filtered. When partial delamination of the waste was obtained, residual LDPE/aluminium ended up on the bottom of the flask together with aluminium; in these cases, not delaminated LDPE/aluminium pieces were manually separated from aluminium after filtration. Recovered LDPE and aluminium were washed with an alkaline NH₄OH (20 mM) solution and then dried overnight under vacuum. The amount of the recovered LDPE and aluminium was expressed based on the amount of the LDPE/aluminium multilayer waste in input (w/w, %). In the case of the delamination with C₁₂-TEA, a washing of the recovered materials with a 20 mM TEA solution was applied to remove residual lauric acid that could eventually be then precipitated through CO₂ bubbling of the washing solution at ambient pressure for 1 h.

2.3. Characterization of the recovered aluminium, LDPE and lauric acid

The quantification of LDPE residues on the recovered aluminium was

Table 1

LDPE amount (% on the input material weight basis, w/w) recovered with an aqueous solution of various surfactants (+/- indicates the partial delamination of LDPE). The data are expressed as the mean of four independent replicates of delamination with each surfactant \pm standard deviation.

Entry	Surfactant	Recovered LDPE amount (%)
1	C ₁₀ -Na	-
2	C ₁₀ -NH ₄	33 \pm 3 (+/-)
3	C ₁₀ -MEA	26 \pm 1 (+/-)
4	C ₁₀ -DEA	26 \pm 2 (+/-)
5	C ₁₀ -EBA	-
6	C ₁₀ -TEA	70 \pm 2
7	C ₁₂ -K	-
8	C ₁₂ -NH ₄ ^a	76 \pm 1
9	C ₁₂ -MEA ^a	41 \pm 1 (+/-)
10	C ₁₂ -DEA ^a	42 \pm 1 (+/-)
11	C ₁₂ -EBA ^a	24 \pm 3 (+/-)
12	C ₁₂ -TEA	73 \pm 4
13	SDS	-
14	CTAB	-
15	Tween 20	-
16	Triton X-100	-

^a 2 h at 90 °C, followed by 1 h of ultrasounds.

performed through calcination at 550 °C for 5 h, attributing the weight loss to both residual LDPE and lauric acid/laurate residues. X-ray photoelectron spectroscopy (XPS) depth profiling analyses of the recovered aluminium were carried out by using an ESCALAB 250 Xi spectrometer (ThermoFisher Scientific, UK), equipped with a monochromatic Al X-ray source and a 6-channeltrons detection system. The spectra were collected in magnetic lens standard mode and constant pass energy of 50 eV. The base pressure in the analyser chamber was $p = 1 \times 10^{-10}$ mbar and it was increased to $p = 1 \times 10^{-8}$ mbar during the depth profile. The depth profile was performed with alternating spectra acquisition and cycles of ion (Ar⁺) sputtering by EX06 ion source. The binding energy scale was calibrated positioning the adventitious carbon contribution (before ion sputtering) at BE = 285.0 eV. The accuracy of the binding energy (BE) scale was ± 0.1 eV. All samples were attached to the sample holder by a metallic clip. All data were acquired and processed by Avantage software v.5.979

Hi-ResTM Thermogravimetric analyses (TGA) of the recovered LDPE were carried out using a TGA Q500 thermogravimetric analyser (TA Instruments) from room temperature to 600 °C, at a starting heating rate of 50 °C min⁻¹, and a resolution index of 4 and a sensitivity index of 1, under airflow. The onset decomposition temperature was defined as the temperature at 5% weight loss (T_d , 5%), while the % residue at 600 °C was used to determine the inorganic impurities present on the recovered LDPE. Thermal transitions were measured using a differential scanning calorimeter (DSC Q2000; TA Instruments), equipped with a refrigerated cooling system (RCS). Samples, under nitrogen flow, were subjected to a first heating scan at 20 °C min⁻¹, from -90 to 155 °C, to erase the polymer thermal history. The samples were then cooled at 10 °C min⁻¹ and a second heating scan was applied. The melting temperature (T_m) was taken at the peak maximum of the melting endotherm from the second heating curve. The crystallinity degree (X_c , %) was calculated as follows:

$$X_c = 100 \times \frac{\Delta H_m}{\Delta H^0}$$

where ΔH_m is the melting enthalpy of the sample (J g⁻¹) and ΔH^0 is the melting enthalpy of 100% crystalline polymer (J g⁻¹). For LDPE, ΔH^0 was considered equal to 294 J g⁻¹ (Fischer and Hinrichsen, 1966).

Tensile stress-strain measurements were carried out on LDPE films produced by compression moulding starting from the recovered LDPE; LDPE manually removed from the waste in input to the process was analysed for comparison. The polymer was placed between two polytetrafluoroethylene plates, with an appropriate spacer, at 150 °C for 1

min under a pressure of 2 ton m⁻² (Carver C12, laboratory press). An Instron Tensile Testing Machine 4465 was used at an extension rate of 5 mm min⁻¹ for the mechanical testing of dog-bone specimens (width 5 mm, gauge length 20 mm) die-cut from hot-pressed samples (180 °C, film thickness 0.2 mm). The average specimen thickness, measured by using a digital micrometre, was used to construct the stress-strain curves from the raw load-displacement data. The stress was measured with a standard 100 N cell while the deformation was precisely monitored. The elastic modulus was calculated from the initial slope of the stress-strain curve. The statistical significance of the differences amongst the tensile stress-strain values was tested by one-way analysis of variance (ANOVA). Once ANOVA resulted significantly, Dunn's post-hoc test was carried out to identify which LDPE samples were significantly different from each other.

Lauric acid and laurate residues on the recovered LDPE and aluminium were quantified by placing samples of both (6–8 mg) in ethyl acetate (0.1 mL), adding pyridine (0.04 mL) and BSTFA-1%TMCS (*N,O*-bis(trimethylsilyl)trifluoroacetamide with 1% trimethylchlorosilane, 0.08 mL), and heating at 80 °C for 1 h. The same approach was applied to analyse the purity of lauric acid recovered from the washing solution containing TEA.

The analysis of the silylated solutions was performed by GCMS—using an Agilent HP 6850 gas chromatograph connected to an Agilent HP 5975 quadrupole mass spectrometer. Analytes were separated on an HP-5MS fused-silica capillary column (stationary phase poly[5% diphenyl/95% dimethyl]siloxane, 30 m, 0.25 mm i.d., 0.25 μ m film thickness), with helium as the carrier gas (at constant pressure, 36 cm s⁻¹ linear velocity at 200 °C). Mass spectra were recorded under electron ionization (70 eV) at a frequency of 1 scan s⁻¹ within the 12–600 *m/z* range. The injection port temperature was 250 °C. The temperature of the column was initially kept at 50 °C for 5 min, then increased from 50 to 325 °C at 10 °C min⁻¹. A calibration curve prepared with silylated lauric acid was used for the quantification.

2.4. Delamination of different multilayer waste with aqueous solutions of C₁₂-TEA

C₁₂-TEA aqueous solutions at various concentrations (0.15, 0.3 and 0.5 wt%) and with different amounts of TEA (1.5, 2, 2.5 and 3 eq.) were used for delaminating three classes of other multilayer materials containing aluminium: PE/aluminium/PET used for coffee packaging bags, PO/aluminium/PET and PP/aluminium for food and pet food packaging. The ratio waste/aq. solution was 1 wt% for all the materials in input; a ratio of 10 wt% was tested just for PE/aluminium/PET from coffee packaging bags. The main polymeric layers and the adhesives eventually present in the materials in input were characterized by attenuated total reflection (ATR) (see analysis details and Figures S1 and S2 in ESI). The inorganic content of the materials, including aluminium, was determined by calcination at 550 °C for 5 h. In the case of PE/aluminium/PET used for coffee packaging bags, six different samples available on the market were embedded in KBr, cross-sectioned by dry polishing and then observed under optical microscopy to acquire visible and fluorescent images and study the stratigraphic morphology (see analysis details and Figure S3 in ESI).

3. Results and discussion

3.1. Surfactant preparation and switching procedure

The hazard statements of the tested carboxylic acids, the water solubility of all SAS combinations and the recovery of the carboxylic acid from aqueous solutions after CO₂ addition was initially checked (Tables S1 and S2 in ESI). Caprylic (C₈) acid was poorly recoverable after the addition of CO₂, in apparent disagreement with what was reported in the literature (Chen et al., 2017) and despite its pKa value being close to the ones of the other carboxylic acids here tested. A possible explanation

of the low recovery here achieved (8–13%) in comparison with the expected high recovery given by its low water solubility (680 mg/L at 20 °C) could be attributed to an increase of solubility of the carboxylic acid in the “salted” solution resulting from the switch procedure that contains HCO_3^- base⁺ salt, probably due to the H-bond interactions between RCO_2H and HCO_3^- . On the other hand, long-chain carboxylate salts ($\geq 14\text{C}$) were poorly water-soluble form at the tested concentration (0.5 wt%), neither as triethylammonium nor as NH_4^+ or Na^+ salts. Amongst those tested, capric (C_{10}) and lauric (C_{12}) acids were the only carboxylic acids that behaved like SAS: the switching of lauric acid salts from the ionic form to the neutral one with CO_2 gave the precipitation and the following recovery of the acid close to 90% with all the tested organic and inorganic bases, while a 70% recovery of the acid was achieved from the switching of capric acid. This was due to the higher water-solubility of capric acid in the neutral form than the water-solubility of lauric acid (61.8 vs 4.81 mg/L). Capric and lauric acids were thus selected as the best carboxylate scaffolds for SAS preparation and then tested for the delamination of LDPE/aluminium multilayer waste.

3.2. Delamination of LDPE/aluminium waste with aqueous solutions of SAS

LDPE/aluminium waste subjected to the delamination came from post-consumer aseptic packages used in the food and beverage industry; in the EU, the majority of these cartons are composed of paper, LDPE and aluminium, and follow a well-defined EoL management: they are collected separately in the paper collection and directly sent to a paper mill for being subjected to hydropulping and paper recycling. The remaining waste here used is a sandwich-like structure composed of two external layers of LDPE (26 μm thickness each) and an inner layer of aluminium (14 μm thickness, partially oxidized on the surface) (see Figure S5 in ESI) (Samorì et al., 2023). ATR analyses and UV-Vis microscopy of the samples embedded in KBr and cross-sectioned did not reveal the presence of a distinct layer of adhesive between LDPE and aluminium in all the samples analysed (see Figures S4 in ESI), suggesting that the adhesion of the layers in the material could have been achieved through other approaches (i.e. extrusion coating/lamination or ozonisation to partially oxidizing the polymer surface and improving the adhesion between the oxidized polymer and the oxide layer of the aluminium, Mumladze et al., 2018; Zhang et al., 2015).

The delamination of LDPE/aluminium waste was firstly performed by keeping constant the sample loading (1 wt%) and surfactant concentration (0.5 wt%), and evaluating the performance of each SAS solution in terms of the amount of the recovered LDPE at the end of the dissolution process (expressed on the amount of the multilayer waste in input to the process itself) (Table 1). The results have been compared with the performance of four commercial surfactants, sodium dodecyl sulfate (SDS, an anionic surfactant), cetyltrimethylammonium bromide (CTAB, a cationic surfactant), Tween® 20 and Triton™ X-100 (both non-ionic surfactants). Amongst the capric acid-based SAS, only the C_{10} -TEA solution separated the waste in its polymeric layers and aluminium foil, giving an average amount of recovered LDPE and aluminium of 70 ± 2 and $26 \pm 3\%$ on the input material weight basis, respectively; ATR analysis and calcination of the recovered aluminium testified the absence of LDPE residues on both faces of aluminium, confirming the delamination. The other C_{10} -SASs did not delaminate the material at all (Na or EBA salts) or did it only partially (NH_4 , MEA and DEA salts). In the group of lauric acid-based SASs, the potassium salt did not work, while the primary and secondary ammonium salts partially delaminated the material. C_{12} -TEA gave the best delamination performances; in the case of C_{12} - NH_4 , one additional hour under ultrasounds was necessary to achieve good delamination, probably due to a lower kinetic of swelling with the more polar NH_4^+ ion pair. A correlation between the low polarity of the carboxylate/cation pair and the delamination ability of the SAS can be envisioned from the above-reported trend, where the

C_{12} -TEA pair should be the least hydrophilic (log P value of lauric acid is 4.60, while those of capric and caprylic acid are 4.09 and 3.05, respectively) and the best-performing pair. Additionally, literature results suggest that C_{12} -TEA has a particular behaviour within the class of triethanolamine carboxylates (C_8 – C_{18}), largely used as emulsifying agents particularly in the field of cosmetics, having the lowest surface tension, the highest capacity of producing foam, and an optimal emulsifying power (Fiero, 1938). However, more quantitative data are necessary to fully support this statement.

No one of the commercial surfactants performed delamination under the tested conditions, suggesting that neither the presence of a C_{12} -backbone in the surfactant (like SDS and Tween® 20) nor the presence of an organic ammonium cation (like CTAB) is enough when present alone, but the combination of both these features is essential for achieving the swelling of the layers and, thus, their delamination. These results support the evidence already found by other authors that even low concentrations of long-chain monocarboxylic acids like lauric acid (0.2–1 wt%) contained in milk or vegetable oils are sorbed by LDPE and can cause delamination of metalized laminated packaging materials by reducing the interlayer adhesion between LDPE and aluminium of about 40% (Olafsson and Hildingsson, 1995).

Given the lower recovery of capric acid from C_{10} -TEA solution after CO_2 addition (see Table S2 in ESI), and the lower delaminating performance of C_{12} - NH_4 that needed ultrasounds for completing the delamination, C_{12} -TEA was selected for deeper exploring the conditions of the delamination (Table 2), characterizing the recovered materials (Fig. 1 and Table 3), and evaluating the recyclability of the surfactant solution (Fig. 2).

The surfactant concentration was varied at 0.15, 0.3 and 0.5 wt%, while the lauric acid:TEA molar ratio was varied from 1:1 to 1:3. Partial delamination was achieved at the C_{12} -TEA concentration of 0.15 and 0.3 wt%, also by increasing the amount of TEA or the delamination time (entries 1–8). Surprisingly, at both 0.15 and 0.3 wt%, the addition of excess TEA decrease the delamination performance of SAS (compare trends of entries 1 to 4 and 5 to 8). A tentative rationale for such a trend could be that the additional TEA modifies the overall water environment, making it less polar and reducing the tendency of the ion pair to penetrate the LDPE layers and hold it back in the water phase. Only when the ion pair C_{12} -TEA amount is larger (0.5 wt%), a sufficient quantity dissolve in LDPE to swell the layers and perform delamination, irrespective of the amount of TEA still present in the water phase. At the concentration of 0.5 wt%, all the lauric acid:TEA molar ratios tested

Table 2

LDPE amount (%) on the input material weight basis, w/w) recovered with an aqueous solution of C_{12} -TEA. The data are expressed as the mean of four independent replicates of delamination with each condition \pm standard deviation; +/- indicates the partial delamination of LDPE.

Entry	Surfactant	Conc. (wt%)	TEA eq	Time (h)	Recovered LDPE amount (%)
1	C_{12} -TEA	0.15	1.5	2	28 ± 2 (+/-)
2	C_{12} -TEA	0.15	2	3	29 (+/-)
3	C_{12} -TEA	0.15	2.5	3	19 ± 2 (+/-)
4	C_{12} -TEA	0.15	3	3	10 ± 2 (+/-)
5	C_{12} -TEA	0.3	1.5	3	64 ± 4 (+/-)
6	C_{12} -TEA	0.3	2	3	42 ± 1 (+/-)
7	C_{12} -TEA	0.3	2.5	3	38 ± 2 (+/-)
8	C_{12} -TEA	0.3	3	3	30 ± 5 (+/-)
9	C_{12} -TEA	0.5	1	0.5	72 ± 4
10	C_{12} -TEA	0.5	1.1	1	72 ± 5
11	C_{12} -TEA	0.5	1.5	0.5	74 ± 2
12	C_{12} -TEA	0.5	2	3	70 ± 4
13	C_{12} -TEA	0.5	2.5	3	71 ± 3
14	C_{12} -TEA	0.5	3	1	58 ± 2 (+/-)
15	C_2 -TEA	0.5	1	0.5	–
16	C_8 -TEA	0.5	1	0.5	–
17	TEA	0.5	1	0.5	–

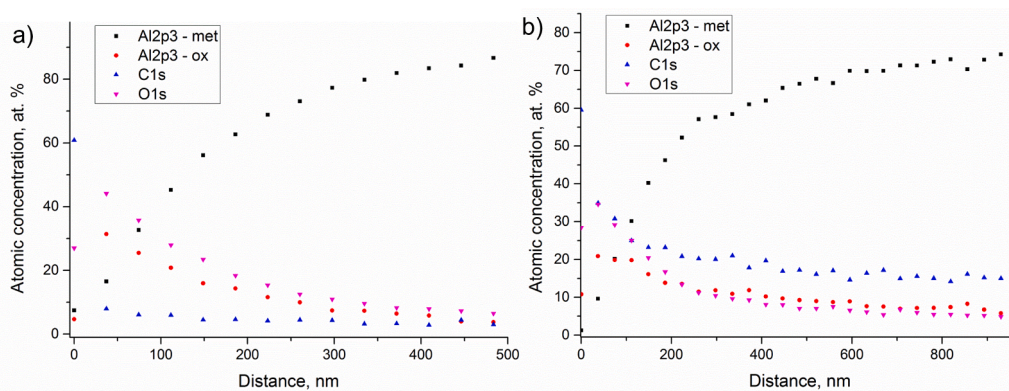


Fig. 1. XPS depth profile of a) aluminium initially present in the waste material in input (the sample was obtained by manually removing one layer of LDPE), and b) aluminium recovered by delamination with C_{12} -TEA (0.5 wt% with 1 eq. of base) after 30 min at 90 °C.

Table 3

Mechanical and thermal properties of LDPE recovered with C_{12} -TEA and LDPE initially present in the waste before the delamination (manually separated from aluminium), determined by tensile stress-strain measurements, TGA and DSC analyses.

LDPE	Elongation at break (%)	Tensile strength at break (MPa)	Young's module (MPa)	T_d 5% (°C)	Residue at 600 °C (%)	T_m (°C)	T_c (°C)	ΔH_m (J/g)	X_c (%)
Before delamination	140±34	7.3 ± 1.6	121±20	332	0.3	103	95	108	37
Recovered by delamination with C_{12} -TEA	132±17	7.0 ± 1.0	128±32	345	0.8	104	96	107	36

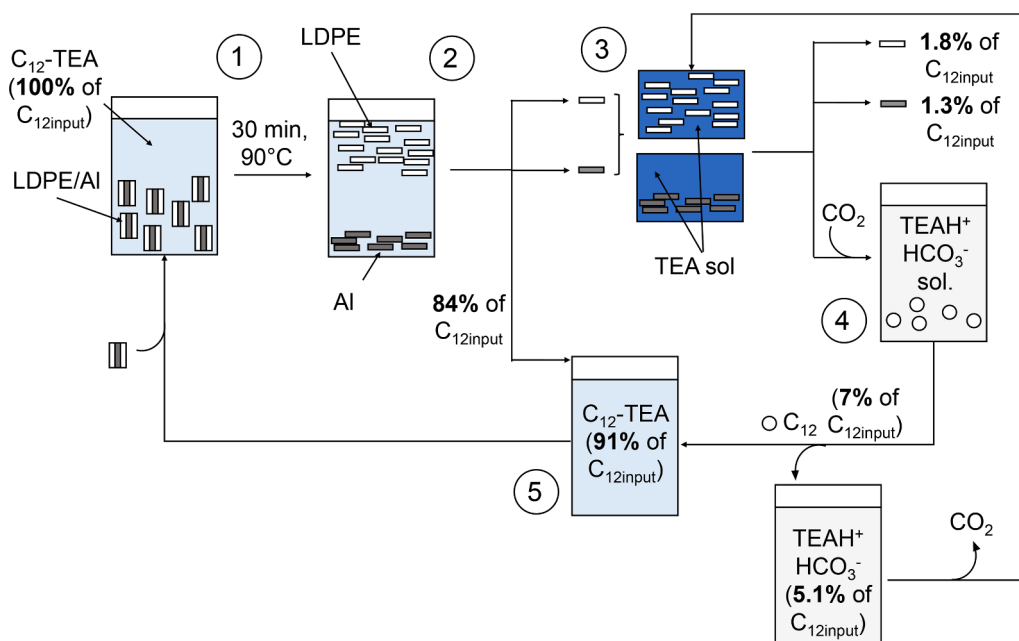


Fig. 2. Delamination process and lauric acid fate.

delaminated the waste in input giving the separation of both LDPE and aluminium (entries 9–13): the optimized conditions were reached by using a lauric acid:TEA molar ratio 1:1 for 0.5 h. A partial delamination was obtained just with 3 eq. of TEA (entry 14). For understanding the role of the base in the delamination process, TEA alone was tested (entry 17), while for understanding the role of the chain length of the carboxylate, triethylammonium octanoate (C_8 -TEA, entry 16) and triethylammonium acetate (C_2 -TEA, entry 15) were prepared and tested. None of these compounds, tested alone, was capable of delaminating the

waste under the best conditions used for C_{12} -TEA (30 min at 90 °C, 1 eq. of the base, entry 9), confirming the essential role of both SAS components for fully separating LDPE and aluminium layers.

3.3. Characterization of the recovered LDPE and aluminium

The purity of aluminium recovered under the optimized conditions, measured after calcination at 550 °C, was 95%; this data was confirmed by XPS analysis that highlighted the presence of organic contaminations

Table 4

Polyolefin (PO) amount (% on the input material weight basis, w/w) recovered with an aqueous solution of C₁₂-TEA from various multilayer waste. The data are expressed as the mean of four independent replicates of delamination with each condition \pm standard deviation, by considering just the transparent polymeric layers recovered. In the case of coffee packaging, each bag was treated as separated; therefore, the data are expressed as the mean of the six individual experiments (+/-: partial delamination of PO; -: no delamination of PO).

Entry	Waste	Waste conc. (wt%)	C ₁₂ -TEA conc. (wt%)	TEA eq	Time (h)	Recovered PO amount (%)
1	PE/aluminium/PET (coffee bags)	1	0.15	1.5	0.5	71 \pm 1
2		10	0.15	1.5	3	70 \pm 2
3		1	0.15	2	0.5	70 \pm 2
4		10	0.15	2	3	69 \pm 1
5		1	0.15	2.5	0.5	70 \pm 1
6		10	0.15	2.5	3	30 \pm 3 (+/-)
7		1	0.15	3	3	69 \pm 1
8		10	0.15	3	3	39 \pm 1 (+/-)
9		1	0.5	1.5	0.5	69 \pm 2
10		10	0.5	1.5	3	71 \pm 1
11		1	0.5	2	0.5	67 \pm 2
12		10	0.5	2	3	70 \pm 2
13		1	0.5	2.5	0.5	72 \pm 1
14		10	0.5	2.5	3	68 \pm 3
15		1	0.5	3	0.5	73 \pm 1
16		10	0.5	3	3	72 \pm 1
17	PO/aluminium/PET	1	0.15	1.5	3	6 \pm 2 (+/-) ^a
18		1	0.15	3	3	9 \pm 5 (+/-) ^a
19		1	0.5	1.5	3	37 \pm 2 (+/-) ^a
20		1	0.5	3	3	39 \pm 5(+/-) ^a
21	PP/aluminium	1	0.5	1.5	3	43 \pm 4 (+/-)
22		1	0.5	2	3	38 \pm 2 (+/-)
23		1	0.5	2.5	3	36 \pm 1 (+/-)
24		1	0.5	3	3	23 \pm 3 (+/-)

^a In some cases, traces of PET were found together with PO.

on the surface of the recovered aluminium (Fig. 1, and Tables S3 and S4 in ESI). After a washing step with an aqueous solution of TEA (20 mM), the residues of lauric acid on the recovered aluminium was 1.5 wt% (corresponding to 1.3% of the initial amount of lauric acid used in the delaminating solution). The surface oxidation was comparable to that of aluminium in the waste material in input (Fig. 1): this is a crucial point since flexible packaging waste like the one also tested here (par. 3.5) has a low aluminium content, which is difficult to recover with other methods (like pyrolysis or incineration) without extensive oxidation, reducing its market value from > 2 €/t (metallic aluminium) to 0.3 €/t (aluminium oxide) (lme.com/en/Metals).

As far as LDPE quality is concerned, after a washing step with an aqueous solution of TEA (20 mM), the residue of lauric acid on the recovered LDPE was 0.7 wt% (corresponding to 1.8% of the initial amount of lauric acid used in the delaminating solution). This value is in line with the amount of lauric acid (0.2 mg per g of polymer) sorbed by LDPE in 2 days of contact at rt reported in the literature (Olafsson and Hildingsson, 1995). The recovered LDPE did not statistically differ in terms of thermal and mechanical properties from the LDPE manually separated from aluminium in the original waste material, as demonstrated by the comparable values of elongation at break, tensile strength at break, Young's modulus, degradation temperature, melting temperature and crystallinity degree (Table 3).

3.4. Lauric acid recovery by using CO₂

A focus on the fate of lauric acid in the entire process with C₁₂-TEA revealed that after the delamination (step 1, Fig. 2) and material separation (step 2), 84% of the amount that entered the process remained in the aqueous solution while the rest remained on the recovered materials (step 3). Therefore, the recovered materials were washed with a TEA solution (20 mM, step 3); this washing solution was then bubbled with CO₂ (the switching step 4) to recover 7% of the initial lauric acid (5.1% remained in the washing solution, also because of the water solubility of the neutral acid i.e. 4.8 mg/L). This amount was added to the aqueous

solution separated from LDPE and aluminium, giving a combined C₁₂-TEA solution containing 91% of the amount of lauric acid that entered the process that can be reused for a new delamination process (step 5). This value is lower than the recycling rate of other switchable compounds like DMCHA used for separating multilayer materials (e.g. > 98%, Mumladze et al., 2018; Yousef et al., 2018; > 96%, Samori et al., 2017) but we believe that it could be increased on a larger scale, where the losses in each step could be minimized, especially improving the washing step of the recovered materials with alkaline solutions followed by the switching step with CO₂. It is worth mentioning that eventual losses of C₁₂-TEA solutions will be less dangerous for human health and the environment in comparison to the majority of the switchable compounds developed so far: no hazard for humans has been identified for TEA, it is not harmful to aquatic organisms and its chronic effects are not expected to be harsh, while lauric acid has a low-risk profile. Moreover, triethanolamine laurate and in general triethylamine carboxylates are widely used as safe emulsifiers, surfactants, skin-conditioning agents, or hair-conditioning agents in the cosmetic industry (Fiume et al., 2013).

3.5. Delamination of different multilayer waste with C₁₂-TEA solution

To verify the versatility of C₁₂-TEA in the delamination of multilayer materials, a set of different flexible waste containing aluminium was also tested (Table 4); three classes of materials were identified by manual sorting of plastic mix waste and then analysed by ATR to confirm their polymeric composition (see Figure S1 in ESI):

- PE/aluminium/PET used for coffee packaging bags (entries 1–16). Six different types of bags (i.e. different brands of coffee) were collected and used (see Figure S3 in ESI). In all samples analysed, aluminium was present as a layer corresponding to 13–20% of the weight of the whole material, while PE and PET were 60–70 and 10–20% of the weight, respectively. Between PE and aluminium, and between aluminium and PET, the distinct presence of tie layers of polyurethane was detected (see Figure S2 in ESI);

- Polyolefins (PO)/aluminium/PET (entries 17–20). This class included a variety of packaging used for food and animal feed applications, in which we observed a large heterogeneity in terms of i) PO composition (PE or PP), ii) number of polymeric layers, iii) tie layer composition, even between different layers of the same material (polyurethane or acrylate), iv) aluminium content (1–20%), v) aluminium mode of deposition (as a layer or applied by vapour coating). In all samples analysed, POs are always layered in the internal part of the packaging.
- PP/aluminium (entries 21–24). This kind of multilayer structure was identified in the packaging of food like snacks, biscuits, chips, and dried fruits; it had a more homogeneous composition since aluminium was always < 4% in the weight of the whole material and applied by vapour coating.

The delamination of PET containing multilayer waste resulted dependant on the specific packaging type. PE from coffee packaging bags was separated as transparent layers under all the tested conditions (C_{12} -TEA concentration of 0.15–0.5 wt%; TEA eq. 1.5–3; waste loading of 1 and 10 wt%) and no PE traces were found on the residual aluminium/PET layers by ATR analysis (data not shown). The lower the waste loading and the faster the delamination was (0.5 h vs 3 h). A partial PE delamination was obtained when the lowest surfactant concentration (0.15 wt%) and the highest TEA eq. (2.5 and 3) were used (entries 6 and 8), a behaviour already observed for LDPE/aluminium waste. The removal of PET from aluminium was not obtained in all conditions tested and this finding could be due to two reasons already described in the literature: i) a different composition and behaviour of polyurethane adhesives present between PE/aluminium and aluminium/PET (see Figure S2 in ESI), and ii) a lower diffusion rate of longer-chain carboxylates through PET than PO (Ügdüler et al., 2021).

On the other hand, PE or PP layers of all the other multilayer waste of the group of PO/aluminium/PET were only partially or not delaminated under the tested conditions (entries 17–20), presumably because of the different components of the adhesives used for these packaging types in comparison to the tie layers composition in coffee packaging bags: polyacrylate- and polyurethane-based adhesives were found in the internal and external layers of samples belonging to the group of PO/aluminium/PET packaging materials, while just polyurethane-based adhesives were found between the layers of coffee packaging bags (see Figure S2 in ESI). In the majority of the samples analysed aluminium, glues and inks remained on the polymeric layers; so the resulting specimens were not considered as “delaminated” (i.e. they were not considered “transparent polymeric layers”). Better delamination results were obtained with a C_{12} -TEA concentration of 0.5 wt%, independently from the molar ratio between TEA and lauric acid.

The same holds for the delamination of multilayer waste containing just PP and aluminium (< 4% on the input material weight basis): a C_{12} -TEA concentration of 0.5 wt% gave a partial recovery of PP present in the waste in input (entries 21–24), while the recovered polymer was < 10% when a lower surfactant concentration (0.15 wt%) was used (data not shown).

4. Conclusions

C_{12} -TEA solutions were here used to delaminate multilayer plastic waste containing aluminium, recovering good yields of non-oxidized aluminium and polyolefins with optimum thermal and mechanical properties from such problematic plastic waste. In particular, C_{12} -TEA resulted highly efficient in the separation of PE from de-pulped food and beverage cartons, a kind of waste already separately collected in the EU and sent to paper mills for recovering paper through hydropulping. C_{12} -TEA worked well also in the delamination of PE/aluminium/PET materials like coffee packaging bags, a waste for which it is feasible adopting a separate collection in several countries (like Italy), considering its large consumption amount. The other flexible multilayer

materials here analysed (i.e. PO/aluminium/PET and PP/aluminium) were more complex and challenging, but promising results were achieved in the recovery of polyolefins. It is worth mentioning that the delamination performance of C_{12} -TEA is a peculiarity of this surfactant: other commercial surfactants as well as other combinations of carboxylic acids and bases here tested were poorly performing in comparison to C_{12} -TEA; moreover, it proved to be the only SAS recoverable from the aqueous solutions simply by adding CO_2 at ambient pressure. Finally, C_{12} -TEA is safer for human health and the environment than the majority of other switchable systems developed so far, and its application as a diluted aqueous solution largely reduces the usage risk. These aspects are particularly important since a sustainable and safe EoL approach orientated to the recovery of individual materials in multilayer plastic waste is currently lacking at the industrial level.

CRedit authorship contribution statement

Martina Vagnoni: Methodology, Data curation, Investigation, Writing – review & editing. **Walter Pitacco:** Methodology, Data curation, Investigation. **Vincenzo Arpaia:** Data curation. **Emilio Catelli:** Data curation. **Chiara Gualandi:** Writing – review & editing. **Roberta Mastroddi:** Investigation. **Alessio Mezzi:** Writing – review & editing. **Chiara Samorì:** Conceptualization, Writing – original draft, Writing – review & editing, Supervision. **Giorgia Sciutto:** Data curation. **Emilio Tagliavini:** Supervision, Funding acquisition. **Paola Galletti:** Supervision, Funding acquisition, Writing – review & editing.

Declaration of Competing 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.

Data availability

No data was used for the research described in the article.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.resconrec.2023.107141](https://doi.org/10.1016/j.resconrec.2023.107141).

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