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Research Article

Electrohydraulic Fragmentation of Aluminum and Polymer Fractions from Waste Pharmaceutical Blisters

Vivek Agarwal, Petteri Halli, Sampsa Helin, Fiseha Tesfaye, and Mari Lundström*



and TGA) showed that the investigated fractions consisted of approximately 10-12 wt % of aluminum, with the clear majority of waste blister mass in the polymer fractions, which consisted of two overlapping layers. Moreover, the polymer layer also gave indications of the presence of Cl. WPBs were subjected to electrohydraulic fragmentation, where the effects of the gap between electrodes (10-40 mm), the amount of pulses (50-500 pulses), pulse frequency (2-5 Hz), and discharge voltage (100-180 kV) on the separation process were systematically investigated. It was shown that at optimal conditions (electrode gap of 40 mm, 300 pulses, frequency of 3 Hz, 130 kV of discharge voltage, and 40 g of initial WPB mass), up to 88% of Al ($\geq 99.4\%$ purity) and polymers were recovered from the investigated waste blister samples. The current study contributes toward improving the circular economy of aluminum as well as the reduction in energy consumption by a new application of electrohydraulic fragmentation for pharmaceutical blister packages.

KEYWORDS: Recycling, Sustainability, Waste management, Circular economy, Energy efficiency

1. INTRODUCTION

Pharmaceutical packaging waste has been continuously increasing over the years with the increase in the number of its applications in everyday life. Globally, the pharmaceutical packaging market grew from USD 47.8 billion in 2010¹ to USD 71.0 billion in 2018.² Moreover, it is projected that the pharmaceutical packaging market will reach USD 111.9 billion by 2026 at a compound annual growth rate (CAGR) of 8.75%.² Pharmaceutical blisters are one of the major segments of pharmaceutical waste packages^{3,4} and consist of an average of 80-85 wt % of plastic films and 10-20 wt % of aluminum foils.⁵ Due to their structural complexity, waste pharmaceutical blisters (WPBs) are mainly disposed of by incineration and landfilling in municipal solid waste.⁶ Incineration of plastics may cause the release of hazardous gases such as dioxins, hydrogen chloride, and nitrous oxide, which ultimately results in environmental problems.^{7,8} Furthermore, incineration also deprives aluminum of recycling in the context of the circular economy of metals. Therefore, the sustainability of WPB recycling along with sustainability in the whole pharmaceutical

investigated to make Al available for direct recycling at a secondary Al facility. The characterization of WPBs (ICP-OES, SEM-EDS,

industry sector has become an environmental, social, and economic concern. $^{9\!-\!11}$

The aluminum recycling process is one of the most common recycling processes, cans being the most recycled consumer product in the world.¹² Transforming aluminum scrap into recycled aluminum requires only 5% of the energy used to produce primary aluminum from bauxite and also produces only 5% of the greenhouse gas emissions compared with the primary process.¹² Recently, only a few research efforts have been devoted to separating metallic and nonmetallic fractions from waste packages to allow aluminum and/or polymer recycling. In thermal processing, which includes incineration and pyrolysis,^{13,14} nonmetallic compounds are evaporated to separate them from aluminum. The main drawbacks of this

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process are high energy consumption, generation of toxic gases, and loss of aluminum along with oxidation.

Separation of aluminum from plastic laminates through exfoliation has also been studied using different solvents such as organic solvents, acid solutions, basic solutions, and water. The hydrometallurgical separation of aluminum from polyvinyl chloride (PVC) has been investigated by dissolving the aluminum from WPBs using hydrochloric acid⁴ and sodium hydroxide¹⁵ as lixiviants. In addition, separation of aluminum from polyethylene in the aseptic composite has been reported using methanoic acid (2-4 mol/L) at temperatures of 60-80 $^{\circ}$ C, a separation time of 25 min, and a liquid to solid ratio of 30 L/kg.¹⁶ Zhang et al.¹⁷ investigated the separation of aluminum-plastic laminates in Tetra Pak using a mixed organic solvent system of benzene-ethyl alcohol-water as separation chemicals. Similarly, Mumladze et al.¹⁸ treated WPBs using acetic acid, acetone, dimethylformamide, and ethyl acetate at 90 °C for 8 min to separate the aluminum layer from the polymer layers.

Another separation approach for waste packages is the application of switchable hydrophilicity solvents, which separate themselves from the extracted product by changing their polarity with the addition and removal of CO_2 . Yousef et al.⁶ and Samori et al.¹⁹ applied *N*,*N*-dimethylcyclohexylamine (DMCHA) to separate polymer and aluminum layers from packaging materials. In another investigation, waste vegetable oil was utilized for the separation of aluminum and polyethylene from Tetra Pak as an environmentally friendly approach.²⁰ Although all of the above-mentioned techniques have some success in separating Al from waste packages, the use of chemicals, solvents, and harsh conditions as well as the generation of contaminated solutions or solvents can significantly reduce the feasibility of the process and also the quality and purity of the aluminum and polymer fractions. This may challenge their recyclability and/or require excess washing water to further increase the burden on the process of water balance and solution purification. This can ultimately adversely affect the sustainability of the process. Therefore, straightforward mechanical processes are addressed to develop effective and environmentally friendly separation processes to recover aluminum and polymers from WPBs.

Electrohydraulic fragmentation (EHF) or high-voltage pulse crushing or shockwave technology is an emerging liberation technique, where shockwaves are generated between electrodes using a high-voltage pulse generator, propagating waves in a liquid medium. The shockwaves ultimately pass through the solid (the target material placed in the liquid medium) spherically and break it down into small fragments from the mechanical weak points or interface of metal and nonmetals.²¹ EHF has been traditionally investigated for the liberation of minerals or rock fragmentation;^{22–26} however, this technique has shown potential for effective separation in the area of waste electronics recycling such as printed circuit boards (PCBs),^{27,28} liquid crystal displays (LCD),²⁹ and solar panels.^{30,31}

The application of EHF in WPB recycling has not been reported in the open literature; therefore, in the present investigation, we introduce a novel approach in the area of aluminum recycling from WPBs. The primary goal of this research is to determine the potential of WPBs as a secondary aluminum source that can be integrated into a state-of-the-art aluminum recycling process. First, characterization was performed to investigate the mineralogy and chemistry of this urban waste and to understand the structural complexity of WPBs. Second, application of the EHF technique for WPBs was investigated and optimized to allow Al recycling from this overlooked metal-containing waste fraction.

2. EXPERIMENTAL SECTION

2.1. Sample Collection and WPB Type. Two types of WPBs were collected from a Finnish firm (Pharmac Finland Oy), see Figure 1. The collected WPBs were manually sorted based on color and



Figure 1. Two different sample types of waste blister packages used as raw materials in the experiments.

divided into two samples: A and B. Sample A was silver in color and 90 mm \times 60 mm in dimensions, while sample B was white with dimensions of 90 mm \times 50 mm.

2.2. Characterization. To determine their chemical composition, the WPBs were cut into small pieces followed by total leaching in a mixture of aqua regia and phosphoric acid in a 4:1 volume ratio. The resulting solution was analyzed using inductively coupled plasmaoptical emission spectroscopy (ICP-OES, PerkinElmer Optima 7100 DV, USA) for metal analysis. Morphological analysis and crosssection measurement of WPBs were conducted using a scanning electron microscope (SEM, Tescan Mira³ GM, Czech Republic) coupled with energy-dispersive spectroscopy (EDS, UltraDry 50 mm² EDS detector and NSS-software, Thermo Scientific, USA). The WPB samples were prepared for SEM analysis using a sharp cutter and embedded in epoxy for 24 h. Next, polishing was performed on the epoxy-containing sample, and the samples were carbon coated in order to avoid charging during analysis. In addition, EDS was employed to analyze the metallic and polymer composition of the WPBs.

For TGA analysis, samples A and B were cut into a particle size fraction of <1 mm and homogenized by mixing. After homogenizing, 20 ± 0.2 mg sample sizes were introduced into to a CHN(O) analyzer (FLASH 2000, Thermo Scientific, UK) using quartz capsules at Åbo Akademi University, Finland. For combustion, technical air (79% N₂ and 21% O₂) was supplied to the reactor chamber. The investigated temperature range varied between 300 and 650 °C, and the total combustion time was 5 min. The formed gas mixture was separated in a chromatographic column and detected by a thermal conductivity detector (TCD). The Eager Xperience software package (version 1.2) was employed to process the electrical signal from the TCD, and the values for C, H, N, and S are reported on a wt % original dry mass basis.

2.3. Electrical Fragmentation Experiments. Industrial WPB samples were fragmented using high-voltage pulse power laboratory fragmentation (selFrag-Lab, Switzerland).⁵⁰ The WPB as-received samples were placed in a 2 L capacity vessel and then filled completely with tap water (\sim 2 L). In each experiment, 20 g of sample was used for fragmentation. After each EHF experiment, the fragmented samples were collected and placed in a muffle furnace (A Scandia oven, Type K4/PDI 40, DK) for drying at 60 °C for 24 h. The dried fragmented WPBs were sieved using a vibratory sieve shaker (Fritsch Analysette, Germany) with a mesh size of 1 mm (Figure 2). The overflow material (>1 mm) was then sorted into three different fractions: (i) separated aluminum fraction, (ii) separated polymer fraction, and (iii) unseparated fraction (Figure 2). Separation of the



Figure 2. Images of example fractions from sample B obtained after EHF experiments: (a) aluminum, (b) polymer, (c) unseparated fraction, and (d) underflow ($\leq 1 \text{ mm}$) material.

aluminum fraction from other fractions was performed based on the density separation method. A solution with a density of 1.78 g/cm³ was prepared by dissolving zinc chloride salt (200 g/100 mL, VWR Chemicals, Belgium, technical purity) into deionized water (Merck Millipore, USA). Overflow material was then added to the prepared zinc chloride solution, and then the solution was centrifuged (Thermo Scientific Heraeus Megafuge 16R Centrifuge, Germany) at 1000 rpm for 5 min. The separated aluminum fraction (at the bottom of the tube) was collected and rinsed with deionized water before drying. The remaining polymer and unseparated fractions (at the top of the tube) were collected and separated manually after drying with the help of an optical microscope. The dry mass of each fraction was recorded. The underflow material (≤ 1 mm) was analyzed for aluminum using ICP-OES with the total leaching method described earlier (section 2.2). The total mass of each fraction was calculated by adding the mass of the separated respective fractions in the underflow and overflow material.

The following equations (eqs 1 and 2) were used to calculate the recovery of aluminum and polymer fractions from WPBs, respectively

recovery of AI (%) =
$$\frac{m_{\rm AI}}{MW_{\rm AI}} \times 100$$
 (1)

recovery of polymer (%) =
$$\frac{m_{\rm PI}}{MW_{\rm PI}} \times 100$$
 (2)

where $m_{\rm AI}$ and $m_{\rm PI}$ are the total masses of aluminum and polymer fraction after separation, respectively, and M is the total mass of unseparated WPBs before separation. $W_{\rm AI}$ and $W_{\rm PI}$ are the total weight fractions of aluminum and polymer in the WPBs.

The parameters of the EHF process investigated were the gap between electrodes (10–40 mm), amount of pulses (50–500 pulses), pulse frequency (2–5 Hz), and discharge voltage (100–180 kV). The experimental series and investigated parameters are presented in Table 1. After finding the optimized parameters, the optimal initial mass of the sample (10–40 g) was investigated. For these experiments, all of the WPBs from the same manufacturer and with a similar structure were chosen in order to have a homogeneous sample type for both samples A and B.

3. RESULTS AND DISCUSSION

3.1. Characterization of Blisters. Chemical analysis of the WPBs was performed using ICP-OES. The results show that the metallic layer of WPBs is mainly made of aluminum in both samples A and B at a purity of over 99.4%. A small

Table 1. Experimental	Parameters in	EHF	Experiments	and
Their Respective Leve	els			

experimental parameters		leve	els	
electrode gap (mm)	10 ^a	20	30	40
amount of pulses	50	100 ^a	300	500
frequency (Hz)	2	3 ^a	4	5
discharge voltage (kV)	100	130 ^a	160	180
^a Value kept constant while	other par	ameters we	re studied.	

amount of Fe was also detected in the range of 0.4-0.6 wt.%. No other element could be detected in significant amounts. The aluminum layer carries 10.9 and 12.1 wt % of the total mass of samples A and B, respectively. SEM-EDS was employed to gather information about selected WPB samples. Figure 3 shows the cross-section area of the investigated WPBs for both sample types A and B. It was observed that in both sample types, two polymer layers were present to cover the drug and one metallic layer or aluminum foil was also present at the bottom to provide support. With the help of SEM micrographs, the thickness of each layer was also measured: the total thickness of the WPBs was in the range of $300-350 \ \mu m_{e}$ that of the aluminum layer in the range of $35-40 \ \mu\text{m}$, and the thickness of the polymer layers varying in the range of 250-300 μ m, being mainly dependent on the number of layers. EDS analysis also confirmed that the metallic layer was mainly formed of aluminum in both samples A and B (Table 2). However, the presence of Fe in small quantities was also detected, which was in agreement with the ICP-OES results. A significant amount of chlorine was detected in the polymer layers, suggesting that the polymer was polyvinyl chloride (PVC) or polyvinylidene chloride (PVDC).

The WPBs were also subjected to TGA analysis to investigate the thermal behavior and the results for mass loss as a function of temperature, as shown in Figure 4, together with determination of the contents of C, N, S, and H in the WPBs. The thermal degradation profile of both samples A and B showed a similar behavior, and significant weight loss was observed in the range of 300–500 °C. At 600 °C, sample A had lost ~90% while sample B had lost ~87% of the total weight, suggesting that most of the polymer fraction had gasified. The remaining mass of the metal-rich residue was 10– 13 wt %, while the ICP-OES results suggested that the aluminum fraction was in the range of 10–12 wt % of the total weight of the WPBs.

With the help of Eager Xperience software (Thermo Fisher, USA), the wt % of C, N, S, and H in the polymer structure was also recorded. The results show that for sample A, the wt % of C, N, S, and H in the polymer layers were 42.1%, 0.07%, 0.09%, and 5.35%, respectively, while for sample B the wt % of C, N, S, and H were 34.7%, 0.07%, 0.04%, and 4.44%, respectively. These results suggest the presence of PVC in the polymer layers, as the theoretical composition in the PVC structure is C \approx 38.4% and H \approx 4.8%, and from the blister with 10% of Al, the same composition became C \approx 34.6% and H \approx 4.3% of the total mass.

Figure 5 presents the concentration of the CO and CO_2 formed during combustion as a function of time at different temperatures. The combustion was conducted in synthetic air for 5 min. As shown in Figure 5, the highest temperature provided the highest content of carbon oxidation. Moreover, at higher temperatures (500–650 °C) for each sample, there are two peaks observed for carbon, most likely corresponding to

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Figure 3. Morphological analysis of a cross-section of the blisters investigated (samples A and B).

Table 2. Composition	(wt %) of Metallic and Nonmetallic La	yers in WPB	Samples A	and B Usi	ng EDS Analysis
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	_					_	-			
element	0	F	Na	Al	Si	Cl	Fe	S	Ti	
sample A										
spectrum 1	10.94	1.67	0.11		0.75	71.5				
spectrum 2	17.63	1.02	1.1	0.11	1.72	78.42				
spectrum 3	8.25	0	0.01	88.68	2.01	0.04	1.01			
sample B										
spectrum 1	2.44	0.26		96.89		0.07				
spectrum 2	5.14	1.99	0.98	0.15	0.34	90.6		0.52		
spectrum 3	9.14	0.79		0.11	0.35	86.79		0.26	2.56	



Figure 4. WPB mass loss during TGA measurement as a function of temperature.

the layered structure of the blisters. These results are in agreement with the report of Yousef et al.,⁶ where the authors also observed two degradation peaks during TGA analysis of PVC-containing blisters. It is noteworthy that all oxidation started approximately at the same time (~ 25 s) regardless of the temperature set. Although the temperature did affect the total amount of carbon oxidation and also the combustion rate—at a higher temperature a higher combustion rate was

observed—the starting point was the same for all samples within a margin of error of ± 2 s.

3.2. Electrohydraulic Fragmentation of WPBs. The WPBs were subjected to EHF, and the results obtained after the EHF of samples A and B can be seen in Figure 6. Figure 6a demonstrates that with an increasing electrode gap from 10 to 40 mm, the mass of separated aluminum (4.4-5.2%), polymer (31.5-31.4%), and unseparated fraction (62.0-55.7%) did not change significantly in sample A. However, the mass fraction of the underflow (≤ 1 mm) was increased from ~2% to ~8% at the higher electrode gap, suggesting an increase in the material fragmentation phenomenon and also in the separation efficiency of aluminum to polymer. Similar results were obtained for sample B, where the mass of separated aluminum (7.2-7.9%), polymer (46.4-59.1%), and underflow (4.5-5.5%) did not change significantly at the elevated electrode gap. However, the mass fraction of the unseparated fraction decreased from 41.8% to 27.4%, suggesting that the separation efficiency of the process was increased at the higher electrode gap. The possible explanation of this behavior could be related to the restriction of particle movement at a lower electrode gap, resulting in lower fragmentation, while more volume for particle movement in the processing vessel was available at a higher electrode gap as suggested by Wielen et al.³² Therefore, under the investigated experimental conditions, a 40 mm

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Figure 5. CO and CO_2 produced versus time during combustion of (a) sample A and (b) sample B. Combustion was conducted in a muffle furnace with a supply of synthetic air composed of 21 vol % of O_2 and 79 vol % of N_2 .



Figure 6. Effect of (a) electrode gap, (b) amount of pulses, (c) frequency, and (d) discharge voltage on percentage mass of aluminum, polymer, unseparated fraction, and underflow (≤ 1 mm particle size) after EHF experiments performed on samples A and B (electrode gap = 10 mm, pulses = 100, frequency = 3 Hz, discharge voltage = 130 kV, and initial WPBs mass = 20 g).

electrode gap would be recommended for improved separation of aluminum from polymers.

In contrast, the effect of the amount of pulses on the mass percentage of each fraction was considerable (Figure 6b). A large amount of pulses resulted in an increasing mass percent of aluminum (3.0-6.7%), polymer (14.1-54.2%), and underflow (4.3-25.7%) fractions, while the mass percent of the unseparated fraction decreased from \sim 78% to \sim 15% in sample A. The effect of the amount of pulses was even more significant in sample B as the mass fraction of the unseparated fraction decreased significantly from ~42.8% to only ~3.5%. This suggests a significant improvement in the separation efficiency of the process. It should also be noted that in both samples A and B the mass fraction of the underflow increased drastically at a higher number of pulses. With the increased amount of pulses, total energy applied also increased, which resulted in an increased number of potential sites for fractures and microfractures. Similar results were also observed by Touzé et al.³³ while studying the application of EHF to concrete waste.

Similarly, Figure 6c and 6d shows that the pulse frequency and discharge voltage have only a slight effect on the mass percentage of the separated overflow fractions compared to the amount of pulses, whereas the fragmentation of blisters into smaller pieces (≤ 1 mm) increases with high pulse frequency and discharge voltage. Not every pulse generated during EHF experiments induces fragmentation in the blisters, and therefore, every misfired pulse results in loss of energy. At elevated pulse frequency (number of pulses per second) and discharge voltage, the energy provided to the system also increases, and therefore, fragmentation of blisters was increased at a higher pulse frequency. The mass fraction of the unseparated fraction was also unaffected by the change in frequency and discharge voltage, and therefore, a pulse frequency of 3 Hz and discharge voltage of 130 kV could be recommended for achieving a higher separation of aluminum from polymers in WPBs. These results are in line with previous findings in the literature where pulse frequency and discharge voltage had a moderate effect on the liberation of rocks after



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Figure 7. Effect of (a) electrode gap, (b) amount of pulses, (c) pulse frequency, and (d) discharge voltage on percentage recovery of aluminum and polymer after EHF experiments performed on samples A and B (electrode gap = 10 mm, pulses = 100, frequency = 3 Hz, discharge voltage = 130 kV, and initial sample mass = 20 g).



Figure 8. Effect of the initial sample mass on percentage (a) mass of aluminum, polymer, unseparated fraction, and underflow and (b) recovery of aluminum and polymer after EHF experiments performed on samples A and B (electrode gap = 40 mm, pulses = 300, frequency = 3 Hz, and discharge voltage = 130 kV).

applying EHF.^{32,33} The results shown in Figure 6 demonstrate that the amount of pulses had the most substantial effect,

reducing the unseparated mass fraction and increasing the separated aluminum and polymer fractions.

On the basis of the results shown in Figure 6, the recovery (%) of separated aluminum and polymer fractions present in both the overflow and the underflow fractions of samples A and B was calculated and is compared in Figure 7. Figure 7a reveals that both aluminum and polymer recovery or separation was better for white color blisters (sample B) than for silver blisters (sample A). By increasing the electrode gap, the aluminum and polymer recovery improved slightly from \sim 42% to \sim 52% and from \sim 37% to \sim 43%, respectively, in sample A, while higher aluminum (from \sim 70% to \sim 72%) and polymer (from ~57% to ~72%) recovery was reported in sample B. However, a noticeable increase in aluminum and polymer recovery was observed when the amount of pulses was increased from 50 to 500 (Figure 7b). Aluminum recovery of up to \sim 72% was achieved at 300 pulses and then increased slightly up to \sim 74% at 500 pulses for sample A. In contrast, for sample B, \sim 84% of the aluminum was recovered at 300 pulses and maximum Al recovery of up to $\sim 87\%$ was achieved at 500 pulses. It can be seen from Figure 7b that the polymer recovery percent increased for both samples A and B at the elevated number of pulses with maximum polymer recovery being recorded at ~86% and ~98% at 500 pulses for samples A and B, respectively. It should be noted that the use of high pulses increases the cost of the process due to higher energy consumption. Therefore, 300 pulses was selected as the optimal condition.

Both pulse frequency (Figure 7C) and discharge voltage (Figure 7D) had only a moderate effect on aluminum and polymer recovery. At a pulse frequency of 3 Hz, aluminum and polymer recovery was ~42% and ~37% from sample A and ~67% and ~56% from sample B, respectively. In contrast, at a voltage of 160 kV, recoveries of ~53% and ~56% aluminum and polymer were achieved for sample A while for sample B, higher voltage resulted in lower aluminum recovery. Interestingly, it was observed that polymer recovery increased continuously with increasing discharge voltage for samples A and B. However, increasing the voltage further will increase the energy consumption of the process.

Overall, based on the EHF experimental results, it was concluded that the most suitable EHF experimental conditions to support the high separation of aluminum from polymer were as follows: electrode gapm 40 mm; amount of pulses, 300; frequency, 3 Hz; discharge voltage, 130 kV. These results also suggest that the obtained optimized EHF conditions could be used for WPBs different in color, chemical composition, and internal structure, such as samples A and B. Typically, aluminum yield or separation was higher in white color blisters (sample B) under similar conditions. Moreover, for all of the overflow samples obtained after the centrifuge treatment, an optical microscope was employed to determine the unseparated fraction from the polymer fraction.

In the optimized conditions (electrode gap = 40 mm, amount of pulses = 300, frequency = 3 Hz, and discharge voltage = 130 kV), EHF experiments were further performed on WPB samples A and B to optimize the initial mass of the sample. The initial sample mass was varied from 10 to 40 g to define the optimal solid to liquid (S/L) ratio for the treatment. The results are given in Figure 8 for both samples A and B. Figure 8a represents the respective mass fractions of aluminum, polymer, unseparated fraction, and underflow material for both samples A and B. In sample A, the mass fraction of the overflow fraction did not change significantly by changing the initial sample mass from 10 to 40 g and remained at the

following level: aluminum, from \sim 5.3 to \sim 5.6; polymer, from ~63.8% to ~65.4%; unseparated fraction, from ~7.5% to \sim 12.7%. The mass fraction of the underflow material decreased slightly from $\sim 23\%$ to $\sim 16\%$, suggesting a decrease in fragmentation phenomena when the higher initial mass was employed. Similar EHF results were obtained from sample B, where no significant difference in mass fraction (%) was observed when varying the initial mass. Figure 8b represents the aluminum and polymer recovery (%) from both samples. Aluminum recovery was higher from sample B than from sample A in all of the experimental conditions. By increasing the initial mass, the recovery of aluminum from the WPBs increased from $\sim 67\%$ to $\sim 85\%$ for sample A and from $\sim 71\%$ to ~88% for sample B. Polymer recovery was higher from both samples when a low initial sample mass (10 g) was used. There was no significant difference in recovery when the initial sample mass was varied from 20 to 40 g. Therefore, an initial sample mass of 40 g (S/L ratio of 20 g/L) is recommended for optimal conditions to lower the energy consumption of the process. Moreover, the water analysis results show that 19.8 mg/L of Al, which corresponds to $\sim 1\%$ of total Al in the investigated blisters, was lost in solution during the EHF experiments under the optimized conditions. It can be hypothesized that Al dissolution may be supported by O₂ evolution during the pulses. In the upscaled EHF process, naturally, closed-loop water usage could result in a slightly higher Al concentration in the process water due to accumulation. However, in more concentrated amounts, dissolved Al can be removed by state-of-the-art methods like hydroxide or phosphate precipitation.^{34,35} Aluminum purity was \geq 99.4% in the separated aluminum fraction, while there was no aluminum found in the separated polymer fraction.

3.3. Process Energy Consumption. Energy consumption in the WPB EHF process is an important factor in order to scale up the process to recycle WPBs. Primary aluminum production is a highly energy-intensive process, and in Europe around ~15 kWh of energy is required to produce 1 kg of aluminum.³⁶ Moreover, as mentioned earlier, aluminum recycling requires only 5% of the total energy consumed in primary production (~0.75 kWh/kg).¹² This suggests that to compete against primary production, the total energy consumption in EHF should not exceed ~14.25 kWh/kg Al. On the basis of the EHF results, energy consumption was calculated at optimized experimental conditions (electrode gap of 40 mm, 300 pulses, frequency of 3 Hz, 130 kV of discharge voltage, and 40 g of initial mass of WPBs) and is reported in Table 3. It can be seen that the optimized EHF treatment energy consumption was from 6.99 to 7.92 kWh/kg depending on the WBP sample. Although the calculated energies are onehalf of the energy consumption compared with the primary aluminum production process, it should also be noted here that on a laboratory scale, a maximum of 40 g (S/L ratio of 20 g/L) sample mass was used in the EHF. At this low initial mass, most of the energy was lost in the water. Therefore, the possible energy consumption was also calculated on an industrial scale assuming 1 kg of WPBs (S/L ratio of 500 g/ L) could be treated using similar experimental conditions and with similar process efficiency for both samples A and B. In such a case, only ~1 kWh of energy would be required to produce 1 kg of aluminum. Consequently, in any of the cases, ~0.75 kWh/kg would be required to refine the separated aluminum in the secondary aluminum recycling process.

Table 3. Energy Consumption Calculations for the EHFProcess of Waste Pharmaceutical Blisters

S/L ratio (g/L)	energy required to treat WPBs (kWh/kg)	efficiency at optimized conditions (%)	energy required for Al separation (kWh/kg)	energy required for Al production ^b (kWh/kg)
sample A	L			
20	0.66	85	7.16	7.92
500 ^a	0.026	85	0.29	1.04
sample B				
20	0.66	88	6.23	6.99
500 ^a	0.026	88	0.25	1.00

^{*a*}Scenario with increased solid content in the experiment, not experimentally verified. ^{*b*}Total energy consumption including separation of Al with EHF and recycling of Al in secondary smelter.

The experimentally verified results suggest that EHF treatment can provide a novel route to improve the circular economy of aluminum—recovering it through recycling with only \sim 50% of the energy consumption compared with primary aluminum production. Besides, the increase in solid/water ratio during treatment may further decrease the specific energy consumption to a notably low level, see Table 3.

4. CONCLUSIONS

- The current study presents for the first time the application of electrohydraulic fragmentation (EHF) on waste pharmaceutical blisters to separate aluminum from the polymer fraction to allow its recycling in a secondary Al plant.
- ICP-OES characterization suggested that the WPB samples investigated contained 10–12 wt % of aluminum and 88–90 wt % of polymer fraction. SEM-EDS results revealed the presence of two polymer layers (possibly PVC) and one aluminum layer in the internal structure of the investigated blisters. TGA analysis of the WPBs showed that 87–90% of mass loss occurred at high temperatures (600 °C) due to the gasification of the polymer fraction.
- The amount of pulses was the EHF parameter having the most significant effect on increasing the separation and recovery of aluminum and polymer fractions as well as reducing the amount of unseparated fraction where aluminum is lost.
- Under optimized experimental conditions (electrode gap of 40 mm, 300 pulses, frequency of 3 Hz, 130 kV of discharge voltage, and 20 g/L of S/L ratio), it was possible to recover 85–88% of aluminum (≥99.4% purity) from the investigated WPB samples. There was 11–14% lost to the mixed fraction with no Al recovery and 1% to the treatment solution, corresponding to 19.8 mg/L. Recovery or separation of Al was always better from white color blisters compared with silver ones.
- Energy calculations of the process demonstrate that EHF could be a potential technique on industrial scale for the recycling of aluminum from WPBs, as approximately one-half of the energy—compared to the primary aluminum production—is required based on laboratory scale results with a solid/water ratio of 20 g/L. With the potentially successful increase of solid content, possibly only ~7% of energy would be required

on an industrial scale compared to the energy required to produce 1 kg of aluminum in the primary process.

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Notes

The authors declare no competing financial interest.

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