applied sciences

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Integrated Process for Ballot Bin Waste Valorization for High-Quality Cellulose Acetate Recovery

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Abstract: Cigarette butt littering poses a significant environmental challenge, with billions of butts discarded each year, fouling ecosystems with slow-to-decompose cellulose acetate filters that absorb and release harmful compounds. In response, an innovative, sustainable approach for valorizing ballot bin waste (BBW) by extracting high-quality cellulose acetate from cigarette butts was investigated. This green approach eliminates the need for hazardous acids and toxic solvents, resulting in a yield of 30% (w/w) and a degree of substitution (DS) of 2.0–2.5, which is comparable to pure cellulose acetate. The following four essential processes are involved in this process: filter separation, water washing to remove impurities, ethanol purification, and acetone precipitation of the cellulose acetate. This approach not only mitigates environmental harm, but also supports circular economy goals by transforming waste into valuable resources.

Keywords: ballot bin; cigarette butt; cellulose acetate; waste valorization; Italy; Milan; circular economy

Citation: Lamanna, N.; Zuccante, G.; Rossetti, M.; Franzetti, A.; Santoro, C.; Zoia, L. Integrated Process for Ballot Bin Waste Valorization for High-Quality Cellulose Acetate Recovery. *Appl. Sci.* **2024**, *14*, 10955. https://doi.org/10.3390/app142310955

Academic Editors: Marco Carnevale Miino, Amanda Laca Pérez, Yolanda Patiño and Daniel Sol

Received: 7 October 2024
Revised: 15 November 2024
Accepted: 22 November 2024
Published: 26 November 2024

1. Introduction

In the last few decades, global tobacco consumption has skyrocketed, reaching a peak of 6.25 trillion cigarettes in 2012 [1]. Despite the decrease in cigarette consumption to 5.7 trillion reported by Tobacco Atlas [2], this number is expected to rise to ca. 9 trillion in 2025 due to the future uptrend of the market along with population growth [3,4]. The exponential increase in cigarette manufacturing has led to the ubiquitous accumulation of toxic waste cigarette butts (WCBs) in the environment [5]. If properly disposed of, WCBs are discarded in ashtrays, receptacles, and bins [6]. According to the current definitions, WCBs are litters composed of unsmoked tobacco residue, charred tobacco on the end, a used filter (cellulose acetate, CA), and a wrapping paper [7], and one of most important components from an environmental point of view is the concentration of heavy metals, as suggested by Quéméneur et al. [8]. Although a single WCB weighs 0.4 g on average, ca. 2 million tons of WCBs are being discarded yearly in the environment. Over time, cigarette butts have become the number one most littered item on earth [9]. Therefore, they are considered to be one of the biggest environmental concerns due to their toxic effects on living organisms [10]. Due to its slow degradability, the cellulose acetate filter can take up to 30 years before completely degrading. Its high adsorption capability leads to the incorporation of a high number of hazardous substances from the external environment over time that are added to those retained during smoking [11]. All these toxic chemicals can be leached in relatively

short periods in the environment and enter the food chain [12–15]. According to recent studies, WCBs can release up to 7000 chemicals, including highly ecotoxic substances such as lead, cadmium, polycyclic aromatic hydrocarbons (PAHs), aromatic nitrosamines, and cyanides [16,17]. In summary, the combination of high consumption rates, careless disposal practices, environmental persistence, and toxicity makes cigarette butts the most littered item globally [11], highlighting the urgent need for effective waste management and recycling strategies, such as those proposed in this study.

Therefore, a large-scale system to collect cigarette butts is still missing, and currently, collection campaigns are mainly carried out by small clean-up crews of volunteers [9]. Strong technological efforts have been made to upscale these collection campaigns with the introduction of Artificial Intelligence (AI). However, these studies are still in the embryonic phase, and they cannot immediately solve the collection problem of WCBs [18]. Moreover, the traditional disposal pathways mainly involve landfilling, which is still a direct violation of environmental safety standards due to its potential for soil toxicity [5] Avoiding the occurrence of cigarette butts in landfills would alleviate the dispersion of numerous pollutants in the environment through WCB leachate [5,19]. For instance, in the aquatic environment, the LC50 of smoked cigarettes corresponded to 1 cigarette per L specifically for *Atherinops affinis* and *Pimephales promelas* [20]. In this regard, incineration is another method that, at first glance, can be considered as a helpful way to generate electricity from unwanted waste, but this practice uncontrollably releases hazardous side products and greenhouse gases into the atmosphere [21]. Moreover, the uncontrollable and impulsive habit of littering, i.e., the abandonment of small objects in public places, has led to the widespread dispersion of WCBs in the environment, which poses serious challenges in collection campaigns [22]. In Italy alone, 72 billion cigarette butts are littered in the environment every year, and environmental policies struggle to contrast the littering habits of citizens [23].

Hence, all solutions that support a smart and sustainable way to collect and recycle cigarette butts must be implemented. The complex compositions and low-value properties of WCBs present a challenge to the recycling process. Several possibilities for recycling have been identified for various technological applications. Some of them include the creation of clay bricks through WCB incorporation [24,25], the addition of WCBs to bituminous asphalt as a high-performing fiber modifier [26], the transformation of the butt into cellulose pulp to feed the paper supply chain [27,28], the fabrication of porous and sound-absorbing material [29], and the production of superhydrophobic filters to clean up sites when oil spills occur [30,31]. Recent research progress has shown that WCBs could be upcycled into some value-added materials with different amounts of success. Applications of treated WCBs for hydrogen storage material, hydrochars, and high-performance supercapacitors have been demonstrated due to their high porosity, ultrahigh surface area, pore volume, and oxygen-rich nature [32,33]. Nanocrystalline cellulose and cellulose pulp were also isolated from cigarette filters based on their potential high-purity cellulose content [34]. The recycling of cellulose filter into a semi-finished cellulose acetate product with a specific granule formulation is of great interest from an industrial point of view, since this can be further processed for several applications [35], such as membranes [36–40], drug delivery systems [41], thin films [42], chemicals [43], optical sensors [44], safety glasses and shields [45], optical lenses [46,47], LCD polarizing panels, costume jewelry, combs, buttons, necklaces [48], and food packaging [49].

In this context, the concept of a circular economy emerges as a transformative approach, where waste is redefined as a resource that can be recovered and reused. Sustainable waste management practices not only minimize environmental impacts, but also foster economic growth by creating new opportunities for resource recovery and recycling [5]. As highlighted by Cruz [50], the integration of sustainability principles into waste management is essential for addressing the challenges of modern waste systems. Considering this, the present work aimed at developing a green chemical approach [51], and the main objectives of the green protocol include the usage of non-toxic solvents with a focus on

sustainable waste management from an environmental and human point of view in order to recover cellulose acetate as a semi-finished, odorless product in the form of granules from cigarette butts.

2. Materials and Methods

2.1. Materials

2.1.1. Collection and Storage of Ballot Bin Waste (BBW)

Our university identified and set up, on an experimental basis, special areas for smokers, marked by specific signs and equipped with specific containers known as ballot bins. Eight active smoking areas were equipped with ballot bins for the separate collection of butts and smoking products, serving six buildings on the Milano-Bicocca campus (Figure 1). Smoking areas within the campus were placed at no less than 7 m and no more than 25 m from the entrances of the buildings, delimited, and signaled through special vertical and horizontal signs. Within the identified areas, special containers for the selective collection of cigarette butts were installed with a tiny ballot bin slot of around 1 cm.

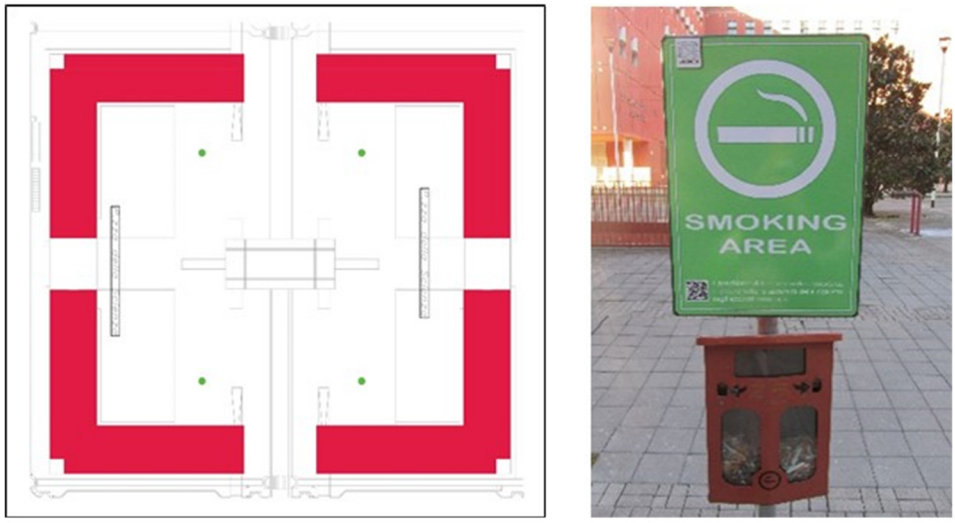

Figure 1. On the **left**: map of Piazza della Scienza, part of the Milano-Bicocca University campus; the green dot indicated the four "smoking area" (of the total eight area) where the ballot bins were installed (on the **right**).

The selective collection of cigarettes butts was achieved by the above-mentioned special containers installed in the smoking areas within the University of Milano-Bicocca Campus. Periodic emptying (roughly one collection per week) allowed for recovering different samples over a period of 6 months (April–September). The samples were air-dried for 3 days under a fume hood and stored in plastic bags at −20 °C.

2.1.2. Reagents

All the chemicals and reagents used for the heavy metal analysis were of analytical grade. Nitric acid (65%, Sigma-Aldrich, Darmstadt, Germany), and hydrochloric acid (35–38%, Sigma-Aldrich) were used to digest the organic matter in the samples. The mono-element certified standard reference materials of Cd, Co, Cu, Ni, Fe, Mn, Zn, Cr, Pb, and As at 1000 mg/L (PerkinElmer Pure, Waltham, MA, USA) were used for calibration and quality control.

2.2. Methods

2.2.1. Sieving

The waste refinery started with a sieving treatment. A stainless-steel sieve with a 5.6 mm opening was used to filter various portions of the ballot bin waste (from 20 g to

100 g), thus separating the cigarette butts from the ashes, unsmoked tobacco, and other residues. To sort the waste by size, the sieve was manually shaken for 5 min. The sieve caught cigarette butts; ashes and tobacco residues were recovered separately. The different fractions were weighed and stored in plastic bags at 20 °C.

2.2.2. Water Washing

The cigarette butts were put in bidistilled water (5% *w*/*v*) and vigorously mechanically stirred at 50 °C for 1 h. Then, the suspension was sieved by a stainless-steel sieve with a 5.6 mm opening in order to separate the smoked filter (retained by the sieve) from the tipping paper in form of a pulp. The smoked filters were washed with clean water, air-dried in oven at 60 °C for 1 day, and weighed. The pulp paper was recovered by Büchner filtration, washed with water, air-dried overnight under a fume hood, and weighed. The same procedure was applied to unsmoked filters, manually removed from cigarettes, as a reference.

2.2.3. Ethanol Washing

The smoked filters were put in absolute ethanol (10% *w*/*v*) and vigorously mechanically stirred at room temperature for 1 h. Then, the suspension was filtered by Büchner. The clean filters were washed with fresh ethanol (100 mL), air-dried overnight under a fume hood, and weighed. The ethanol filtrate was rotavaporized in order to recover the solvent. A yellow-brown, viscous, oily material was recovered in a round-bottom flask and weighed. The same procedure was applied to unsmoked filters as a reference.

2.2.4. Cellulose Acetate Precipitation

Cellulose acetate solution was prepared by dissolving 20 wt% of a cigarette filter in acetone (ACS reagent, ≥99.5%, Sigma-Aldrich) at a concentration of 100 g in 1 L, at room temperature under continuous stirring (300 rpm), over a period of 24 h. The solution was then centrifuged at 3000 rpm for 15 min in order to remove suspended and non-acetone-soluble black particles. The supernatant was then dropwise added to 2 L of bidistilled water with continuous stirring to re-precipitate the cellulose acetate, which was recovered by filtration, dried in oven at 60 °C for 1 day, and weighed. The acetone in the filtrate could be recovered by distillation from water and continually recycled in the process.

2.2.5. Characterizations

Attenuated total reflectance (ATR) FT-IR measurements were performed using a NICOLET iS5 spectrometer (Thermo Scientific, Waltham, MA, USA) equipped with an iD7 ATR accessory and diamond crystal. All infrared spectra were recorded within the range of 4000–600 cm^{-1} with a 4 cm^{-1} resolution and 32 scans. The estimation of the degree of substitution (DS) was performed following the procedure reported by Fei et al., 2017, specifically developed for highly acetylated samples [52].

For all the fractions, Cd, Co, Cu, Ni, Fe, Mn, Zn, Cr, Pb, and As were quantified using an inductively coupled plasma-optical emission mass spectrophotometer (ICP-OES Optima 7000 DV PerkinElmer, Waltham, MA, USA). All samples were prepared according to the following procedure: about 750 mg of oven-dried representative material was precisely weighted, placed in a PTFE vessel, and digested in a Milestone Ethos TC Microwave digestion system by adding 4 mL of aqua regia (HNO_3 65% and HCl 37%) in a closed system to reduce the risk of contamination [53]. The system was programmed to use up to 1000 W of power to increase the detected temperature to 220 °C, at which time, the temperature was maintained for 15 min. After digestion, samples were made up to a total of 10 mL and analyzed by ICP-OES. The instrument's detection limit was 0.1 μg/g. For every sample, three replicates were taken, and the average value and standard deviation were calculated. The data are expressed as the average of 2 purification experiments, analyzed 3 times each.

For GPC characterization, the pulverized material (roughly 50 mg) was put in an 8 mL dried sample bottle equipped with a magnetic stirrer with 1 mL of pyridine and 250 µL of benzoyl chloride. The solution was vortexed until it became homogeneous and allowed to cool to room temperature. The sample was kept under magnetic stirring at room temperature for 2 h. To precipitate the benzoylated material, a deionized water–ethanol solution (1:3 v/v, 20 mL) was added and the mixture was vigorously shaken and vortexed for 5 min. The solid was filtered off through a sintered funnel (grade 3), washed with further ethanol, and purified with methanol. The benzoylated samples were solubilized in THF and passed through a 0.45 µm GHP Acrodisc syringe filter (Waters, Milford, CT, USA) for GPC analyses. Gel Permeation Chromatography analyses were performed on an Agilent HP1100 series (Agilent, Santa Clara, CA, USA) equipped with a UV-Vis detector set at 240 nm and RID (Refract Index Detection). The injection port was a Rheodyne loop valve equipped with a 20 µL loop. The GP-column system was composed of a sequence of an Agilent PL gel of 5 µm, 500 Å, an Agilent PL gel of 5 µm, 5000 Å, and an Agilent PL gel of 5 µm, 10^4 Å. The solvent used was THF (Fluka 99.8%, Sigma-Aldrich). The PL Polymer Standards of Polystyrene from Polymer Laboratories (Church Stretton, UK) were used for calibration. An evaluation of the number-average molecular weight (M_n) and the weight-average molecular weight (M_w) of the samples was performed. The peak molecular weight M_p is defined as the molecular weight of the species with maximum absorbance. Moreover, the ratio $D = M_w/M_n$, defined as the dispersity index, was also calculated. The M_n, M_w, and M_p values reported are the averages of three analyses ($p = 0.05$, n = 3).

For ^{1}H-NMR characterization, 10 mg of cellulose acetate was dissolved in 750 µL of d_6-Acetone (Sigma-Aldrich, 99%) and placed in a 5 mm NMR tube. ^{1}H-NMR spectra were collected on a Bruker-500 spectrometer (Bruker, Billerica, MA, USA) operating at 500.13 MHz with the following acquisition parameters: 90° pulse width of 10 ms, spectral width of 12 ppm, and relaxation delay of 2 s. The total number of scans was 64 (four dummy scans) and the acquisition time was 2.60 s. All ^{1}H-NMR chemical shifts reported in this work are relative to the residual peak of the solvent. The estimation of the degree of substitution (DS) was performed following the procedure described in the literature by Zhang [54].

3. Results and Discussion

Since the problem of littering related to cigarette butts affects any type of environment, both urbanized and non-urbanized, the University of Milan-Bicocca has, thus, decided to implement a smoking-free policy by approving the regulation for the application of the smoking ban, which came into force in January 2020. The design and implementation of models for the collection of cigarette butts have been conducted in private areas for public use owned by the University of Milano-Bicocca. As reported by Yousefi et al. [55], the most important attempt at proper cigarette butt collection is the smoker's behavior modification in order to reduce littering. The installations were in line with the smoke-free policy adopted by the University of Milano-Bicocca. The ban on smoking was implemented in all internal areas and in the external amenities of university buildings, except for the external areas reserved for smokers (smoking areas), which were located at a reasonable distance from the entrances and suitably equipped. The first stage of the experimentation was based on the collection of cigarette butts using a receptive system such as ballot bins, with the double aim of recovering the highest number of butts and ensuring the health of those attending Milano-Bicocca University. In fact, the goals of this policy are the following: first, the protection and safety of all, thus reducing exposure to second-hand smoke; and second, the growth of a culture of health within the institution. Moreover, following the principle of circular economy, combining the possibility of recovering cigarette butts with an anti-littering policy associated with recycling could represent a win-win situation for the university, for all those who attend it, and for the environment that surrounds the university itself.

3.1. Collection and Purification Overview

Only a few studies on cellulose acetate valorization have reported the direct use of waste cigarette butts collected in ballot bins as a raw material. In this work, we set up an integrated process, starting with the materials gathered from the smoking areas. Figure 2 reports the purification procedure for the treatment of the ballot bin waste to obtain cellulose acetate. The different steps are highlighted in oval blue, while the different materials along the mass balance are highlighted in rectangular white. The mass balance data reported were the average of five experiments with a 95% confidence interval. Representative pictures are also depicted in the scheme.

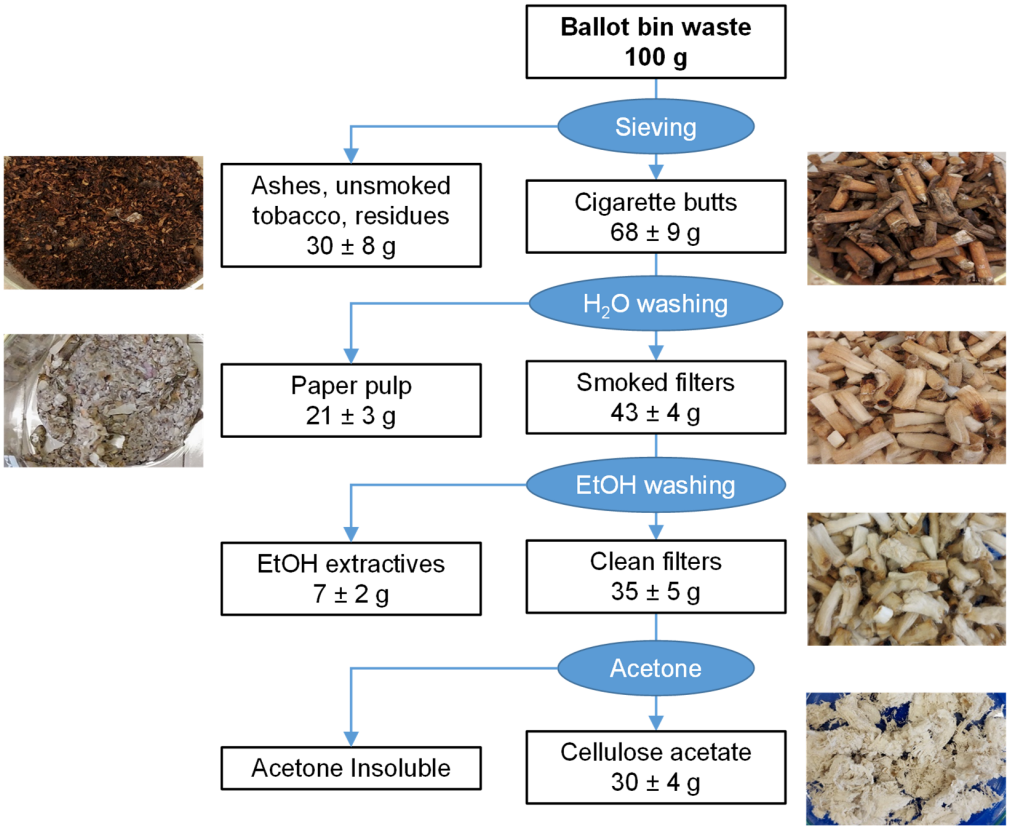

Figure 2. Representative scheme of the purification process for the treatment of the ballot bin waste to cellulose acetate. The different steps are highlighted in oval blue, while the different materials along the mass balance are highlighted in rectangular white.

3.2. Sieving

In order to start the waste refinery, a sieving treatment was performed. A sieve with a 5.6 mm opening was able to separate the cigarette butts from the ashes, unsmoked tobacco, and other residues. This simple separation was made possible because the tiny ballot bin slot (around 1 cm) allowed for a high selectivity of the collected waste. The sieving was able to fractionate 100 g of ballot bin waste into the following two fractions: 68 ± 9 g of cigarette butts and 30 ± 8 g of other residues (Figure 2). The total recovery yield was higher than 98%. Among the many harmful and toxic compounds found in tobacco smoke and cigarette butts, metals seem to play an important role, in terms of direct toxicity either for smokers or in terms of environmental impact [56,57].

As reported in Table 1, row 1, the ballot bin waste had a significant content of heavy metals, mainly Fe and Mn. As, Cd, and Co were never detected (detection limit 0.1 μg/g) [56]. The contamination of the ballot bin waste by the steel of the structure could not be excluded, but this was not further investigated. With a simple sieving step, we were able to remove the ashes, unsmoked tobacco, and other residues from the cigarette

butts. The heavy metal content after sieving was measured on the two collected fractions, namely the cigarette butts and ashes. Considering the mass balance in Figure 2 and the heavy metal content in Table 1, rows 2 and 3, the sieving was able to remove, respectively, 36% of Cu, 15% of Cr, 15% of Fe, 41% of Mn, 17% of Ni, 0% of Pb, and 44% of Zn. For the comparison of the absolute heavy metal concentrations in the ashes, good agreement was found with the work of Dahlawi et al. [53]. It is interesting to note that the lowest removal efficiency was related to heavy metals with a high smoke transfer rate (Pb in particular, but also Ni and Cr) [58]. Those heavy metals had a tendency to transfer in the smoke phase during the burning phase, and low concentrations in ashes were, therefore, detected. Mn, on the other hand, was characterized by a low transfer rate and remained in the ashes (removal efficiency of 41% by sieving). These data indicate the importance of setting up an integrated management strategy for cigarette butt collection based on the use of ballot bins to minimize water contact with the waste, either in terms of the minimization of the leaching of heavy metals in the environment or in terms of recovery and upgrading. Water contact could mobilize heavy metals between fractions, but also promote the cellulose acetate degradation of cigarette filters during the aging of the material [59].

Table 1. Heavy metal content expressed in μg/g (* or mg/L) in the different fractions recovered during the purification process. Data are expressed as an average ± interval of confidence (n = 3, $p = 0.05$).

	As	Cd	Co	Cu	Cr	Fe	Mn	Ni	Pb	Zn
Ballot bin waste	nd	nd	nd	5.0 ± 3.4	0.4 ± 0.3	351.0 ± 328.5	39.0 ± 26.6	2.7 ± 1.5	0.6 ± 0.4	7.9 ± 5.6
Cigarette butts	nd	nd	nd	5.0 ± 3.4	0.3 ± 0.2	383.6 ± 284.4	2.4 ± 1.4	2.3 ± 1.5	1.3 ± 1.4	10.9 ± 2.2
Ashes	nd	nd	nd	6.0 ± 1.0	0.2 ± 0.1	170.5 ± 14.7	53.6 ± 9.7	1.5 ± 0.6	nd	11.5 ± 0.7
Smoked filters	nd	nd	nd	0.6 ± 0.5	0.2 ± 0.1	49.4 ± 39.0	nd	1.7 ± 1.4	0.8 ± 0.6	5.6 ± 3.9
Washing waters *	nd	nd	nd	2.5 ± 0.4	nd	49.25 ± 27.5	8.6 ± 5.6	1.56 ± 0.9	1.6 ± 1.1	nd
Clean filters	nd	nd	nd	0.7 ± 0.5	0.3 ± 0.2	39.6 ± 9.9	nd	2.0 ± 1.4	1.8 ± 0.2	5.9 ± 2.2
Cellulose acetate	nd	nd	nd	1.3 ± 0.4	0.3 ± 0.2	9.1 ± 0.9	nd	2.0 ± 0.3	0.3 ± 0.2	2.0 ± 0.3

3.3. Water Washing

Water washing has been recognized as an effective method for the cleaning of cigarette butts [35,47] without the addition of any other chemicals, thus minimizing treatment costs and mainly preserving the chemical properties of the cellulose acetate. Alkaline and acidic conditions have been recognized to help in the purification of cigarette butts [56], efficiently removing heavy metal contaminants and/or organic compounds. Nevertheless, these conditions are likely to promote deacetylation and/or depolymerization reactions on the cellulose acetate polymer [60]. Since the goal of this research was to recover pure cellulose acetate with pristine chemical characteristics, we avoided the use of such conditions. The water washing was also able to remove, besides heavy metals and other inorganics, water-soluble organic compounds (such as glues, plasticizers, and smoke residues) and defibrillate the tipping paper under heating and vigorous stirring.

It was possible to recover a fraction called smoked filters with an average weight of 43 ± 4 g (starting from 100 g of BBW), along with 21 ± 3 g of pulp paper (Figure 2). The total recovery yield was around 90%; the lost parts were likely the water-soluble components reported above. Simple water washing, as reported in Table 1, row 4, was able to remove 91% of Cu, 59% of Cr, 92% of Fe, 100% of Mn, 55% of Ni, 62% of Pb, and 68% of Zn. These findings are consistent with other research reported the literature that was conducted using the same conditions [56,61]. In fact, as reported by Merman [56], Cu, Fe, and Mn (removal efficiency higher than 90% in this work) have the highest concentrations of metals leached from smoked cigarette material measured after a specified period of soaking. The effect of water washing in the purification of cellulose acetate was also explored by Benavente et al. [35]. Factors like pH and ionic strength could affect the amount of metal released [57] and a clear effect of conditions (NaCl vs. sodium acetate vs. sulfuric acid) was observed in terms of metal extraction, with the best results with H_2SO_4 0.02%. According to the authors, the acidic solution seemed to be the most efficient for the extraction of Al, Fe,

Cu, and Zn. Our results indicated that, even in neutral conditions (without the addition of any chemicals), a high removal efficiency was obtained; this was explained by the higher temperature of the water, as used by De Fenzo et al. [47]. Even though the scope of the present research is not to detect the amount of metals leached by WCBs in aquatic environments, in this article, we measured the heavy metal concentrations in the washing water and the data are reported in row 5 in Table 1, which are consistent with literature reports [56].

3.4. Ethanol Washing

Ethanol washing has also been recognized as an effective and environmentally friendly method for the cleaning of cigarette butts, mainly from organic contamination. As a green and bio-based solvent, ethanol is able to remove organic compounds such as plasticizers and smoke residues (e.g., nicotine and PAHs) [35,39,46,62]. This is why, once the filters were characterized for their heavy metal concentrations, they were subjected to another washing step with ethanol. The final product is called a clean filter, and it was recovered from 43 g of smoked filters in 35 ± 5 g of white/yellowing cellulose acetate fiber filters, while after solvent evaporation, 7 ± 2 g of oily, yellow organic material was removed (Figure 2). In this way, ethanol was completely recycled during the purification process. Preliminary FT-IR investigations (Figure 3) indicated that this material seems to be mainly composed of alkyl esters, ethanol soluble cellulose acetate, or most probably, plasticizers and other chemicals used in tow [47]. Other organic compounds, such as nicotine and the residue of combustion, could also be present, but the material was not further investigated. Based on the solubility properties of ethanol, it is not surprising that heavy metal concentrations were minimally affected (Table 1, row 6). In a previous study, it was found that the extraction of heavy metals is more favored in a slightly acidic ethanol solution (0.02% w/v H_2SO_4) than in a neutral one [35]. However, the use of ethanoic acidic solution can modify cellulose acetate (trans-esterification) and, thus, affect the quality as well as the yield of the final product.

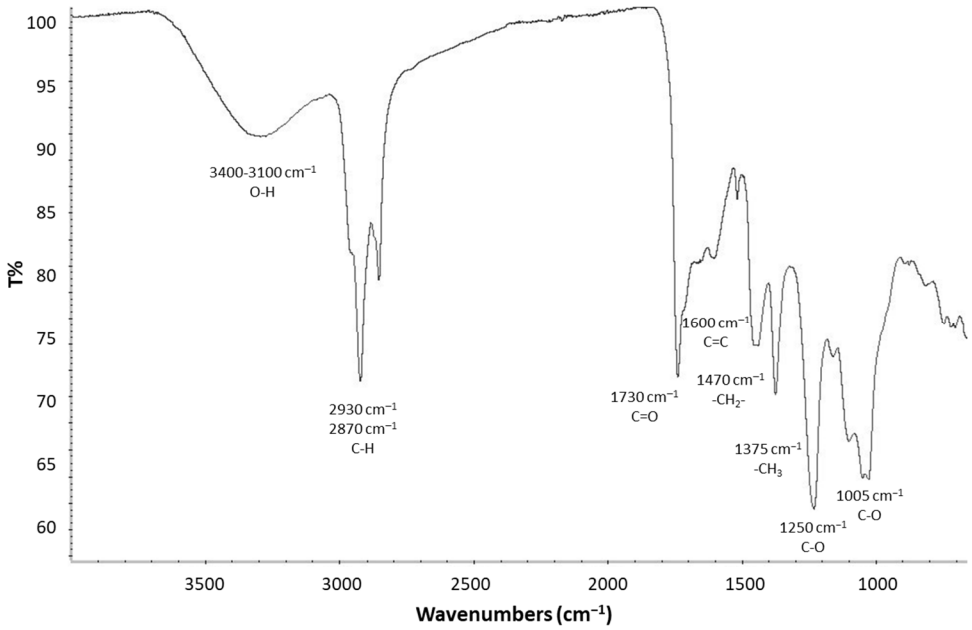

Figure 3. FT-IR spectrum of ethanol extractives form BBW.

3.5. Acetone Precipitation of Cellulose Acetate and Characterizations

The final stage in cellulose acetate purification was based on the dissolution/precipitation strategy. It should be mentioned that cellulose acetate (CA) is soluble in different organic solvents depending on the number of hydroxyl moieties substituted by acetyl groups, i.e., the degree of substitution (DS). Generally, when the DS is larger than two (like in

the case of WCBs), common solvents such as tetrahydrofuran, acetone, or dioxane can be used [63]. On the contrary, when DS < 2, CA can be dissolved in fewer solvents, for example, acetic acid [43]. In the present study, CA was dissolved in acetone, since it is a greener, cheaper, and more commercially available solvent compared to the others listed above. The insoluble material (removed by centrifugation) was a dark brown heterogeneous material composed mostly of burned filter parts and other residues, and it was not further investigated. The cellulose acetate was then precipitated by putting the clear solution in water as an antisolvent. Finally, 30 ± 4 g of cellulose acetate was recovered by filtration and drying as a white, fluffy material from 35 g of clean filter (Figure 2). The final product, CA, was characterized by the means of different analytical tools. The precipitation from acetone strongly reduced the metal concentrations (Table 1, row 7), probably due to a double mechanism, as follows: either during the water precipitation or by removing insoluble parts where heavy metals were chelated. The levels of heavy metals in the final cellulose acetate were generally quite low. The cellulose acetate recovered (from five experiments) was also deeply chemically characterized by ^{1}H-NMR, GPC, FT-IR, and UV and compared to the cellulose acetate present in an unsmoked filter (Figure 4).

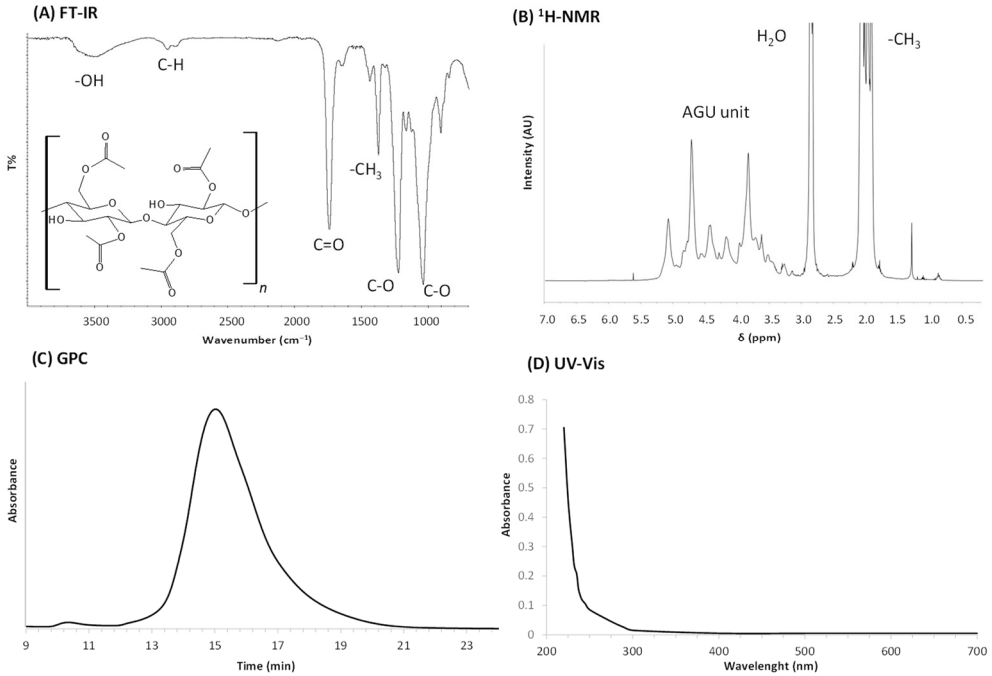

Figure 4. Typical analyses of the final CA recovered. Panel (**A**): FT-IR; panel (**B**): ^{1}H-NMR; panel (**C**): Gel Permeation Chromatography (GPC); and panel (**D**): UV-Vis. The structure of diacetylcellulose in used as a reference.

The FT-IR spectrum (Figure 4, panel A) confirms that the recovered material was cellulose acetate. This was evidenced by the appearance of peaks related to acetate groups, i.e., those at 1740 cm^{-1} and 1220 cm^{-1}, referring to the C=O stretching and C-O stretching in acetyl esters, and those at 1369 and 1420 cm^{-1}, peaks arising from the symmetric bending vibration of C-H in the methyl groups, respectively [64,65]. The signals located at ca. 2950 and 1030 cm^{-1}, resulting from the C-H stretching vibrations of sp^3 hybridized C atoms and the C-O stretching of alcoholic and etheric bonds, respectively, are related to the structure of the cellulosic backbone [61]. Finally, the presence of the broad band at 3000–3600 cm^{-1} refers to O-H stretching and, therefore, enlightens us that there is a partial substitution of -OH with acetyl acetate groups. The ^{1}H-NMR spectrum (Figure 4, panel B) further confirms the structure of the cellulose acetate through the appearance of strong signals within the chemical shift range of 1.8–2.2 ppm assigned to methyl hydrogens in acetate groups. Moreover, the presence of many superimposed peaks in the chemical

shift range of 3.5–5.5 ppm, which refers to the protons belonging to the anhydride glucose units (AGUs) of the cellulose chain, was detected [52]. The peak at ca. 2.8 ppm is due to water traces, as reported by Xu et al. [63]. The data regarding the degree of substitution determined by the FT-IR analysis, the molecular weights, and the dispersity index (D) achieved by the GPC analysis are reported in Table 2. The data from the unsmoked filter (used as a reference), in terms of the DS, molecular weights, and dispersity index, confirmed that the purification procedure retained the chemical properties of pristine cellulose acetate. The DS and GPC outputs were measured after each step of the purification process, highlighting no significant modifications. The data from the five experiments on ballot bin waste purification confirmed that the collection procedure (ballot bin configuration, time of collection, and storage) retained the chemical properties of cellulose acetate. In fact, the DS and molecular weight values were always comparable with the reference. Furthermore, the UV-Vis spectra (Figure 4, panel D) of the recovered cellulose acetate displayed evidence that the optical properties were also retained, as the samples showed absorption only in the UV region (<300 nm of wavelength, i.e., in the UVC zone) and high transmission in the visible region [44].

Table 2. Degree of substitution (DS) calculated by FT-IR along with molecular weight outputs from GPC of unsmoked samples and ballot bin waste. The Mn, Mw, and Mp values reported are the average of three analyses (p = 0.05, n = 3). D is the dispersity index determined by GPC analysis. --- analysis not performed.

	Treatment	DS	M_p (10^3) g/mol	M_n (10^3) g/mol	M_w (10^3) g/mol	D
Unsmoked filter	---	2.10 ± 0.1	343 ± 15	423 ± 19	1750 ± 180	4.1
	H_2O washing	2.12 ± 0.1	353 ± 20	474 ± 21	1822 ± 190	3.8
	Ethanol washing	2.11 ± 0.1	346 ± 22	454 ± 26	1721 ± 175	3.8
	Acetone	2.10 ± 0.1	343 ± 16	487 ± 18	1684 ± 185	3.5
Ballot bin waste	---	2.07 ± 0.2	---	---	---	---
	H_2O washing	2.10 ± 0.3	---	---	---	---
	Ethanol washing	2.11 ± 0.2	---	---	---	---
	Acetone	2.07 ± 0.3	369 ± 25	427 ± 22	1594 ± 225	3.7

It is worth mentioning that the extraction process proposed herein meets the environmental goals of green chemistry using green solvents in a simple process (Figure 5).

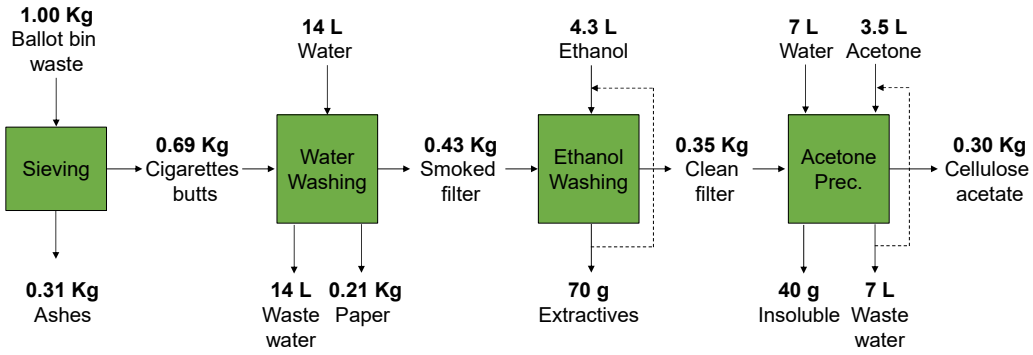

Figure 5. Schematic diagram for the purification process of 1 kg of ballot bin waste along with mass flow data experimentally determined, input (BBWs), and output materials.

In addition, in 2023 and 2024, Zuccante et al. [66,67] explored transforming the whole cigarette and various fractions of this process, such as ashes, paper, and filters, into an oxygen reduction reaction electro-catalyst, further enhancing the concept of the circular economy and reducing waste production. Additionally, the ethanol extractives showed potential as a tool against insecticide-resistant mosquito vectors [68,69]. The ability to

recycle solvents like ethanol and acetone minimizes the process's environmental impact, with wastewater (62 L/kg) treatment being competitive in terms of water consumption compared to producing (only processing water for cellulose acetylation was considered) (76 L/kg) virgin cellulose acetate [70]. Finally, the effective monitoring and enforcement of these standards are essential to promote adherence, accountability, and sustainable waste management.

4. Conclusions

In light of the achieved results, it should be underlined that the extraction of CA from cigarette butts presented herein is an effective method for obtaining a high-quality semi-finished CA with the same physical and chemical properties as an unsmoked cigarette filter. This is an interesting achievement, especially from the circular economy point of view, since the ballot bin waste was converted into a high-quality product whose chemical characteristics could permit the recycling of a semi-finished cellulose acetate product of great interest from an industrial point of view, since it can be further processed for several applications. In addition, the implemented collection strategy could avoid and/or reduce littering and the enormous environmental problems related to the potential release of microplastic fibers and toxic substances such as heavy metal, nicotine, carcinogenic tar, and polycyclic aromatic hydrocarbons. With this comprehensive approach, cigarette waste can be transformed from an environmental burden into an opportunity for sustainable resource recovery.

Supporting research and development is another cornerstone, as funding and collaboration with academic institutions drive innovations in recycling technologies, making the recovery of valuable materials like cellulose acetate (CA) more efficient. Notably, the extraction of CA from cigarette butts has proven to be an effective method for obtaining a high-quality semi-finished CA that retains the same physical and chemical properties as unsmoked cigarette filters. This achievement is particularly significant from a circular economy perspective, as it transforms waste from ballot bins into a high-quality product suitable for various industrial applications. Integrating circular economy principles into these policies shifts the focus from waste disposal to resource recovery, benefiting both ecosystems and the economy. The implemented collection strategy not only addresses the challenge of littering, but also mitigates the environmental hazards associated with the potential release of microplastic fibers and toxic substances, including heavy metals, nicotine, carcinogenic tar, and polycyclic aromatic hydrocarbons. Moreover, the proposed extraction process aligns with the environmental goals of green chemistry by utilizing green solvents in a straightforward method. The achievement of the objectives in terms of the yield of the purified acetate (about 30%), the quality of the semi-finished product with a DS of 2.0-2.5 comparable to virgin acetate, the removal of hazardous substances (70 g ethanol extracts), and the excellent removal of heavy metals, 91% copper (Cu), 59% chromium (Cr), and 92% iron (Fe), demonstrates the effectiveness of the CA purification process.

Author Contributions: N.L.: methodology, writing—original draft preparation. G.Z.: data curation, writing—reviewing and editing. A.F.: conceptualization, supervision. C.S.: conceptualization, writing—reviewing and editing, supervision. M.R.: conceptualization, supervision. L.Z.: conceptualization, writing—reviewing and editing, supervision. All authors have read and agreed to the published version of the manuscript.

Funding: The cigarette butts fractionation has been performed within the MUSA—Multilayered Urban Sustainability Action—project, funded by the European Union—NextGenerationEU, under the National Recovery and Resilience Plan (NRRP) Mission 4 Component 2 Investment Line 1.5: Strengthening of research structures and creation of R&D "innovation ecosystems", set up of "territorial leaders in R&D".

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

Acknowledgments: G.Z. acknowledges a Ph.D. scholarship on the Italian National Ph.D. program "Scientific, technological and social methods enabling circular economy", Curriculum "Technical materials for circularity" funded by EU's Recovery and Resilient Plan. C.S. would like to acknowledge also the Cariplo Foundation, Call for Circular Economy through the project "Transformation of plastic waste in Electrocatalysts, Supported by exhausted gases recovery Layout" (TESLA).

Conflicts of Interest: The authors declare no conflicts of interest.

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