Wiley
Advances in Polymer Technology
Volume 2025, Article ID 8477994, 8 pages
https://doi.org/10.1155/adv/8477994

WILEY

Research Article

Chemical Procedures for Paint Removal in Used Thermoplastics for Revalorization as a Raw Material in the Automotive Industry

Carla Zambrano Membrives,[1,2] Pablo Tamarit,[1] Ana Inés Fernandez,[2] and Camila Barreneche[2]

[1] *CITSALP, S.L., Calle Antonio Gaudi, 74, Nave 5 Pl. Ind. Rubi Sur., Rubí 08191, Barcelona, Spain*
[2] *Departamento de Ciencia de los Materiales y Química Física, Facultad de Química, Universitat de Barcelona, C/Martí i Franquès 1 08028, Barcelona, Spain*

Correspondence should be addressed to Camila Barreneche; c.barreneche@ub.edu

Received 24 October 2024; Accepted 20 February 2025

Academic Editor: Chenggao Li

The recycling of thermoplastic polymers from end-of-life vehicles is crucial to achieve a circular economy within the sector and prevent the accumulation of plastic waste by reusing it. However, several challenges in these processes present difficulties, making it challenging or impossible to recycle these materials for the same applications, impeding the closure of the life cycle and valorization of waste. The primary problem faced by plastic converters is the presence of superficial paint. In this study, we evaluate the implementation of chemical methods with varying conditions to remove paint from used bumpers thermoformed with polypropylene, with the aim of valorizing the plastic waste from these bumpers. We examine the various process variables, such as reagent concentration, temperature, time, and pH. Additionally, we analyze procedures to quantify the paint content in the different recycled samples and use this as a tool to compare the effectiveness of different paint removal processes.

Keywords: automotive; paint; polymers; recycling; sustainability

1. Introduction

1.1. Recycling of Automotive Plastics. The production of plastic and the amount of polymer waste have doubled worldwide, resulting in 460 million tons in 2019, of which only 9% was recycled, 19% was incinerated, and nearly 50% was sent to landfills. The remaining 22% ended up in uncontrolled landfills or entered the ecosystem [1]. Several waste management associations in the European Union indicate as efficient starting points the design of products that favour recycling, the management of exports at the end of their useful life, the end of the landfill of plastic waste, and the expansion of disposal and recycling systems [2].

The automotive industry is one of the major users of plastics, with about 10%–15% of the weight of a vehicle being comprised of plastic materials and composites [3]. Therefore, a specialized design of management of this waste is essential at the end of the vehicle's useful life. The recycling of end-of-life vehicles (ELVs) is one of the most efficient ways to manage automotive plastic waste, and in the European Union, 80%–90% of ELVs can be recycled [4]. In 2019, it was reported that end-of-life recycled vehicles in the European Union (EU) had a total weight of 6.9 million tonnes [5].

In the automotive sector, postindustrial and postconsumer recycling are the two main sources of raw material for polymer recycling. Different types of recycling methods exist for reusing these polymeric raw materials, including mechanical, chemical recycling, and energy recovery. Chemical recycling converts polymers into their source monomers, which are then used to synthesize the same or different polymers through subsequent polymerizations. In contrast, energy recovery involves transforming plastic waste into energy in the form of heat, steam, or electricity. Mechanical recycling entails a series of operations that transform waste into raw materials through collection, sorting or separation, washing, shredding, agglutinating, extruding, molding, injecting, or thermoforming. The preference is to use mechanical and chemical recycling over energy recovery, except when recovery processes fail due to economic or technical restrictions. Mechanical recycling is the most used method in automotive industry [6].

Mechanical recyclers face several problems, such as degradation, heterogeneity (different polymers, complex multilayer structures, reinforced, or assembled), and contamination by impurities of another type of polymer or chemicals such as adhesives, paints, or others requiring pretreatments of classification and cleaning [7]. The nature of automotive paint depends on the type of plastic to be painted and its characteristics, such as porosity, surface roughness, and surface tension [8]. These paints comprise various components, including binders, resins or polymers, fillers, pigments, solvents or thinners, additives, and some varnishes [9]. The resins are typically alkyd, acrylic, amine, polyurethanes, epoxy, chlorinated rubber, silicone, or silicates. Likewise, the most used pigments in automotive paint are titanium dioxide, barium sulfate, silicon dioxide, zinc oxide, zinc chromate, etc. The solvent used can be aqueous or organic solvents such as alcohols, glycols, esters, hydrocarbons, among others. This component is the volatile part of the paint and disappears once the dry film is obtained. Additives contain several components that provide specific characteristics to the paint, such as moisturizers, dispersants, antioxidants, etc.

The presence of paint in the recycling process leads to difficulties at various stages. In mechanical recycling, the sorting stage is particularly problematic, as paint interferes with the density of painted polymers and makes it difficult to separate them by density differences between mixtures of different polymers. It also reduces the quality of recycling due to the presence of colored spots on the surfaces of the thermoformed parts, among other effects on the material's mechanical properties.

Over the years, several studies have attempted to eliminate the amount of paint on polymer substrates using different chemical methods. In 1995, Löhr et al. [10] explained a chemical stripping procedure from alkalized ethylene glycol and polypropylene glycol for plastic bumpers. In 2001, Gecol et al. [11] used anionic, cationic, zwitterionic, and nonionic surfactants to remove dyes from plastic films. In 2002, Songsiri et al. [12] used a cationic surfactant to remove solvent-based ink on high-density polyethylene surfaces, studying experimental parameters affecting depainting. In 2004, Seong Taek Na et al. published a patent [13] in which they developed an alkalized phenoxyethanol and isopropyl alcohol procedure for the depainting of bumpers painted with thermoset paint. In 2007, Chotipong et al. [14] used various alkyltrimetylamonium bromides to remove binder ink of an epoxy nature from HDPE bottle surfaces and the effects of pH, temperature, and salinity were observed. In 2010, Montie [15] patented the use of various chemical fluids to remove the paint from plastic substrates.

However, none of the techniques has been entirely satisfactory since factors such as efficiency, performance, economic and industrial viability, or nondegradation of the polymer of interest are not met in each of the procedures. In addition, in many cases, the mechanism of action of these substances is unknown.

The present study aims to assess chemical procedures for eliminating the amount of paint contained in recycled polyolefins, specifically polypropylene (PP), to revalue it in later applications after its end of life within the automotive sector. The study aims to optimize some of these methods by varying the experimental conditions and considering factors such as environmental impact, toxicity, possible damage to the substrate of interest, and the need to handle hazardous chemicals. Additionally, it aims to elucidate the mechanism of action of these methods on the material.

2. Materials and Methods

2.1. Materials. Polypropylene-based postindustrial recycled bumpers with a 20% talc load and painted in different colours, such as red, white, blue, gray, and black, were used as samples. It is assumed that the difference in sizes of the samples treated with the chemical procedures are not significant differences for the results obtained since they have irregular and similar sizes.

The following reagents were employed: sodium dodecyl sulphate (SDS), hexadecyltrimethylammonium bromide (CTAB), sodium hydroxide (NaOH), benzyl alcohol, polyethylene glycol, potassium hydroxide (KOH), phenoxyethanol, and isopropyl alcohol. Deionized water was used throughout the experiments.

2.2. Methods: Paint Removal

2.2.1. Procedures to Remove the Amount of Paint. Procedures for paint removal involved the use of chemical methods, typically using reflux and stirring in an aqueous solution. Various experimental conditions were employed to assess the effectiveness of the depainting process. Factors such as the concentration of the reagent, temperature, stirring time, and pH were found to impact the effectiveness of the depainting procedure. Table 1 provides a summary of the experimental conditions used in this study.

Procedure A involves immersing the samples in a CTAB surfactant solution, which is then alkalinized with NaOH at different concentrations, temperatures, times, and pH levels during reflux at high temperatures. Procedure B is similar to Procedure A, but it involves immersing the samples in a mixture of CTAB and SDS surfactants using the same procedure. Procedure C is also an immersion bath, but it uses a mixture of deionized water with benzyl alcohol at different temperatures for 2 h. The solution is then alkalinized with NaOH until it reaches a pH of 12 and is kept under reflux at high temperatures without adjusting the pH. Procedure D involves immersing the samples in a solution of alkalinized polyethylene glycol at 60°C. Procedure E is an immersion bath that uses a mixture of phenoxyethanol and isopropyl alcohol, which is then alkalinized with KOH in different proportions and times at 70°C while being stirred with ultrasound.

2.2.2. Characterization of Samples. To analyze the initial quantity of paint in the samples, as well as assess the

Table 1: Summary of experimental conditions used for chemical paint removal procedures.

	Concentration of reagent (%)	Temperature (°C)	Time (h)	pH
A (CTAB, NaOH, deionized water)	2.0	40.0	3.0	>12
	5.0	—		10
	10	115		
B (CTAB, SDS, NaOH, deionized water)	2.5	80.0	3.0	>12
	5.0	115	1.0	
C (Benzyl alcohol, deionized water)	50/50	80.0	2.0	>11
		115		
D (Polyethylene glycol, KOH)	PEG 400	60.0	15 min	>10
			1.0	
E (Phenoxyethanol, isopropyl alcohol, KOH)	70/30	70.0 (ultrasonic bath)	1.0	12
	90/10	115	3.0	

Abbreviations: CTAB, hexadecyltrimethylammonium bromide; SDS, sodium dodecyl sulphate.

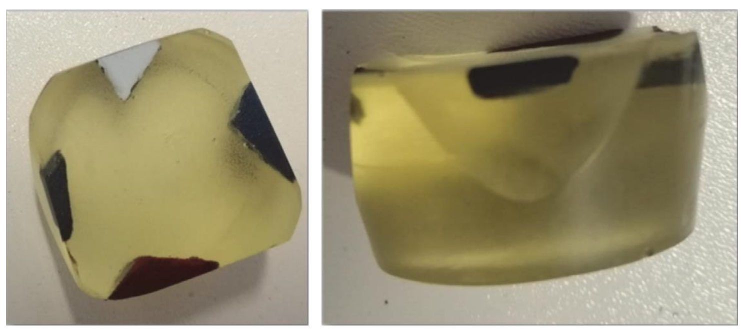


Figure 1: Images of the shredded embedded in the epoxy resin.

Table 2: Description of the paint adhesion categories following the ISO 2,409:2020.

Category	Summary
0	No changes
1	The paint becomes slightly more pliable, but its removal still requires vigorous scraping
2	While the paint has become slightly more pliable, removal now requires less aggressive scraping
3	The paint is easily removed with minimal abrasion
4	Removal of the paint can be accomplished effortlessly by running a finger over it
5	The paint has completely deteriorated, resulting in the fading of the underlying material

effectiveness of depainting and adhesion of the painted samples, a range of analytical tools was employed. These included digital imaging using an electron microscope, spectrocolorimeter, vernier callipers, and the cross-cut test.

To determine the average weight percentage of paint in the shredded samples, multiple statistical samples were taken. The thickness of the paint layer was measured using electron microscope images of the samples embedded in epoxy resins as shown in Figure 1.

The thickness of the PP substrate, the area covered by paint, the percentage of volume, and weight of paint were also measured. Furthermore, adhesion categories were assigned based on the appearance of the cross-cut test results, as summarized in Table 2.

The study includes an analysis of the effects of heat treatments at different temperatures on the adhesion of paint to a polypropylene substrate over a 1-h period. Adhesion is measured using the cross-cut test, a standardized method defined in ISO 2,409 : 2020. Additionally, the efficacy of the paint removal procedures is evaluated by analyzing digital images of the treated samples using open software *Image J*. The % efficacy of depainting is calculated using the following equation:

$$\%\ \text{Efficacy of Depainting} = 100 - \left(\left(\frac{\text{paint pixels (unpainted sample)}}{\text{paint pixels (painted sample)}} \right) \times 100 \right). \quad (1)$$

The image processing of one of the decoated samples by procedure A is shown in Figure 2.

FIGURE 2: Digital conversion of unpainted sample images.

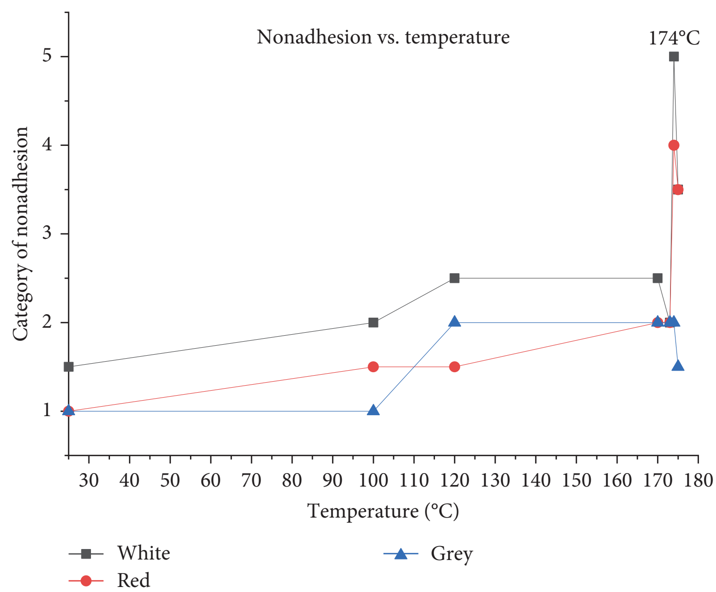


FIGURE 3: Nonadhesion vs. temperature between paint and polypropylene substrate of bumper samples.

The efficacy of depainting procedures that alter color intensity without resulting in partial or total paint removal is quantified using the CIELAB method [11] with a spectrocolorimeter. This method enables comparison of the efficacies of different colors using the same procedure. The CIELAB color space, recognized by the International Lighting Commission (CIE), provides an absolute theoretical reference for light components. The L* component represents luminosity, ranging from 0 (black) to 100 (white). The a* component represents the gamma of coordinates from red (positive) to green (negative), while the b* component represents the gamma of coordinates from yellow (positive) to blue (negative). The spectrocolorimeter used in this study was calibrated to the D65 illuminator, representing daylight with a color temperature of 6500K and a standard observer of 10°. To quantify the efficacy of depainting, the total color difference between the standard (unpainted sample), painted sample, and decoated sample is calculated using the DE* equation:

$$DE^* = [(DL^*)^2 + (Da^*)^2 + (Db^*)^2]^{1/2},$$

where:

$$\begin{aligned} DL^* &= L^*(\text{sample}) - L^*(\text{standard}) \\ Da^* &= a^*(\text{sample}) - a^*(\text{standard}) \\ Db^* &= b^*(\text{sample}) - b^*(\text{standard}) \end{aligned} .$$

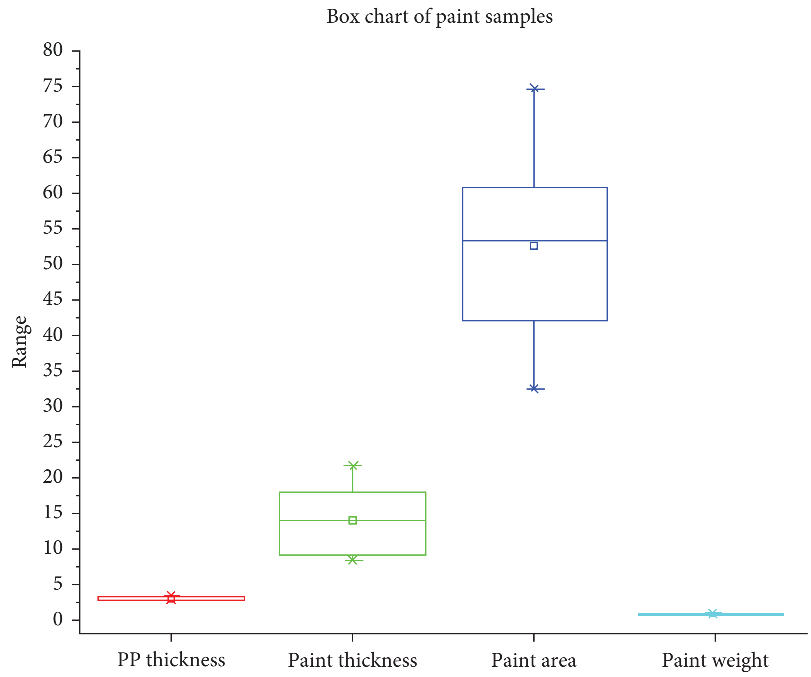
Here, "sample" refers to the painted or decoated sample, while "standard" refers to the reference sample without paint. A higher DE* value indicates a greater level of depainting.

3. Results and Discussion

3.1. Characterization of the Samples to be Stripped. Figure 3 shows the effect of temperature on the adhesion between the paint and substrate, categorized as defined in Table 2. At temperatures higher than 174°C, the polymer matrix becomes too viscous because it has reached melting temperature, resulting in a more complex separation process. At 173°C–174°C, the paint softens, making it easier to remove through friction, weathering, or shear. Additionally, we observed that white paint exhibits lower adhesion to the polymeric matrix compared to other paints tested due to its unique composition.

Figure 4 presents the average PP and paint thicknesses in terms of paint area, volume percentage, and weight percentage. We report both the average and the range of deviation for each sampling measurement. The paint area has the highest error percentage due to the irregular shredded areas resulting from the uncontrolled cuts of the blade mills, as expected. On average, the weight percentage of paint is 0.49%.

The average particle size of shredded pieces is around 5 mm (range from 1 to 10 mm), with a granulometry below 12 mm.



Total N	12			
Mean	2.914 mm	14.01 µm	52.60 mm	0.487 %
Standard Deviation	0.254	4.684	13.42	0.171
Minimum	2.690	8.360	32.50	0.271
Maximum	3.420	21.63	74.75	0781

Figure 4: Box charts of statistics about geometric characterization of shredders samples with paint.

3.2. Efficiencies of Paint Removal Procedures. The efficiencies of paint removal procedures are presented in Table 3, which also shows the change in appearance between a painted and a decoated sample.

The best efficacy of 97.13% was achieved with procedure A using alkalized cationic surfactant (CTAB) at a reagent concentration of 10% in an aqueous-based solution and a basic pH of 13.5 for 3 h. This process is driven by surface tension, surfactant adsorption, wetting, dispersion, and solubility phenomena [12]. The surfactant is adsorbed on the surface of the polymer and ink, decreasing the interfacial tension of both the polymer–water and ink–water contact. Figure 5 shows the detachment of the paint due to the action of the surfactant in the samples.

The CTAB is more effective than the mixture with SDS (anionic surfactant) because the binder of the paint contains the acrylate ion of the acrylic resins in solution, forming an anion and being more compatible with cationic surfactants. Shear forces or friction on the surface aid the detachment of the paint, and the surfactant improves moisturization by forming a water layer between the ink and the polymer surface once the paint is released. Procedure B could not be tested with a reagent concentration higher than 5%, unlike procedure A, because the solution remained saturated and foamed up the reflux column.

Procedures C, D, and E resulted in partial paint elimination, with efficiencies of 43.99%, 2%–5%, and 49.44%, respectively. The C procedure with benzyl alcohol, when the solution is not alkalized, only presented a change in the color intensity of the paint and not a partial or total elimination of it, so the efficacy was calculated with the CIELAB method and not with the pixel count. The efficiencies of each color differ because of different formulations (at least the pigment) and painting processes. In different conditions, red and white colors presented higher efficiencies due to lower adhesion of the paint with the polymeric matrix.

Regarding the effect of these chemical treatments on the polymer properties, according to study of Hirokazu et al. [16], a change in the modulus was observed up to 21 days with NaOH, maintaining ~2 GPa without treatment and increasing slightly to 2.2 GPa with NaOH. The effect of alkaline treatments, as in procedures A, D and E, is considered and expected low since the exposure time in this article is significantly shorter than the treatment durations reported in the literature.

4. Conclusions

In general, the process of chemical depainting of polymer surfaces is based on the detachment of the paint and its dispersion in a washing bath through mechanical action. The effectiveness of the depainting procedures is influenced by factors such as the time and speed of agitation, pH, reagent concentration, and temperature. Higher pH values lead to increased negative

Table 3: Efficacies of the chemistry stripping procedures with its conditions and images of real samples before and after chemical stripping using process A.

Procedures	Experimental conditions	Results of efficacies		Comments on the calculation of efficacies
A (CTAB, NaOH, deionized water)	5% 40°C pH = 13.5 3 h	0%		By pixel count
	5% 115°C pH = 13.5 3 h	95.16%		
	10% 115°C pH = 13.5 3 h	97.13%		
	2% 115°C pH = 13.5 3 h	70.99%		
	2% 115°C pH = 10 3 h	40.98%		
B (CTAB, SDS, deionized water)	2.5% 115°C pH > 12 1 h	0%		
	2.5% 115°C pH > 12 3 h	0%		
	2.5% 80°C pH > 12 3 h	0%		
	5% 80°C pH > 12 3 h	0%		
C (benzyl alcohol, deionized water)			DE*	
	50:50 80°C 2 h	Blue	5.133	*By CIELAB* (causes swelling in the paint and changes in its intensity)
		Red	20.02	
		White	6.934	
		Silver	1.491	
	50:50 115°C 2 h	Blue	5.475	*By CIELAB* (causes swelling in the paint and changes in its intensity)
		Red	26.40	
		White	28.88	
		Silver	4.121	
	50:50 115°C pH > 11 2 h	43.99%		*By pixel count*
D (polyethylene glycol, KOH)	60°C pH > 10 1 h	0%		*By pixel count*
		2%–5%		
E (phenoxyethanol, isopropyl alcohol, KOH)	90:10 70°C pH = 12 1 h	0%		*By pixel count*
	90:10 70°C pH = 12 3 h	0%		
	90:10 115°C pH = 12 3 h	49.44%		

Abbreviations: CTAB, hexadecyltrimethylammonium bromide; SDS, sodium dodecyl sulphate.

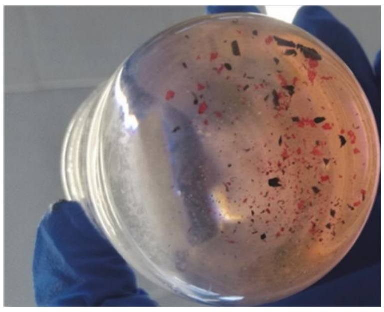


Figure 5: Base of the balloon with the CTAB solution after depainting shredded polypropylene. CTAB, hexadecyltrimethylammonium bromide.

charges on both the impurity and substrate, promoting repulsion between them. Increasing the temperature reduces the adhesion of the paint to the polymer, as demonstrated by the cross-cut test.

Multiple chemical techniques have demonstrated effective depainting of thermoplastic bumpers, with the utilization of cationic surfactants exhibiting a superior efficacy rate of 97.13%. Using aqueous solutions of surfactants instead of organic solvents for decoating plastics can lead to significant environmental benefits. One of the challenges in understanding the action of surfactants in the striping of recycled materials is the complexity of the patented formulations of paints and their components, as well as the poor traceability of their origin after use or industrial production. However, an understanding of the surfactant characteristics and the composition and nature of the polymer matrix and paints provides insight into the depainting process.

Given the multiple combinations of materials, copolymers, additives, fillers, and variability in paint compositions, optimizing depainting procedures for each composition is necessary to increase the effectiveness of the depainting process. Standardizing optimal depainting methods for each situation is essential, given that the adhesion of each type of paint also varies.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest.

Funding

The authors would like to thank the Catalan Government for the quality accreditation and funding given to their research group DIOPMA (2021 SGR 00708).

The authors would also like to acknowledge the financial support provided by the *Agencia de Gestió d'Ajuts Universitaris i de Recerca* (AGAUR) through the Industrial Doctorate (DI) Grant 2020 DI 017, which made this work possible.

Acknowledgments

The authors would like to thank the Catalan Government for the quality accreditation and funding given to their research group DIOPMA (2021 SGR 00708). DIOPMA is a certified agent TECNIO in the category of technology developers from the Government of Catalonia. Additionally, a special thanks to CITSALP S.L. company, which supported the project by providing material.

The authors would also like to acknowledge the financial support provided by the *Agencia de Gestió d'Ajuts Universitaris i de Recerca* (AGAUR) through the Industrial Doctorate (DI) Grant 2020 DI 017, which made this work possible.

References

[1] OECD, *Global Plastics Outlook: Economic Drivers, Enrivonmental Impacts and Policy Options* (OECD Publishing, Paris, 2022).

[2] Archive Ellen McArthur, "Ellen McArthur Foundation," https://archive.ellenmacarthurfoundation.org/explore/plastics-and-the-circular-economy, Accessed: May 08, 2022.

[3] A. Patil, A. Patel, and R. Purohit, "An Overview of Polymeric Materials for Automotive Applications," *Materials Today: Proceedings* 4, no. 2 (2017): 3807–3815.

[4] ARA, *Automotive Recycling Industry: Environmentally Friendly, Market Driven, and Sustainable* (Automotive Recyclers Association, 2012).

[5] Eurostat, "End-of-Life Vehicle Statistics," https://ec.europa.eu/eurostat/statistics-explained/index.php?title=End-of-life_vehicle_statistics#Total_weight_of_end-of-life_vehicles, Accessed: Apr 04 2022.

[6] S. M. Al-Salem, P. Lettieri, and J. Baeyens, "Recycling and Recovery Routes of Plastic Solid Waste (PSW): A Review," *Waste Management* 29, no. 10 (2009): 2625–2643.

[7] M. B. Biddle and P. Dinger, *An Overview of Recycling Plastics from Durable Goods: Challenges and Opportunities* (Identi-Plast II, Brussels, 1999): 1–25.

[8] K. B. Ferreira, A. G. G. Oliveira, and J. A. Gomes, "Raman Spectroscopy of Automotive Paints: Forensic Analysis of Variability and Spectral Quality," *Spectroscopy Letters* 50, no. 2 (2017): 102–110.

[9] J. Calvo Carbonell, *Pinturas y Recubrimientos: Introducción a Su Tecnología* (Díaz de Santos, Madrid, 2009).

[10] K. Lohr, B.-U. Kettemann, Z. Jorg, and M. Melchiore, "Chemo-Mecanical Stripping Painted of Paint from Plastic Parts," (1996).

[11] H. Gecol, J. F. Scamehorn, S. D. Christian, and F. E. Riddell, "Use of Surfactants to Remove Solvent-Based Inks from Plastic Films," *Colloid and Polymer Science* 189, no. 1–3 (2001): 55–64.

[12] D. Songsiri, S. S. Min, J. F. Scamehorn, S. Osuwan, and J. W. Ellis, "Use of Cationic Surfactant to Remove Solvent-Based Ink from Rigid High Density Polyethylene Surfaces," *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 204, no. 1–3 (2002): 261–269.

[13] J. H. Hong and S. T. Na, "Hong and Seong Taek Na, Chemical Method of Removing Paint Film on Plastic Resin Using Isopropyl Alcohol," (2004).

[14] A. Chotipong, J. F. Scamehorn, T. Rirksomboon, S. Chavadej, and P. Supaphol, "Removal of Solvent-Based Ink from Printed Surface of High-Density Polyethylene Bottles by Alkyltrimethylammonium Bromides: Effects of pH, Temperature, and Salinity," *Colloids and Surfaces* 297, no. 1–3 (2007): 163–171.

[15] P. Montie, "Process for Removal of Paint from Plastic Substrates," (2010).
[16] H. Wada, Y. Suzuki, K. Okamoto, and M. Ishikawa, "Effects of Chemical Solutions on the Toughness of Polypropylene," *Polymer Journal* 37, no. 12 (2005): 877–886.