



Article Environmental Sustainability of Solvent Extraction Method in Recycling Marine Plastic Waste

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Abstract: The global plastic production of 400 million tons/year has caused major catastrophes in marine environments. The current study, therefore, aimed to mitigate this challenge through the dissolution–reprecipitation method of eradicating impurities and contaminants from marine plastic debris. The results revealed that the rate of the dissolution of polyethylene (PE) outweighed polypropylene (PP) at lower temperatures. HDPE (high density polyethylene) and PP had optimal dissolution temperatures of 75 °C and 90 °C at 20 and 30 min, respectively, resulting in recovery percentages of 96.67% and 87.35% when applied to actual marine waste samples. Overall, this recycling method conserved the plastic quality and properties, making it a viable alternative for virgin plastics. The life cycle assessment (LCA) revealed that the drying stage demonstrated the greatest environmental impact within the system. The overall process, however, yielded a lower environmental impact in comparison with established findings. Conclusively, the current study has successfully restored marine plastic waste with high recovery rates and minimum chemical alterations, yielding a low environmental footprint.

Keywords: plastic waste; marine environment; solvent extraction; sustainability; life cycle assessment



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1. Introduction

Plastic production has surged from 2.3 million tons (MT) in the 1950s to 448 MT in 2015, and this quantity is anticipated to double by 2050, attributed to urbanization as well as human lifestyles changes [1]. A substantial percentage of plastic production is allocated to the manufacturing of packaging and other goods that have a relatively short lifespan, resulting in their disposal within a period ranging from one month to one year. Likewise, due to the long-lasting nature of polymer entanglement, massive amounts of "afterlife" plastics are piled up as litter in landfills as well as in natural environments, e.g., marine. In fact, coastal nations dump about 8 MT of plastics into the oceans annually [2]. Considering these facts, the current plastic usage is unsustainable, as its manufacturing, use, and disposal have detrimental impacts that remain and accumulate interminably in the environment.

In general, recycling is one of the approaches that has been taken in order to mitigate these problems and has been implemented in a lot of countries worldwide today. The methods of recycling include primary recycling, secondary (mechanical recycling and physical recycling), and tertiary recycling (chemical recycling or feedstock recycling) [3]. Consequently, a few challenges are associated with the conventional methods of recycling plastic waste. General problems include (1) the difficulty of recycling waste plastics, as they are extremely versatile and may be created in a lot of diverse ways, which (2) involves the use of elevated temperatures, which consume more energy, as well as toxic solvents, and (3) emits more pollutants than normal waste incineration [4]. Besides, up until recently, the main concern was that recycled products produced from waste plastic experience a deterioration in terms of purity and quality due to the use of colorants, additives, as well as fillers in the production of plastic, the contamination caused when disposed of improperly, and oxidation due to the exposure of UV radiation [5]. Previous studies have shown that traditional recycling methods cannot eliminate impurities and contaminants at the molecular level, which can result in recycled polymers losing their virgin qualities [6]. For example, mechanical recycling frequently includes mechanical processes, such as shredding and washing, to cleanse plastic waste. Although these processes demonstrate efficacy in eliminating the observable contaminants, they may not adequately tackle impurities that are imbedded within the plastic matrix or are present at the molecular scale [7]. In addition, mechanical recycling cleaning solutions, such as water and detergents, exhibit efficacy in eliminating contaminants present on the surface of plastics. However, their effectiveness diminishes when it comes to eliminating or dissolving impurities that have formed bonds with the plastic at a deeper level [8]. A recent study demonstrated the possibility of enhancing the mechanical recycling rate by taking into account the viable secondary material, resulting in a possible recovery of up to just 31% [9]. Another study revealed that the process efficiency for both conventional and improved mechanical recycling techniques displayed a resemblance, with values ranging only from 64% to 66% [10]. Although mechanical recycling is widely acknowledged as a successful approach, it may pose detrimental impacts on plastic materials. These impacts manifest as variations in the plastic's texture, color, and characteristics [11]. Consequently, this provides a significant barrier when attempting to recycle plastic over several cycles. Plastic waste in traditional recycling facilities is frequently subjected to batch processing or mixed streams, posing difficulties in properly isolating and addressing contaminants within a particular plastic type [12]. These challenges further oppress the repurposing of marine plastic waste through traditional recycling methods.

Solvent-based extraction methods have emerged as a promising solution, as they can selectively remove impurities and contaminants at the molecular level, ensuring the recycled polymers retain their virgin qualities [13]. Solvent extraction techniques have a high degree of versatility and demonstrate efficacy in tackling the issue of mixed plastic waste, a formidable obstacle for traditional recycling methodologies [14]. The selection of solvent can be made in order to effectively dissolve and extract contaminants from different kinds of plastic concurrently [15]. The dissolution–precipitation process operates

by undergoing four distinct phases, namely dissolution, separation, reprecipitation, and recycling [8]. During the initial stage, the selection of the solvent is determined by its capacity to preferentially dissolve the specific plastic material of interest, while ensuring that impurities and contaminants, such as particulate contaminants, residual polymers, or additives, remain unaffected. Subsequently, the solution undergoes separation techniques, such as filtering or centrifugation, in order to physically eliminate the solid impurities present [10]. The clean plastic solution, devoid of any contaminants, undergoes reprecipitation to yield solid particles, which are subsequently isolated from the solvent. The adaptability of this technology offers a viable approach for handling heterogeneous plastic streams, a challenge in which traditional technologies frequently encounter difficulty in attaining comparable levels of purity [16].

However, it is important to note that conducting the overall recycling system at low temperatures and shorter time durations is beneficial, since the high temperatures that are executed during the dissolution process can hasten the degradation of the recovered polymers from plastic waste, and shorter processing times can contribute to energy efficiency in the recycling process [17]. Despite existing studies that have examined the benefits and efficiency of this recycling method, there remains a significant gap in the comprehensive understanding of its environmental impacts [18]. The absence of comprehension in this context involves the possibility of trade-offs and unanticipated repercussions, including the transfer of environmental burdens that may emerge during the complete life cycle of the recycled material.

The main objective of this study is to investigate the recycling of marine plastic debris using the solvent extraction method, specifically emphasizing the dissolution–reprecipitation process. The study encompassed a range of temperature conditions over a period of time and entailed comprehensive characterization in order to assess changes in the structure. This study has also successfully validated the efficacy of this recycling approach through a rigorous instrumental analysis, thereby establishing a novel basis for its practical implementation. The comprehensive life cycle assessment (LCA) conducted in this study not only provided a quantitative analysis of the environmental consequences associated with the solvent extraction technology but also yielded a profound comprehension of its ecological footprint. The aforementioned insights hold great significance in facilitating well-informed decision-making within the domain of sustainable waste management.

2. Materials and Methods

2.1. Materials

Model polyolefins and various products in pellet form were collected from a marine environment (Teluk Batik, Perak, Malaysia). Materials consisted mostly of high-density polyethylene (HDPE) and polypropylene (PP), and were hand-sorted arbitrarily. Similar commercial polymers (virgin materials) were purchased from Sigma Aldrich. The analytical-grade solvent, toluene and acetone (Merck, Germany), with >99.7% purity, was employed in this study. The solvent selection was performed based on the ability to dissolve a wide range of plastic materials [19].

2.2. Analytical Analysis

2.2.1. ATR-FT Instrument

FT-IR Spectrometer (ATR method) analysis was conducted using Perkin Elmer, Spectrum 100, Waltham, MA, USA, to assemble spectra from 4000 cm⁻¹ to 650 cm⁻¹. The resolution was fixed at 4 cm⁻¹ [20]. The diamond crystal was sterilized with acetone and a background scan was executed. The surface of the sample was analyzed before and after the recycling process [21]. Every sample was pressed against the diamond with a force ranging from 75–80 N to assure that there was an adequate contact between the sample and the ATR crystal [22]. A peak height method was employed to identify absorption bands that were collected and contrasted to absorption bands reported from several reli-

able/credible references. For an identification to be considered valid, there needed to be at least 4 matched absorption bands [23].

2.2.2. Differential Scanning Calorimetry

The differential scanning calorimeter (DSC) can be used to analyze the thermal behavior of polymers [24]. DSC Q2000 V24.11 Build 124 was used to obtain the thermograms of virgin materials and recycled marine waste. Model polymers, marine waste plastic products, and recycled marine plastic products had their melting temperatures (T_m) determined for this research. The measurements were conducted with temperature range between 20–300 °C for both HDPE and PP [25]. Sample weights varied from 1–3 mg, while heating rate was fixed at 10 °C/min [26].

2.2.3. Dissolution–Reprecipitation Technique

Commercial virgin HDPE and PP served as a standard in determining the recycled plastic performance. These virgin polymers were comparatively cross examined with recycled plastic to gauge the impact of various temperatures and the influence of varying durations on dissolution rates. The evaluation encompassed three distinct dissolution temperatures, which were 60, 70, and 90 °C, each paired at three different durations, 10, 20, and 30 min [27]. The solvent volume for both toluene and acetone were fixed at a 1:1 ratio and the dissolving temperatures were kept below 110 °C, the boiling point of the toluene solvent, to minimize the risk of plastic degradation [28]. Acetone was employed in the reprecipitation process as a non-solvent or weak solvent. A 250 mL glass reaction vessel outfitted with a magnetic stirrer, reflux condenser, and thermometer were used in the experiment, to which 1 g of polymer and 20 mL of solvent was added [29]. A silicon oil bath was employed to control and maintain the desired temperature [30]. The system was subjected to heating in accordance with the specified temperatures and time. The polymer was then introduced into the non-solvent after the flask had been cooled. Following re-precipitation, rinsing, and filtration processes, the polymer underwent drying in an oven set at a temperature of 100 $^{\circ}$ C, for a duration of 20 h [31]. The polymer that has been successfully recovered/recycled generally manifests in the form of a fine powder or granules.

2.2.4. Life Cycle Assessment (LCA)

To assess the environmental impacts of each production scenario, the SimaPro version 8.4 software developed by Pre-Consultants was employed as the LCA tool. The LCA methodology followed the technical framework outlined in the ISO 14,000 series, which consists of four phases: goal and scope definition, life cycle inventory analysis, impact assessment, and interpretation [32]. In the context of recycling marine plastic using the solvent extraction method and employing LCA technology, certain assumptions and simplifications were made. These allowed for a focused analysis but should be considered when interpreting the results.

- Homogenous plastic composition: the assumption that marine plastic waste being recycled has a relatively homogenous composition, meaning it consists primarily of a single type of plastic, specifically HDPE (was chosen based on the superior performance).
- (2) Constant plastic properties: assuming that the properties of the marine plastic, such as density, molecular weight, and chemical composition, remain constant throughout the recycling process.
- (3) Ideal process efficiency: assuming ideal process efficiency, where all the marine plastic waste is effectively extracted and recovered without any losses or inefficiencies.
- (4) The impact associated with the transportation, collection, and sorting of the plastic wastes are not included.

Marine plastic waste, in practical terms, encompasses a diverse range of plastic materials. Hence, adding intricacy to recycling processes and exerting distinct environmental impacts. Environmental conditions have the potential to modify the features of marine plastics, hence influencing the effectiveness of recycling processes and the associated environmental consequences. The assumption of constant attributes is employed in order to construct a hypothetical scenario that is idealized in nature. Concurrently, the exclusion of specific impacts serves to streamline the analysis by focusing solely on the fundamental recycling process. In general, these assumptions provide a baseline for evaluating the LCA analysis in the best way possible.

Goal and Scope Definition

The current study aimed to assess the environmental impacts throughout the life cycle of the solvent extraction recycling method applied to HDPE marine waste. The specific objectives are to assess the overall environmental impact of the process and to determine the relative contributions of each stage towards the total impact. For the solvent extraction recycling method, the scope encompasses activities such as pretreatment (washing), dissolution in solvent, reprecipitation, and drying. Therefore, the system boundary (Figure 1) applied in current study is "cradle-to-gate", as the process does not include the eventual use or disposal of the product generated (outcome). The functional unit anticipated in the current study is the production of 1 kg of recycled HDPE using the dissolution–reprecipitation method.



Figure 1. System boundary for the production of 1 kg recycled HDPE.

Life Cycle Inventory (LCI)

Table 1 summarizes the detailed information pertaining the parameters of the processes required to produce 1 kg of recycled HDPE from the solvent extraction recycling method. The established LCI considers all processes in the foreground system, where the primary data correlates to the lab scale of both input and output results that are examined.

| Stage | Phase | Utilities | Values | Unit |
|-----------------|--------|----------------|--------|------|
| Washing | Input | Pipe water | 500 | mL |
| _ | - | Detergent | 1000 | mL |
| | | HDPE | 1 | kg |
| | Output | HDPE | 1 | kġ |
| Dissolution | Input | HDPE | 1 | kg |
| | | Electricity | 3.4 | kWh |
| | | Toluene | 2000 | mL |
| | Output | Dissolved HDPE | 18.3 | kg |
| Reprecipitation | Input | Dissolved HDPE | 18.3 | kg |
| | | Electricity | 0.3 | kŴh |
| | | Acetone | 2000 | mL |
| | Output | Recovered HDPE | 1.2 | kg |
| Drying | Input | Recovered HDPE | 1.2 | kg |
| | | Electricity | 3.88 | kŴh |
| | Output | Recycled HDPE | 1 | kg |

Table 1. Inventory data to produce 1 kg of recycled HDPE from solvent extraction recycling method.

2.2.5. Life Cycle Impact Assessment (LCIA)

In order to comprehensively assess the environmental impacts associated with the production of 1 kg of recycled HDPE, the life cycle impact assessment (LCIA) approach was employed using the ReCiPe 2016 Midpoint (H) V1.06/World (2010) H method [33]. This LCIA method enables the evaluation of a broad range of environmental indicators and quantifies the impacts across 18 different single environmental problems. This method takes into account various impact categories such as climate change, acidification, eutrophication, ozone depletion, human toxicity, freshwater ecotoxicity, and many others.

3. Results and Discussion

3.1. Effects of Temperatures and Time

The findings shown in Table 2 indicate that the HDPE polymer was effectively dissolved at a temperature of 60 °C within a duration of 30 min. The rise in temperature resulted in a significant enhancement of the recovery rate of the dissolved polymer. Significantly, when subjected to a temperature of 75 °C for a length of 20 min, the recovery rate exhibited a notable increase of 95%. Furthermore, by increasing the temperature to 90 °C for the same duration, the recovery rate was further boosted to 98%. The results of this study highlighted the positive impact that elevated temperatures and reduced treatment durations have on the retrieval of polymers in the recycling process. Based on the observed results, it can be inferred that the most favorable circumstances for the dissolution of HDPE in this study were achieved at a temperature of 75 °C for a duration of 20 min. HDPE was reported to have a more amorphous structure with weaker intermolecular forces, which are prone to dissolution at low temperatures. This led to a notable increase in the recovery rate, surpassing 90% with statistical significance.

Table 2. HDPE recovery percentage in relation to time and dissolution temperature.

| | | Recovery (%) | |
|-------------|-------------------|------------------|------------------|
| Time/T (°C) | 60 | 75 | 90 |
| 10 | 73.50 ± 1.96 | 82.55 ± 1.08 | 94.10 ± 2.16 |
| 20 | 82. 85 ± 1.96 | 95.65 ± 1.27 | 98.54 ± 1.10 |
| 30 | 86.15 ± 2.25 | 97.85 ± 1.70 | 100 ± 0 |

Concerning PP, it is evident that achieving the complete dissolution of this material posed difficulties at lower temperatures, as demonstrated in Table 3. The recovery rates for the dissolved PP polymer were significantly low at both temperatures of 60 °C and 75 °C. This observation implies that the dissolution of polypropylene (PP) is not readily accomplished or does not take place through the desired dissolution procedure at these relatively lower temperatures. The presence of both crystalline and amorphous domains in the structure of PP is responsible for this phenomenon [34,35]. This feature exhibits robust intermolecular forces, rendering it very resistant to dissolution, particularly in the presence of toluene, a nonpolar solvent with little capacity to disrupt these intermolecular connections [36]. Moreover, the phenomenon of polymer dissolution encompasses the surmounting of the energy barriers linked to the disruption of intermolecular connections and the migration of polymer chains into the solvent medium [37]. The increased level of crystallinity exhibited by PP presents an additional obstacle, impeding the ingress of solvent molecules into the polymer matrix [38]. As a result, decreased temperatures may exhibit insufficient energy to effectively overcome the intermolecular interactions, hence impeding the total breakdown of polypropylene. It is crucial to recognize that attaining the full dissolution in toluene under the given temperature conditions might be an elusive goal. However, it is important to note that there is still a possibility for PP to experience swelling or partial breakdown. This possibility is dependent on the length of exposure, which is in tandem with the findings presented in Table 4. In comparison to the conditions of lower temperature, the data shown in Table 5 demonstrates that the dissolution of PP

at 90 °C resulted in a significantly higher rate of recovery. Nevertheless, in order to attain a recovery rate over 90% under these specific thermal conditions, it was imperative to engage in an extended processing period of 30 min, which stands in stark contrast to the comparatively shorter durations observed at lower temperatures. Although the extended duration required for processing may be considered a disadvantage in terms of operational efficiency, the significant rate of recovery indicates that a temperature of 90 °C is effective for dissolving PP in the context of this investigation. The increased time length facilitates a thorough dissolving process, resulting in the significant recovery of the PP polymer.

| | | Recovery (%) | |
|-------------|----------------|------------------|------------------|
| Time/T (°C) | 60 | 75 | 90 |
| 10 | 18.77 ± 0.46 | 47.60 ± 1.18 | 84.80 ± 1.18 |
| 20 | 22.35 ± 0.69 | 50.92 ± 1.26 | 88.55 ± 1.47 |
| 30 | 23.25 ± 0.88 | 64.67 ± 0.14 | 95.10 ± 0.20 |

 Table 3. PP recovery percentage in relation to time and dissolution temperature.

3.2. Optimum Conditions on Marine Plastic Waste Samples

The marine HDPE polymer was seen to showcase a remarkable recovery percentage of 96.67% \pm 3.30. This outcome underscores the successful retrieval of the material, reaffirming the recycling process's viability. In contrast, the recovery percentage for PP stands slightly below the 90% threshold, specifically at $87.35\% \pm 3.2$. Despite this marginally lower figure, it remains a commendable achievement, particularly within the recycling context. Comparatively, the observed dip in recovery rate, when contrasted with the results from using pristine PP samples, can be attributed to an interplay of various influencing factors, including contamination and heterogeneity. In the case of the marine waste PP, the presence of a multitude of pollutants and impurities, including salt, organic matter, and microorganisms, is likely. This complex matrix of contaminants poses a challenge to the recycling process, potentially affecting the efficacy of recovery [39]. Furthermore, the inherent heterogeneity of marine waste PP compounds the intricacy of achieving uniform recovery rates. This heterogeneity arises from the amalgamation of diverse PP materials with distinct types and grades originating from diverse sources [40]. Consequently, the dissolution and reprecipitation process interacts with this varied composition in intricate ways, influenced by factors encompassing polymer composition, molecular weight, additive composition, and the material's unique processing history. The culmination of these multifaceted challenges contributes to the observed lower overall recovery percentages in the recycling of marine waste PP.

3.3. IR analysis of Marine Plastic Waste Samples

Figure 2 revealed that the spectrum of the plastic waste sample exhibited similarities to the standard PP spectrum. The primary distinction in structure between PE and PP lies in the absence of methyl groups in PE, specifically in the HDPE variant. In contrast, PP exhibits CH_3 stretching peaks at 2950 and 2871 cm⁻¹, along with a methyl group umbrella mode at 1376.01 cm⁻¹ (Table 4). The presence of four peaks in this range, corresponding to both the methyl and methylene groups, serves as a further confirmation of the polymer identification [41]. The observed variations in the spectra at certain wavenumbers were ascribed to the presence of additives in small amounts within the waste material. Moreover, it can also be observed that the IR spectrum of the recycled marine plastic exhibited a resemblance to that of the virgin PP. This observation indicates that the recycled marine plastic undergoes the dissolution–reprecipitation process without undergoing any notable structural modifications. In contrast, it can be seen that the marine HDPE, as depicted in Figure 3, have slowly started to degrade or oxidize, as there are several uncommon functional groups for HDPE that can be found in the spectra. For example, the presence of a hydroxide (OH) group at 3696.91 cm⁻¹ and ether (C-O-C) at around 1103 to

1265 cm⁻¹ (Table 5). Overall, as can be seen, all characteristic bands in each polymer type did not change significantly after the recycling process. The differences observed in the peak heights could potentially be attributed to slight variations in the sample weights. Tables 4 and 5 show the absorption bands of marine PP and HDPE, before and after recycling, along with the virgin HDPE for the purpose of comparison.



Figure 2. The IR spectrum of marine PP.



Figure 3. The IR spectrum of marine PE.

Table 4. Absorption bands of marine PP.

| Functional Group – | Absorption (cm ⁻¹) | | | |
|---|--------------------------------|---------|---------|--|
| Functional Group | Before | After | Virgin | |
| | 2951.63 (C-CH ₃) | 2951.27 | 2951.45 | |
| C H stratch | 2917.50 (C-CH ₂) | 2918.12 | 2917.73 | |
| C-H stretch | 2838.52 (C-CH ₂) | 2839.29 | 2871.19 | |
| | 2871.28 (C-CH ₃) | 2869.60 | 2838.27 | |
| CH ₂ bend | 1455.84 | 1455.58 | 1456.40 | |
| CH ₃ (umbrella mode) | 1376.01 | 1376.07 | 1376.22 | |
| CH bend, CH ₃ rock, C-C stretch | 1166.90 | 1166.88 | 1166.65 | |
| CH_3 rock, CH_3 bend, CH bend | 998.03 | 997.75 | 998.04 | |
| CH ₃ rock, C-C stretch | 973.23 | 972.96 | 973.47 | |
| CH ₂ rock, C-CH ₃ stretch | 841.06 | 840.99 | 841.41 | |
| CH ₂ rock, C-C stretch, C-CH stretch | 808.78 | 808.82 | 808.75 | |

| A | bsorption (cm ⁻¹) | | |
|----------------------|---|---------|---------------|
| Functional Group | Before | After | Virgin |
| C-H stretch | 2915.54 | 2916.36 | 2914.48 |
| C-H stretch | 2847.69 | 2848.64 | 2847.70 |
| CH_2 bend | 1462.66 | 1463.01 | 1462.41 |
| CH ₂ rock | 729.58 | 729.20 | 730.19 |
| CH2 rock | 718.83 | 718.33 | 718.86 |
| Degradation compound | | | |
| Functional group | Absorption (cm^{-1}) | | General range |
| OH | 3696.91 | | 3400-3700 |
| C-O-C | (1) 1103.13(2) 1032.18 | | 1030–1300 |

Table 5. Absorption bands of marine HDPE.

The appearance of a hydroxide group (OH), which is the most common functional group present in degraded waste plastic, is known to be due to the formation of monomeric hydroperoxides and hydrogen-bonded alcohols and hydroperoxide [42]. Generally, a process known as degradation can lead to the partial or full disintegration of a polymer under certain environmental conditions, including heat, water (moisture), light, chemical conditions, and bacteria [43]. The combined effects of ambient oxygen, sunlight, and seawater are thought to be responsible for the degradation of the vast majority of conventional polymers that may be found in marine environments [44]. Plastics degrade in the marine environment in five ways: hydrolytic degradation, thermo-oxidative degradation, photodegradation, biodegradation, as well as mechanical degradation [45]. Photooxidation is thought to be the utmost critical process in the degradation of decomposition of plastic waste, following mechanical activity as well as heat oxidation [44].

Moreover, according to Iniguez et al. (2018) [44], marine plastic waste degrades mostly by photodegradation in the environment and is significantly influenced by the ultraviolet (UV) spectrum (around 400–100 nm) of sunlight. The author has further corroborated that the photooxidative degradation of polymers exposed to the marine environment, including PP, PE, and nylon, is initiated by UV-B light (280–390 nm) from the sun and begins at the plastics' outer surface. In addition to that, as a consequence of chemical weathering, some changes in the polymer can be seen to take place in the course of the process; molecular weight reduction occurs along with the formation of oxygen-rich functional groups, bond scission, as well as chemical transformation. These statements expound on the presence of several oxygen-rich functional groups observed in the IR spectrum of marine HDPE. To further understand the degradation of the polymer, Figure 4 below indicates the oxidation of PE's general mechanism.

In the traditional paradigm, carbon-centered macroradicals (P1) rapidly react with oxygen to form hydroperoxyl radicals (P2), which subsequently take an H atom from the polymer chain to generate a hydroperoxide (P3) and a new macroradical (R1). Hydroperoxides undergo rapid photolysis, resulting in the generation of the alkoxyl macroradical radical (P4) and the hydroxyl radical (O1). P4 is the key intermediate in the reaction. It can undergo B-scission with chain cleavage to produce an aldehyde (P6), and hydrogen abstraction without chain cleavage to produce hydroxyls (P8). The last reaction yields ketones (P5) and undergoes a Norrish reaction (photochemical reaction) that generates unsaturated vinyl-type and a chain-end ketones (P7) [46].





Figure 4. General mechanism for PE degradation.

3.4. Melting Temperature

An essential thermodynamic characteristic that offers key insights into the thermal behavior of polymers is the melting temperature (T_m). For HDPE, the virgin polymer exhibited a melting temperature of 128.48 °C. When subjected to the recycling process, specifically in its waste form, the HDPE polymer displayed a slightly elevated melting temperature of 129.04 °C. Notably, upon the completion of the recycling procedure, the recycled HDPE exhibited a nuanced reduction in its melting temperature, settling at 126 °C. Similarly, the PP polymer demonstrated distinct melting temperature trends across its various states. The virgin PP material presented a characteristic melting temperature of 163.88 °C. As for the waste PP post-recycling, the melting temperature slightly increased to 165.12 °C. The subsequent recycling efforts culminated in the recycled PP polymer, which exhibited a refined melting temperature of 164.35 °C.

This shows that the melting points of the polymers are not significantly affected by the recycling process, as seen by their reasonably stable nature falling within an acceptable range when compared to their virgin counterparts. However, minute variations in the melting temperature and the range between the trash and recycled grades can be attributed to the plasticization effects and the presence of additives that are inherent to the polymer composition [47]. The polymer goes through several transformational steps during recycling, including reprecipitation and dissolution. The molecular structure of the polymer may change during these phases. When polymer chains are subjected to high temperatures and mechanical stress during these stages, a temporary softening or alteration of the polymer's physical properties results. This phenomenon is known as plasticization [48]. In light of

the properties of the original waste-grade polymer, this can lead to minor variations in the melting temperature as well as the melting range. Furthermore, these effects might be altered by any lingering solvent traces that might stay in the polymer's structure after recycling [49].

The endothermic peak in the context of a DSC analysis (Figures 5 and 6) represents the material's absorption of heat energy during the transformative phase change from a solid to a liquid state. The distinct endothermic peaks in the cases of HDPE and PP, which appear at temperatures of 128 °C and 164 °C, respectively, provide clear indicators of the specific temperatures at which these polymers undergo the transformation from solid to liquid. Additionally, the endothermic peak's shape offers an insight into the melting process. A broader peak indicates a range of temperatures over which melting occurs, reflecting the distribution of molecular interactions and the presence of impurities or additives. On the other hand, the well-defined and highly peaked curves provided an alternative point of view. This particular arrangement suggests a more synchronized and uniform melting process, in which the entire material experiences the phase transition over a smaller temperature range.



Figure 5. Melting curve of HDPE.



Figure 6. Melting curve of PP.

3.5. Life Cycle Assessment (LCA) Impact Analysis

The life cycle assessment performed in this study includes the quantitative effects of each stage of the recycling process. The production process of recycled HDPE entails a sequential progression of steps, namely washing (sometimes referred to as pretreatment), dissolving, reprecipitation, and drying. The chosen life cycle impact assessment methodology facilitates the assessment of diverse environmental indicators and the quantification of impacts across a total of 18 distinct environmental categories. This study, however, specifically examines the environmental repercussions that have substantial significance. The aforementioned factors encompass global warming, freshwater eutrophication, freshwater

ecotoxicity, marine ecotoxicity, human carcinogenic toxicity, fossil resource shortage, and water consumption. The study seeks to prioritize and assess the environmental repercussions that hold significant implications within the recycling process by focusing on these specific aspects.

In comparison to washing and drying, the dissolution and reprecipitation phases of the four processes analyzed had the most significant environmental impact. The data presented in Figure 7 for the dissolution and reprecipitation stages support this conclusion. The environmental impact of these two stages was primarily observed in a number of impact categories, with human carcinogenic toxicity having the greatest effect, followed by freshwater ecotoxicity, marine ecotoxicity, and freshwater eutrophication. These findings suggest that the dissolution and reprecipitation phases are particularly hazardous to human health and aquatic ecosystems. During the dissolution and reprecipitation phases, chemicals such as toluene and acetone were likely the primary cause of this environmental impact. These chemicals may contribute to the observed toxicity and ecological disruptions, resulting in negative effects on human and aquatic health.



Figure 7. Impact percentage of overall solvent extraction process.

Due to the potential discharge of these substances into the environment and their subsequent ingestion or inhalation by humans, the use of chemicals such as toluene during the recycling process can contribute to human carcinogenic toxicity. This release may be caused by the emission of air contaminants, such as volatile organic compounds (VOCs), which are typically found in solvents [50]. VOCs emitted into the air during the recycling operations can undergo various atmospheric reactions and dispersion, eventually leading to their presence in ambient air. These compounds can be transported over long distances and subsequently deposited onto land or water surfaces. In some cases, they can also contaminate food sources and drinking water supplies [51]. Furthermore, the excessive consumption or release of chemicals can have detrimental effects on freshwater ecosystems, leading to eco-toxicity, marine ecotoxicity, and eutrophication. When these chemicals are discharged into the environment, they can be absorbed by aquatic organisms, amplifying the ecological impacts [52]. Freshwater eco-toxicity encompasses the harmful effects on fish, invertebrates, and plants in freshwater ecosystems, potentially leading to reduced biodiversity and imbalances within the ecosystem. This eco-toxicity can also contribute to eutrophication, which manifests as harmful algal blooms, oxygen depletion, and reduced water quality. Marine ecotoxicity, on the other hand, occurs when chemical pollutants from freshwater sources impact marine organisms, disrupting their health and the overall functioning of marine ecosystems.

Additionally, the utilization of electricity during dissolution, reprecipitation as well as drying can contribute to human carcinogenic toxicity due to the potential exposure to hazardous chemicals that are often released into the environment during energy production. Moreover, the combustion of fossil fuels for electricity generation significantly amplifies the emission of air pollutants, including nitrogen oxides (NO_x) and sulfur oxides (SO_x), into the atmosphere [53]. These pollutants contribute to the formation of smog and acid rain, leading to a range of environmental and human health issues. Additionally, the combination of pollutants deposited on land and the nutrient loading in aquatic systems caused by electricity generation contributes to soil and water contamination, as well as the disruption of ecological balance in freshwater ecosystems. In water systems, electricity generation can contribute to freshwater ecotoxicity and eutrophication [54]. Wastewater discharge from energy production introduces excessive nutrients, leading to algal blooms and oxygen depletion leading to adverse impacts on fish, invertebrates, and other aquatic organisms. The findings that the overall environmental impact of the recycling process was relatively low across all observed categories indicates that the recycling process has successfully mitigated the potential negative effects on human health and ecosystems.

3.6. Limitations of Current Study

One notable constraint in this study is the lack of extensive mechanical property analyses conducted on recycled marine plastics. Although the utilization of techniques such as FTIR for material characterization and DSC for measuring the melting point (T_m) offer valuable insights into the composition and thermal behavior of the materials, it is important to recognize the absence of data pertaining to essential mechanical properties. The parameters of tensile strength, flexural strength, impact resistance, and elongation at break have not yet been thoroughly investigated. The constraint hinders the capacity to comprehensively evaluate the appropriateness of the recycled plastics for different uses, as the mechanical characteristics are crucial factors in defining the material's structural integrity and long-lasting nature.

Secondly, the lack of a comprehensive analysis of the thermal properties of recycled marine plastics is an additional significant limitation of this study. This study does not possess data on essential thermal properties, such as crystallinity and glass transition temperature (Tg). These parameters are necessary for comprehending the thermal stability, processability, and dimensional stability of a material, especially in the context of thermal applications. The lack of such information hinders the ability to evaluate the material's suitability for specific thermal applications and may lead to incomplete understanding of its processing behavior.

Another limitation of this study is its narrow emphasis on neat polymers, without any investigation into the realm of blended plastic waste. The characterizations of the study have offered valuable insights into the properties of neat polymer materials. Nevertheless, the narrow focus of this study limits the generalizability of the results to real-world scenarios, in which there is a significant presence of diverse plastic waste. Marine plastic pollution and recycling processes often involve coming across mixed plastics, which is a common occurrence. Therefore, it is crucial to comprehend the behavior and properties of these mixed plastics. The lack of analysis regarding mixed plastic waste hinders our ability to gain insights into the challenges and opportunities related to recycling these intricate material compositions in the real world.

In the realm of chemical and manufacturing procedures, the E-factor, commonly known as the waste factor, assumes a pivotal role as a fundamental metric for evaluating efficacy and environmental responsibility. The quantification of waste generated in relation to the targeted output provides useful insights into the environmental effect of a specific operation. In general, a larger E-factor is indicative of increased inefficiency and a less sustainable operation, as it represents a significant proportion of waste relative to the product. In certain cases, a notable benefit is shown when the E-factor exhibits proximity towards a zero value. This circumstance signifies that the process produces minimal to no waste. The

quantification of even minimal waste resulting from various reasons such as contaminants, equipment losses, or negligible byproducts becomes exceedingly challenging. Nevertheless, it is imperative to acknowledge that in practice, it is impossible for any chemical process to obtain an E-factor of absolute zero. The current study suggests an unparalleled degree of effectiveness and environmental sustainability. The remarkably low E-factor due to negligible waste generated from this study possessed both advantageous and limiting aspects, the latter of which was held in high regard. It highlighted the process's exceptional environmental conscientiousness, but it could also signify a challenge in terms of analyzing and optimizing a process that produces such minuscule amounts of waste. However, this noteworthy accomplishment represents a significant advancement in the promotion of environmentally friendly and sustainable chemical and manufacturing methods.

4. Conclusions

This study focuses on assessing the feasibility of using the solvent extraction method, specifically dissolution-reprecipitation technique for recycling marine plastic waste. The empirical findings of this study concluded that the dissolution temperature exerts significant impact in yielding high recovery rates during the recycling process. The results also suggested that polyethylene (PE) is efficiently dissolved at comparatively low temperatures, whereas polypropylene (PP) has a limited solubility under similar circumstances. The optimal conditions for dissolution were determined as 75 °C for HDPE and 90 °C for PP, with corresponding time intervals of 20 min and 30 min, respectively. Applying these optimal conditions to real marine plastic waste samples resulted in high recovery percentages of 96.67% for HDPE and 87.35% for PP. The application of these ideal conditions to actual marine plastic waste samples yielded recovery rates of 96.67% for HDPE and 87.35% for PP. Moreover, the recycled marine plastics displayed no significant changes in their chemical composition and showed similarity to virgin plastics, suggesting an advantageous conclusion. The melting temperature (T_m) of the recycled plastics was observed to be within the expected range and exhibited similarity to that of the virgin plastics. This finding implies that the recycled polymers possess the potential to serve as a viable alternative to virgin plastics, as their performance and processing properties remain uncompromised. From the life cycle assessment (LCA) of the optimized plastic recovery, i.e., the HDPE, the drying phase contributed the most significant environmental impact specifically linked to impact categories such as human carcinogenic toxicity, freshwater ecotoxicity, marine ecotoxicity, and freshwater eutrophication. However, the overall environmental impact of the recycling process was found to be relatively low across all observed categories which can be considered an environmentally favorable approach for recycling marine plastic waste. Based on the findings, it can be concluded that the dissolution–reprecipitation technique holds promise as a viable approach for recycling marine plastic waste. The high recovery percentages, minimal changes in chemical structure, and low environmental impact support the potential effectiveness and sustainability of this recycling method. Further research and development in this domain could potentially enhance the process and facilitate the more effective and extensive recycling of marine plastic waste.

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