



UPCYCLING OF LOW-DENSITY POLYETHYLENE

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ABSTRACT

The global plastic waste crisis is intensifying, with low-density polyethylene (LDPE) posing significant recycling challenges due to its chemical inertness, contamination susceptibility, and poor compatibility with conventional mechanical methods. Existing chemical recycling techniques such as pyrolysis and solvolysis, while promising, often suffer from high energy requirements, use of hazardous solvents, and non-selective product yields. This study proposes a hybrid chemo catalytic upcycling approach for LDPE that integrates solvent-assisted purification with low-temperature catalytic hydro-thermolysis using ZSM-5 zeolite. LDPE sourced from post-consumer zip-lock bags underwent a two-step treatment involving dissolution–precipitation followed by depolymerization in an acetone–water medium. The methodology emphasizes energy efficiency, solvent-free reaction environments, and structural preservation of polymer chains. Structural and chemical analyses via XRD and FTIR confirmed the retention of semi-crystalline morphology and minimal oxidative degradation in optimized batches. Batch 4 achieved the highest recovery (88.6%) and lowest conversion (11.83%), highlighting the process's effectiveness in preserving polymer integrity. The results demonstrate the viability of this technique as a scalable, sustainable alternative for LDPE upcycling aligned with circular economy goals.

Keywords:

Catalytic Hydro-thermolysis, Characterization, Dissolution–Precipitation, LDPE Recycling, ZSM-5 Catalyst

I. INTRODUCTION

Plastic pollution has emerged as one of the most pressing environmental challenges of the 21st century, with global plastic production exceeding 400 million tons annually, yet only a small fraction is effectively recycled [1]. Among the various types, low-density polyethylene (LDPE) remains particularly difficult to recycle due to its flexibility, contamination issues, and chemical inertness [2], [3]. Traditional landfilling and incineration not only squander resources but also pose serious ecological and human health hazards [4], [5].

Mechanical recycling, though widely adopted, suffers from limitations such as polymer degradation, incompatibility of mixed plastics, and the generation of low value recyclates [15].

In contrast, chemical recycling methods such as pyrolysis, catalytic cracking, and solvolysis are gaining momentum due to their potential to recover monomers or produce valuable chemicals and fuels [4], [6], [13], [14]. Dissolution–reprecipitation methods have demonstrated over 90% recovery efficiency for LDPE, HDPE, and PP using optimized solvent/non-solvent systems, offering a means



to maintain polymer integrity [11]. However, the energy intensity and solvent recovery challenges remain significant. Complementary to this, studies on polymer swelling and solubility show that LDPE exhibits higher swelling and solubility compared to HDPE in lighter alkanes, influencing its behavior during thermal or solvent-based treatments [12]. Solvolysis, particularly targeting PET, has seen advances using solvents like ethylene glycol and catalysts such as metal salts, DESs, and ionic liquids [17], [18], [19], [13]. However, these methods are often tailored to condensation polymers and may not be directly applicable to polyolefins like LDPE. Several studies explored the use of deep eutectic solvents for depolymerizing recalcitrant plastics by enhancing solubility and catalytic activity while aligning with green chemistry principles [13]. Pyrolysis remains a cornerstone of chemical recycling, converting LDPE into aliphatic hydrocarbons—alkanes and alkenes—that can re-enter the petrochemical supply chain [4], [14], [21]. The use of FCC catalysts and fixed-bed reactors improves conversion efficiency, though issues like coke formation and high-temperature requirements persist [11], [21]. Notably, recent innovations have shown promise with radio-frequency induction heating combined with dual-functional catalysts ($\text{Fe}_3\text{O}_4/\text{Ni}$, Pt), achieving selective depolymerization to light olefins or C7–C14 alkanes with minimal byproduct formation [21]. Low temperature depolymerization techniques are gaining attention. For example, catalytic hydro-thermolysis of LLDPE using H-ZSM-5 and trace water achieved complete selectivity to C1–C4 products at just 220 °C, driven by Brønsted acid site activity [22]. Similarly, solvent-free hybrid chemo-mechanical approaches using reactive twin-screw extrusion (TSE) allow structural tuning of LDPE through catalytic degradation during extrusion [23]. This method shows potential for producing coatings and adhesives from contaminated post-consumer LDPE.

LDPE's mechanical behavior under stress, temperature, and strain rate has been well-characterized to inform its recyclability and product design, especially in high-performance applications [24]. Beyond recycling, novel valorization strategies now include the upcycling of LDPE into highly crystalline artificial graphite with exceptional electrical conductivity and carbon yield [25], expanding its utility into advanced electronics and energy applications. In parallel, various studies have focused on the recovery of monomers via depolymerization (e.g., PET to BHET or TPA) under mild conditions [7], [8], [9], [10]. These studies also highlighted the role of green solvents, ionic liquids, and solid catalysts in improving selectivity and reducing energy consumption. Process modeling and optimization have been applied to catalytic systems, pyrolysis kinetics, and reactor simulations to understand degradation mechanisms and maximize product quality [5], [16], [20].

Despite significant advancements, existing methods for LDPE recycling are either energy-intensive, generate non-specific product mixtures, or require costly solvent systems. Most catalytic methods still demand high temperatures or pressures, and solvent-based techniques face scalability issues due to solvent recovery and contamination. To bridge these gaps, this project proposes a novel low-temperature, hybrid chemocatalytic method for LDPE recycling, integrating localized heating via embedded magnetic materials, solid acid zeolite catalysts, and a solvent-free reactive processing route. Unlike conventional pyrolysis or solvolysis, this approach allows selective depolymerization at lower temperatures, minimal coke formation, and targeted production of short-chain hydrocarbons or oxygenates, which can be reused directly in polymer synthesis or as fuel precursors. By eliminating harmful solvents and reducing energy input, this method enhances both environmental sustainability and economic viability. It represents a significant advancement toward scalable, high-efficiency recycling of LDPE waste in alignment with circular economy principles

II. METHODOLOGY

2.1 Materials and Apparatus

Low-Density Polyethylene (LDPE) zip-lock plastic bags were chosen as the polymeric waste feedstock for catalytic depolymerization, with analytical-grade toluene and *n*-hexane serving as organic solvents for the dissolution–precipitation process. ZSM-5, a microporous zeolite catalyst with strong Brønsted acidity, was employed to facilitate catalytic cracking reactions. All experiments utilized standard

laboratory-grade glassware and apparatus, including a three-neck round-bottom flask fitted with a spiral-coiled reflux condenser, magnetic stirrer, and thermometer for controlled batch reactions. Vacuum filtration was conducted using a Büchner funnel with a connected vacuum pump, while the catalytic hydro-thermolysis step was carried out in a stainless-steel autoclave lined with a Teflon insert to ensure safe, chemically compatible operation under elevated temperature and pressure.

2.2 Pre-Treatment of Waste LDPE

Waste LDPE was sourced from used zip-lock plastic bags and manually cut into 1 cm × 1 cm squares. To remove surface contaminants, the cut plastic pieces were soaked in distilled water for a few minutes, then drained and oven-dried at 75–80 °C for 30 minutes. Dried pieces were manually separated to avoid agglomeration and ensure uniform exposure during further processing.



Fig 1. LDPE Squares Cut from Waste Zip Lock Plastic Bags



Fig 2. Cleaning Using Distilled Water and Filtration of LDPE Squares

2.3 Solvent Assisted Dissolution and Precipitation of LDPE

A weighed sample of 0.5 g dried LDPE was added to 12.5 mL of toluene in a round-bottom flask and stirred magnetically. The mixture was heated in an oil bath maintained at 80 °C for 30–40 minutes to achieve complete polymer dissolution. After cooling to room temperature, 7.5 mL of *n*-hexane was introduced to induce phase separation and precipitate the polymer, forming a colloidal suspension. The mixture was left undisturbed for 15–20 minutes, then filtered under vacuum using a Büchner funnel. The recovered LDPE solid was dried and mechanically broken into smaller pellets. This solvent-mediated recycling step improves purity, enhances surface area, and prepares the polymer for efficient catalytic breakdown. Toluene was selected for its non-polar compatibility with LDPE, while *n*-hexane aids in antisolvent-induced precipitation due to its lower polarity.



Fig 3. Dissolution and Precipitation of LDPE using Toluene and n-Hexane with Vacuum Filtration



Fig 4. Vacuum Filtration and LDPE powder after drying in Oven at 80°C

2.4 Catalytic Hydro-thermolysis of LDPE

The LDPE pellets obtained in Step 1 were added to an acetone–water mixture (total 4 mL, with 3.2 mL water and 0.788 mL acetone) along with 0.04 g ZSM-5 catalyst (8 wt.% relative to LDPE mass). The mixture was stirred to ensure uniform dispersion of both catalyst and polymer. This reaction mixture was transferred into a Teflon-lined stainless-steel autoclave and sealed. The reactor was heated to 70 °C and maintained for a specified period (2–3 hours) to allow depolymerization under mild hydrothermal conditions. The choice of low temperature enhances energy efficiency while minimizing degradation byproducts. Upon completion, the system was cooled to room temperature. To remove volatile and gaseous products, 20 mL of distilled water was added, and nitrogen purging was performed. The vessel was sealed with a septum cork, and gas was flushed using an inlet and outlet needle, creating an inert atmosphere during off-gassing. Acetone improves catalyst dispersion and swelling of LDPE, while the ZSM-5 catalyst provides acidic sites for bond scission. The Teflon liner ensures containment of volatiles and prevents metal contamination.



Fig 4. LDPE mixture before putting in Teflon cover and autoclave vessel

2.5 Recovery and Evaluation

After purging, the contents were filtered and dried. The conversion and recovery metrics were calculated. In contrast to PET depolymerization, where conversion implies useful monomer recovery, LDPE recycling emphasizes material recovery. Therefore, lower conversion values can indicate greater preservation of polymer mass for reuse and are sometimes preferable depending on the recycling objective.

LDPE Recovery (%)

Measures the mass of unreacted LDPE successfully retained after processing:

$$\text{Recovery (\%)} = \frac{\text{Recovered LDPE (g)}}{\text{Feed LDPE (g)}} \times 100 \dots\dots\dots (1)$$

LDPE Conversion (%)

Quantifies the fraction of LDPE lost due to reaction, degradation, or incomplete recovery:

$$\text{Conversion (\%)} = \frac{\text{Feed LDPE} - \text{Recovered LDPE}}{\text{Feed LDPE}} \times 100 \dots\dots\dots (2)$$

III. RESULTS AND DISCUSSION

3.1 XRD Analysis

1. Virgin LDPE

The XRD pattern of virgin low-density polyethylene (LDPE) confirms its semi-crystalline nature, characterized by distinct diffraction peaks superimposed on a broad amorphous background. A prominent peak observed at $2\theta \approx 21.5^\circ$ corresponds to the (110) crystalline plane, which is indicative of the orthorhombic unit cell structure typically found in polyethylene. Additional, less intense peaks at 2θ values of approximately 18.1° , 23.8° , and 29.5° suggest the presence of secondary reflections arising from other crystal lattice orientations, such as the (200) and (020) planes.

The intensity and sharpness of the peak at 21.5° highlight the partial crystalline ordering of polymer chains, while the broad baseline spanning lower and higher 2θ values reflects the amorphous fraction of LDPE, attributed to the disordered chain segments and branching typical of this polymer. This dual-phase composition—crystalline and amorphous—is responsible for LDPE's characteristic mechanical flexibility and thermal behavior. Minor peaks at $2\theta \approx 13.24^\circ$, 14.2° , and 28.8° may arise due to local ordering or structural imperfections, and their presence further confirms the heterogeneous nature of

the polymer morphology. The crystallinity index (CI) can be estimated from the ratio of the crystalline peak area to the total area under the curve to quantitatively analyse the extent of crystallinity.

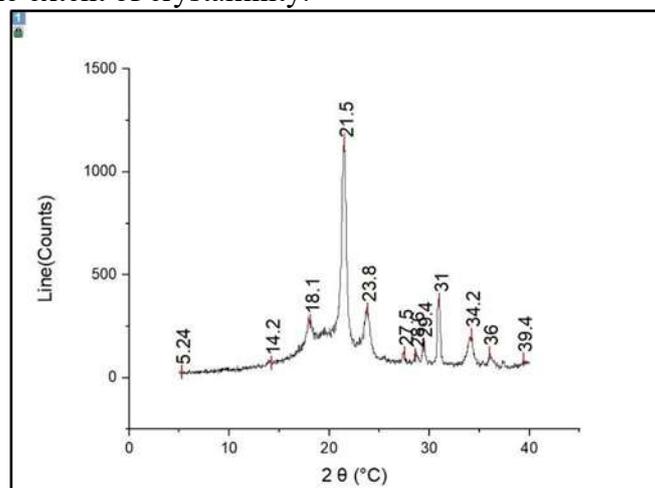


Fig 5. XRD Analysis of Virgin LDPE

2. Batch 1 recycled LDPE (step 2, RLDPE)

Batch 1 shows a main peak at $2\theta \approx 21.8^\circ$ like virgin LDPE, but with additional peaks at 16.4° , 18.5° , and 24.2° , along with a broadened baseline. These changes suggest a reduction in crystallinity and increased amorphous content, likely due to structural alterations from thermal and oxidative degradation during recycling. Batch 1 (Recycled) shows signs of significant degradation. While the primary peak near 21.8° is still present, new peaks and a broadened baseline reflect increased amorphous content and disruption in chain alignment, likely due to thermal or oxidative effects during recycling.

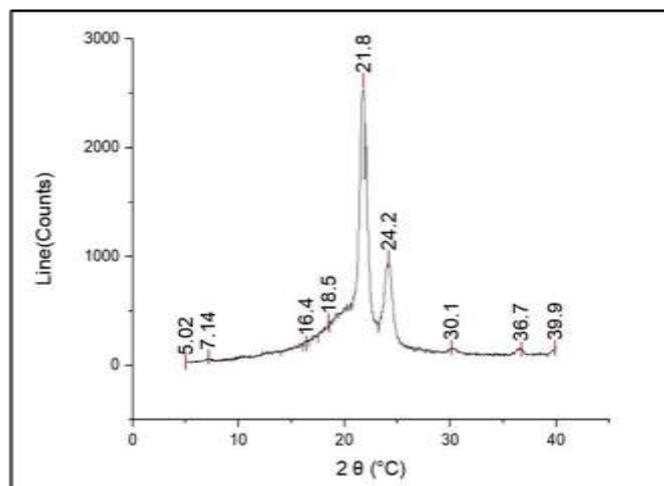


Fig 6. XRD Analysis of B1 ,Step 2 ,R LDPE

3. Batch 2 recycled LDPE (step 2)

The XRD profile of Batch 2 displays dominant peaks at $2\theta \approx 21.8^\circ$ and 24° , corresponding to (110) and (200) planes, respectively. Compared to Batch 1, the peaks are slightly sharper, indicating partial recovery of crystalline regions. Minor peaks at lower angles and elevated baseline intensity still point to the presence of some disorder in the polymer matrix.

Batch 2 exhibits a partial recovery of crystallinity, with more defined peaks at 21.8° and 24° , suggesting improved ordering over Batch 1 but still showing signs of disorder and reduced crystallinity.

compared to virgin LDPE.

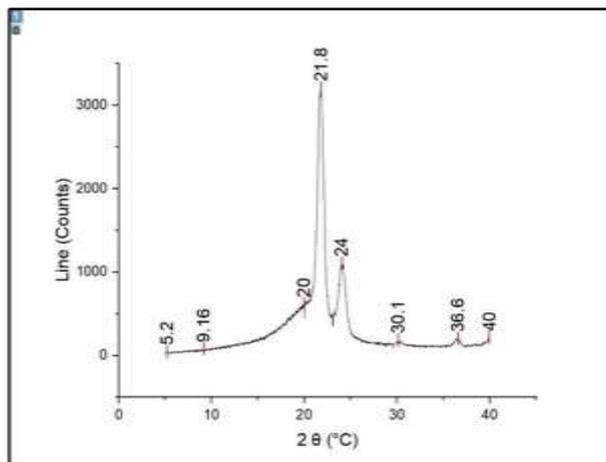


Fig 7. XRD Analysis of B2 ,Step 2 ,R LDPE

4. Batch 4 recycled LDPE (step 2)

The XRD pattern of Batch 4 displays sharp and intense peaks at $2\theta \approx 21.64^\circ$ and 24.02° , which are characteristic of the (110) and (200) planes of LDPE's orthorhombic crystal structure. These prominent reflections indicate a high degree of preserved crystallinity, signifying minimal degradation during the recycling process. Additionally, the presence of distinct secondary peaks at both lower and higher 2θ angles suggests improved polymer chain alignment and partial structural recovery compared to other recycled batches. The narrow peak widths and elevated intensities demonstrate that Batch 4 has retained a crystalline morphology closely resembling that of virgin LDPE. This implies that the recycling conditions for Batch 4 were optimal in preserving the semi-crystalline framework, making it a strong candidate for reuse in high-performance applications.

Batch 4 stands out as the most structurally preserved recycled sample, closely matching the crystallinity profile of virgin LDPE. The sharp, intense peaks at 21.64° and 24.02° , along with reduced baseline noise and minimal broadening, indicate effective preservation of the semi-crystalline structure. This suggests that the recycling conditions applied in Batch 4 were optimal and minimized polymer degradation.

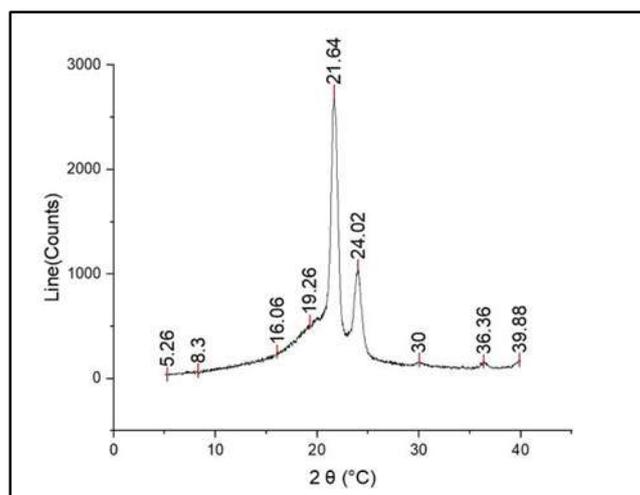


Fig 8. XRD Analysis of B4 ,Step 2 ,R LDPE

3.2 FTIR Analysis

1. Virgin LDPE

The FTIR spectrum of virgin LDPE displays characteristic absorption bands at $\sim 2915\text{ cm}^{-1}$ and $\sim 2849\text{ cm}^{-1}$ corresponding to C–H asymmetric and symmetric stretching vibrations of $-\text{CH}_2-$ groups. A peak around $\sim 1470\text{ cm}^{-1}$ is attributed to bending vibrations of $-\text{CH}_2-$ groups, while a band near $\sim 720\text{ cm}^{-1}$ indicates rocking motion of long $-\text{CH}_2-$ chains. The absence of peaks in the $1700\text{--}1800\text{ cm}^{-1}$ region confirms the lack of carbonyl groups, indicating no oxidative degradation.

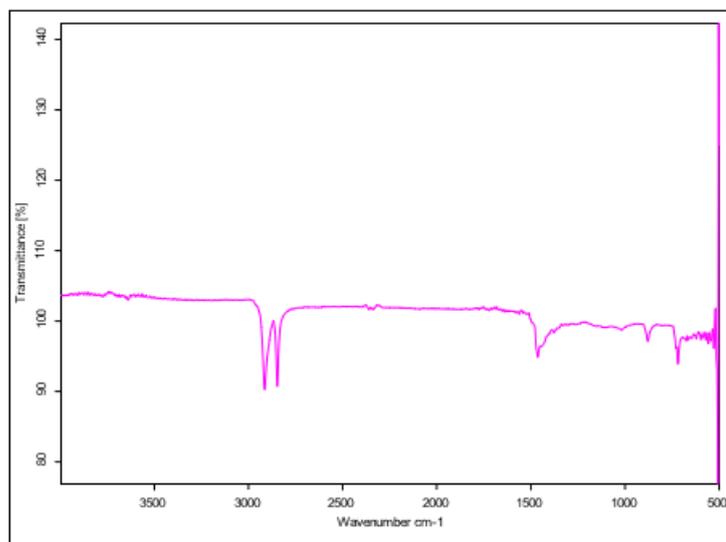


Fig 9. FTIR Analysis of Virgin LDPE

2. Ziplock Bag

The FTIR spectrum of the ziplock bag shows prominent absorption peaks near $\sim 2915\text{ cm}^{-1}$ and $\sim 2848\text{ cm}^{-1}$, characteristic of $-\text{CH}_2-$ asymmetric and symmetric stretching in polyethylene. A bending vibration at $\sim 1470\text{ cm}^{-1}$ and a rocking vibration around $\sim 720\text{ cm}^{-1}$ confirm the presence of long $-\text{CH}_2-$ chains. The absence of significant peaks around $\sim 1700\text{ cm}^{-1}$ suggests minimal oxidative degradation, indicating the material retains typical LDPE structural features suitable for upcycling.

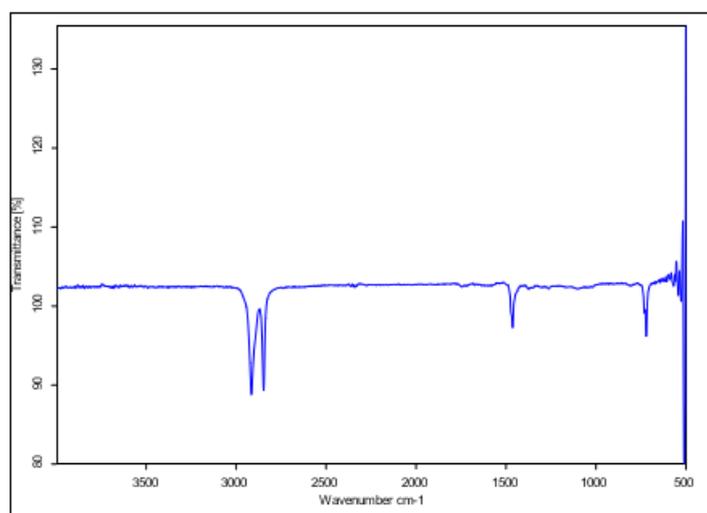


Fig 10. FTIR Analysis of Ziplock Bag

3. Batch 1 Recycled LDPE (step 1)

The FTIR spectrum post-dissolution shows retention of key LDPE functional groups, indicating that the polymer backbone remains largely intact. The characteristic $-\text{CH}_2-$ stretching vibrations at $\sim 2915\text{ cm}^{-1}$ (asymmetric) and $\sim 2848\text{ cm}^{-1}$ (symmetric) are still prominent. Additionally, bending $\sim 1470\text{ cm}^{-1}$

and rocking $\sim 720\text{ cm}^{-1}$ vibrations of $-\text{CH}_2-$ groups are visible, similar to the raw material. The preservation of these peaks suggests that the dissolution step did not chemically alter the LDPE structure, confirming the step's effectiveness in physically processing the material without degradation.

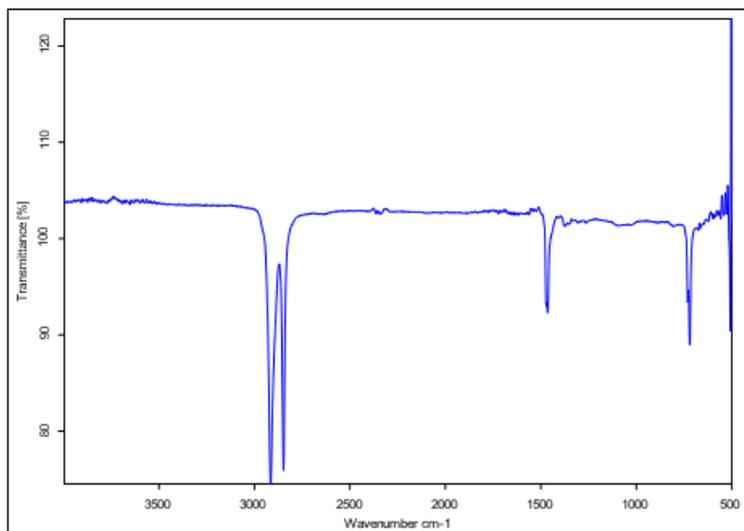


Fig 11. FTIR Analysis of Batch 1 Recycled LDPE

4. Batch 1 Recycled LDPE (step 2)

The FTIR spectrum of recovered LDPE exhibits characteristic absorption bands at $\sim 2915\text{ cm}^{-1}$ and $\sim 2849\text{ cm}^{-1}$, attributable to C-H asymmetric and symmetric stretching vibrations of $-\text{CH}_2-$ groups, respectively. The bending vibration peak at $\sim 1470\text{ cm}^{-1}$ and rocking motion band at $\sim 720\text{ cm}^{-1}$ confirm the preservation of the essential LDPE structure after the recovery process. Minor peaks in the $1700\text{--}1800\text{ cm}^{-1}$ range may indicate slight oxidative degradation during processing, though the intensity remains minimal compared to degraded samples. The overall spectral profile closely resembles virgin LDPE, confirming successful recovery with minimal structural alteration.

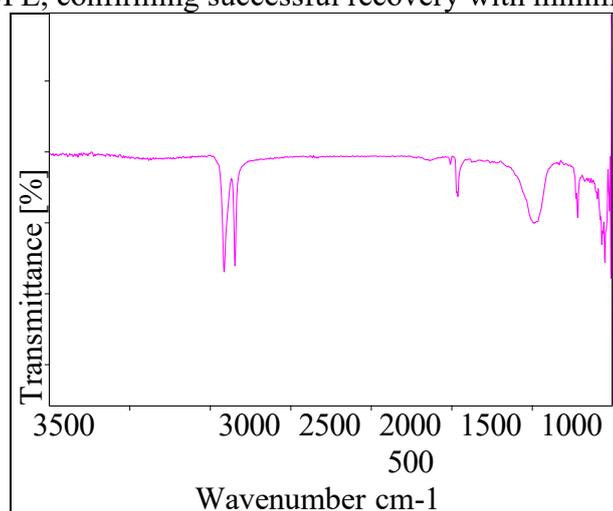


Fig 12. FTIR Analysis of Batch 1 Recycled LDPE (step 2)

5. Batch 4 Recycled LDPE (step 1)

The FTIR spectrum confirms the preserved LDPE structure, with characteristic peaks at 2920 cm^{-1} (asymmetric $-\text{CH}_2$ stretching) and 2851 cm^{-1} (symmetric $-\text{CH}_2$ stretching). Additional bands at 1464 cm^{-1} (CH_2 bending) and 888 cm^{-1} (CH_2 rocking) further validate the polymer backbone. High

transmittance and sharp peaks indicate minimal degradation, while the absence of carbonyl bands ($1700\text{--}1800\text{ cm}^{-1}$) confirms limited oxidation, aligning with the high recovery rate (88.6%) observed in Batch 4.

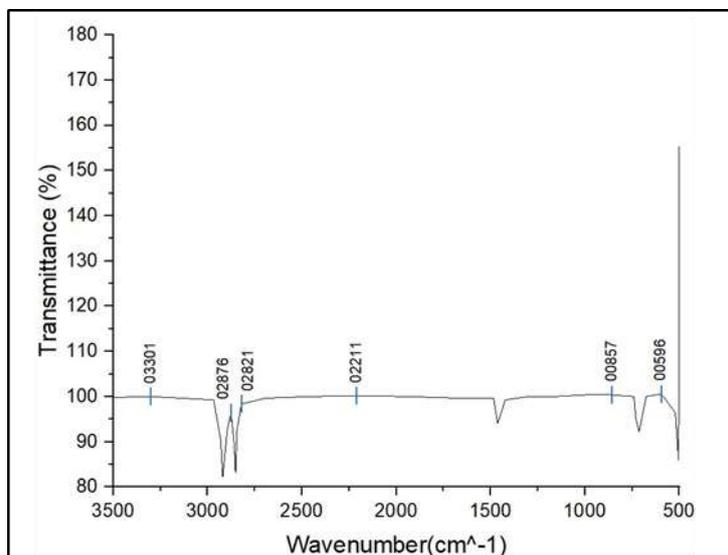


Fig 13. FTIR Analysis of Batch 4 LDPE

6. Batch 4 Recycled LDPE (step 2)

The spectrum shows reduced transmittance and flattened peaks, indicating structural modifications. While key LDPE bands (-CH_2 stretching at $2920/2851\text{ cm}^{-1}$) persist with lower intensity, confirming partial transformation, the retained polymer backbone aligns with the low conversion rate (11.83%). This supports efficient recovery, as most LDPE remained intact during step 2.

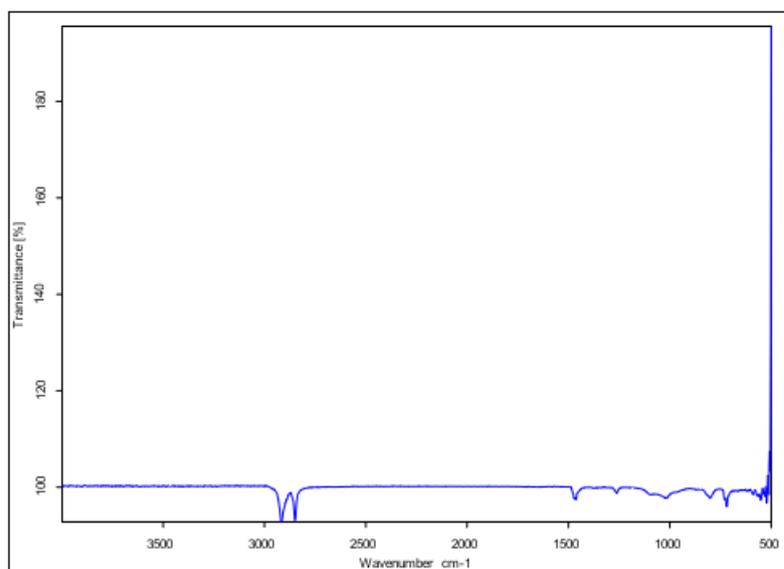


Fig 14. FTIR Analysis of Batch 4 Recycled LDPE (step 2)

3.3 Conversion and Recovery

The recovery and conversion data from the four experimental batches indicate a consistent and effective separation of LDPE, with Batch 4 yielding the highest recovery of 88.6%, making it the most optimized run in terms of product retrieval. Batch 1 and Batch 2 also showed high recoveries of 87.2% and 87.91%, respectively, while Batch 3 had a slightly lower value of 82%, potentially due to process variability. In terms of conversion, Batch 1 demonstrated the highest transformation efficiency at 62.8%, whereas Batch 2 showed a significantly lower conversion of 42.08%, likely due to partial



reaction or incomplete processing in the second step. These findings highlight the importance of process control and uniformity, as minor adjustments can substantially impact both recovery and conversion rates. Overall, the data supports the scalability of the optimized Batch 4 conditions for maximizing LDPE recovery in future applications.

In addition to the observed trends in recovery and conversion, it is important to note that the desired product in this process is the unreacted LDPE powder, as highlighted in the batch calculations. The term “conversion” here represents the fraction of LDPE that underwent reaction or degradation, which is considered a loss rather than a gain in this context. Therefore, a lower conversion percentage is actually favourable, indicating that a greater amount of LDPE remained unreacted and was successfully recovered. For example, Batch 4 not only achieved the highest recovery rate of 88.6% but also recorded the lowest conversion at 11.83%, confirming the process’s effectiveness in preserving LDPE integrity. This inverse relationship between recovery and conversion underscores the process optimization, where minimizing conversion directly correlates with maximizing product yield. The results collectively validate that the methodology is robust and suitable for efficient LDPE recycling, with the potential for scale-up and broader industrial application

Table 1: Optimized Batches Summary

Batch	Feed LDPE (g)	Recovered LDPE (g)	Recovery (%)	Reacted LDPE (g)	Conversion (%)
1	0.50	0.425	84.9	0.082	62.08
2	2.78	1.61	57.91	1.17	42.08
3 (only S1)	4	3.28	82	0.72	18
4	4.50	3.99	88.6	0.51	11.83

IV. CONCLUSIONS

This study successfully demonstrates a selective and efficient method for LDPE waste recovery using a combination of solvent-assisted dissolution–precipitation and low-temperature catalytic hydrothermolysis. Across the four experimental batches, the recovery of LDPE ranged from 57.91% to 88.6%, while conversion values varied between 11.83% and 62.08%. Among these, Batch 4 achieved the highest recovery (88.6%) and the lowest conversion (11.83%), indicating effective polymer preservation with minimal degradation. The superior performance of Batch 4 can be attributed to optimized operating conditions—specifically, an effective balance of acetone-to-water ratio that enhanced LDPE swelling, controlled catalyst dispersion (8 wt% ZSM- 5), and mild hydrothermal treatment at 70 °C. These factors collectively reduced chain scission and favored recovery over depolymerization, making Batch 4 the most efficient condition for selective LDPE recycling.

V. ACKNOWLEDGMENTS

The authors express their gratitude to the authorities of the Vishwakarma Institute of Technology, Pune, and the Department of Chemical Engineering for their essential assistance and continuous support in completing this work.

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