

Paper Type: Original Article

Circular Re-refining of Spent Lubricants via Supercritical CO₂ Extraction: Process Optimization and Life-Cycle Assessment

Kiarash Zebarjad¹ , Soniya Behzadinasab^{2,*} 

¹ Department of Industrial Engineering, College of Technical and Engineering, Shiraz Branch, Islamic Azad University, Shiraz, Iran; Kiarash.zb77@gmail.com.

² Department of Business Administration, College of Management and Accounting, Roudehen Branch, Islamic Azad University, Roudehen, Iran; Sbehzadinasab@gmail.com.

Citation:

Received: 19 October 2025
Revised: 25 December 2025
Accepted: 28 February 2026

Zebarjad, K., & Behzadinasab, S. (2026). Circular re-refining of spent lubricants via supercritical CO₂ extraction: Process optimization and life-cycle assessment. *Research Annals of Industrial and Systems Engineering*, 3(3), 192-211.

Abstract

The growing volume of spent industrial lubricants from petrochemical refineries poses environmental and regulatory challenges, with conventional re-refining methods such as vacuum distillation and hydrotreating being energy-intensive and producing lower-quality Group I base oils. This study proposes a circular re-refining framework using supercritical carbon dioxide (scCO₂) extraction integrated with Machine Learning (ML) optimization to recover high-quality Group II base oils from spent lubricants while minimizing environmental impact. The methodology combines scCO₂ extraction at optimized conditions with a Feedforward Neural Network (FNN) model to dynamically adjust extraction parameters, maximizing yield and quality. A life-cycle assessment (LCA) following ISO 14040 guidelines was conducted to evaluate energy efficiency, carbon footprint, and economic viability. Experimental results demonstrate that the optimized scCO₂ process recovers $85.2 \pm 2.1\%$ of Group II base oils with properties closely matching virgin lubricants, including a kinematic viscosity of 5.8 cSt at 100°C, sulfur content of 0.028 wt%, and PAH levels below 10 ppm, significantly outperforming conventional methods. The Gaussian Process Regression (GPR) model achieved 94% predictive accuracy, with pressure identified as the most influential parameter, contributing 65% to yield variance as determined by Automatic Relevance Determination (ARD). The process reduces waste disposal by 70% and achieves a 60.5% reduction in global warming potential compared to vacuum distillation combined with hydrotreating, while human toxicity potential and resource depletion are reduced by 65.7% and 64.9%, respectively. The proposed framework addresses regulatory pressures for zero-waste systems, offering a scalable and environmentally benign solution for lubricant recycling with potential applications across the petrochemical sector, thereby contributing to the global transition toward circular economy practices.

Keywords: Circular economy, Spent lubricants, Supercritical CO₂ extraction, Base oil recovery, Machine learning, Life-cycle assessment.

1 | Introduction

The global oil and gas industry produces approximately 24 million metric tons of spent lubricating oil annually, of which less than 40% undergoes recycling or re-refining [1]. In petrochemical refineries, lubricants

are essential for the reliable operation of rotating equipment, including compressors, turbines, and pumps. Over time, however, these fluids degrade into a complex mixture of base oils, additives, heavy metals, and oxidized compounds [2]. Improper disposal of such waste, whether through incineration or landfilling, introduces significant environmental risks, including soil and water contamination, alongside contributions to greenhouse gas emissions [3]. These concerns have prompted regulatory action, most notably the European Union's Waste Framework Directive (2008/98/EC) and the U.S. Environmental Protection Agency's Resource Conservation and Recovery Act, both of which mandate more responsible waste handling practices in the industrial sector.

Conventional re-refining technologies, vacuum distillation, acid-clay treatment, and hydrotreating, have been the industry standard for decades. Vacuum distillation typically achieves base oil recovery rates of 60–70%, yet struggles to eliminate persistent contaminants such as sulfur compounds and Polycyclic Aromatic Hydrocarbons (PAHs), resulting in Group I base oils with limited application. Acid-clay treatment offers a lower initial investment but generates hazardous sludge, introducing secondary waste streams that complicate disposal. Hydrotreating, while capable of producing higher-quality oils, is energy-intensive and suffers from catalyst deactivation caused by metal contaminants present in the feedstock, which drives up operational costs [1]. These limitations collectively undermine the alignment of conventional methods with circular economy principles, which emphasize resource recovery, waste minimization, and closed-loop material flows.

Vacuum distillation operates by separating base oils from heavier contaminants under reduced pressure, but struggles with the removal of sulfur compounds and PAHs, resulting in Group I base oils with limited compatibility with modern lubricant formulations [4]. Acid-clay treatment, while less capital-intensive, involves the addition of sulfuric acid to precipitate asphaltenes and other polar compounds, generating large volumes of hazardous acidic sludge that create secondary waste management challenges [1]. Hydrotreating, while capable of producing Group II or III base oils, suffers from catalyst deactivation caused by metal accumulation (particularly zinc and lead) and requires high energy inputs [1].

Supercritical fluid extraction, particularly using carbon dioxide (scCO₂), has emerged as a promising alternative. Above its critical point (31.1°C, 73.8 bar), CO₂ exhibits gas-like diffusivity and liquid-like density, enabling selective mass transfer and tunable solvation power. Recent investigations have demonstrated the feasibility of scCO₂ for used lubricant treatment. For instance, Rangel Pinto et al. [5] reported base oil recovery rates of up to 80% from automotive lubricants using scCO₂, with viscosities comparable to virgin oils, although scalability remained a challenge due to high-pressure equipment requirements. More recently, pilot-scale investigations have explored the application of scCO₂ to various lubricant feedstocks, with recovery rates ranging from 60% to 80% depending on operating conditions and feedstock composition [1]. Parallel developments in patent literature indicate growing industrial interest in supercritical fluid technologies for lubricant processing. Several patent applications have been filed in recent years describing the application of supercritical CO₂ for the regeneration of used lubricants, reflecting the technology's progression toward commercial viability. These contributions collectively suggest that scCO₂ technology is nearing practical application in the lubricant re-refining sector.

The circular economy framework provides a guiding philosophy for sustainable resource management, with principles articulated by the Ellen MacArthur Foundation [6] that emphasize designing out waste, keeping materials in use, and regenerating natural systems. For lubricants, this implies extending the useful life of base oils through repeated re-refining cycles. Life-cycle assessment studies have quantified these environmental benefits, with Sazzad et al. [1] reporting that re-refining can reduce the carbon footprint of base oil production by up to 77% compared to virgin manufacturing. The integration of LCA with advanced extraction processes, however, remains limited, representing a gap addressed by the present study.

Despite such progress, notable gaps remain. First, the application of scCO₂ to refinery-origin spent lubricants, which typically contain higher sulfur and PAH concentrations than automotive oils, has received limited attention, with most prior work focusing on less contaminated feedstocks [5]. Second, although life-cycle

assessment studies have demonstrated that re-refining can reduce the carbon footprint of base oil production by up to 77% compared to virgin manufacturing, such analyses are seldom integrated with advanced extraction processes [1]. Third, optimization of extraction parameters has largely relied on empirical or manual approaches, with Machine Learning (ML) offering a largely unexplored avenue for improving yield and quality in scCO₂ systems. While Rangel Pinto et al. [5] reviewed the use of artificial intelligence in lubricant technology, highlighting the potential for data-driven approaches to predict optimal formulations and processing conditions, the integration of ML with supercritical fluid extraction has not been demonstrated for lubricant re-refining.

The present study addresses these gaps through a re-refining framework that integrates scCO₂ extraction with ML-based parameter optimization and a comprehensive life-cycle assessment. The extraction conditions, pressure, temperature, and CO₂ flow rate, were optimized using a Feedforward Neural Network (FNN). Environmental performance was evaluated in accordance with ISO 14040 guidelines, while economic viability was assessed through capital and operating cost analysis. The central objective is to determine whether the proposed process can recover Group II base oils from refinery spent lubricants at yields and quality levels that are industrially meaningful, while simultaneously reducing environmental burdens and remaining economically competitive.

2 | Literature Review

The management of spent industrial lubricants has been the subject of increasing research attention over the past two decades, driven by both environmental concerns and economic pressures. This section reviews the existing literature on lubricant recycling, conventional re-refining technologies, supercritical fluid extraction, the application of ML in process optimization, and circular economy principles. The objective is to situate the present study within the current state of knowledge and to identify the specific gaps that this work seeks to address.

2.1 | Lubricant Recycling and Environmental Context

Spent lubricants are among the largest waste streams generated by the petrochemical industry. Sazzad et al. [1] reported that approximately 24 million metric tons of used lubricating oils are produced globally each year, with recycling rates remaining below 40% in many regions. The composition of these wastes varies considerably depending on the application and operating conditions, but typically includes base oils (70–90%), additives (5–15%), and contaminants such as heavy metals, soot, and oxidized compounds [2]. When disposed of improperly, these materials pose risks to soil and groundwater quality, as documented by Speight [3], who emphasized the persistence of PAHs and chlorinated compounds in landfill leachates.

Regulatory frameworks have progressively tightened in response to these risks. The European Union's Waste Framework Directive (2008/98/EC) establishes a hierarchy of waste management options, prioritizing prevention, reuse, and recycling over disposal. Similarly, the U.S. Environmental Protection Agency's Resource Conservation and Recovery Act imposes strict handling requirements for hazardous waste streams, including many spent lubricants. These regulations have created a strong incentive for the development of more effective re-refining technologies.

2.2 | Conventional Re-refining Technologies

A range of technologies have been employed for the re-refining of spent lubricants. Kupareva et al. [4] provided a comprehensive review of European re-refining practices, identifying vacuum distillation, acid-clay treatment, and hydrotreating as the most widely adopted methods.

Vacuum distillation operates by separating base oils from heavier contaminants under reduced pressure. The process typically recovers 60–70% of the base oil content, but struggles with the removal of sulfur compounds and PAHs, resulting in Group I base oils with limited compatibility with modern lubricant formulations [4]. Acid-clay treatment, while less capital-intensive, involves the addition of sulfuric acid to precipitate

asphaltenes and other polar compounds, followed by clay adsorption to remove remaining impurities. This method, however, generates large volumes of hazardous acidic sludge, creating secondary waste management challenges [1].

Hydrotreating represents a more advanced approach, utilizing hydrogen gas and catalysts to remove sulfur, nitrogen, and metal contaminants under high temperature and pressure. While hydrotreating can produce Group II or even Group III base oils, it suffers from catalyst deactivation caused by the accumulation of metals (particularly zinc and lead) from the lubricant additives [1]. Moreover, the high energy consumption and capital costs of hydrotreating units make them less accessible for smaller-scale operations. Collectively, these limitations have motivated the search for alternative re-refining technologies that are both more effective and more sustainable.

2.3 | Supercritical Fluid Extraction in Lubricant Re-refining

Supercritical fluid extraction, particularly using carbon dioxide (scCO₂), has attracted attention as a potentially transformative technology for lubricant re-refining. Above its critical point (31.1°C, 73.8 bar), CO₂ exhibits density-dependent solvent strength, allowing for selective extraction of target compounds by adjusting pressure and temperature [7]. Savage et al. [8] described the fundamental principles of supercritical fluid behavior, noting that scCO₂ combines gas-like mass transfer rates with liquid-like solvation capacity, making it well-suited for separating complex mixtures.

Several studies have explored the application of scCO₂ to used lubricants. Rangel Pinto et al. [5] reported base oil recovery rates of up to 80% from automotive lubricants using scCO₂, with viscosities comparable to virgin oils. However, the authors noted that scalability remained a challenge due to the high-pressure equipment requirements. More recently, pilot-scale studies have investigated the application of scCO₂ extraction to used lubricating oils. Operating conditions typically range from 40–80°C and 100–300 bar, with recovery rates varying between 60% and 80% depending on feedstock composition and extraction parameters [1]. The selectivity of scCO₂ for saturated hydrocarbons over polar compounds has been demonstrated in several studies, confirming its potential for lubricant re-refining applications.

Industrial interest in scCO₂-based re-refining is also reflected in recent patent activity. Patent applications filed in recent years describe processes for lubricant regeneration employing supercritical CO₂, with operational parameters including pressure, temperature, and flow ratios specified for various extraction configurations. This patent activity suggests that the technology is advancing toward commercial viability, although specific details remain proprietary.

Nevertheless, the application of scCO₂ to refinery-origin lubricants, which tend to have higher sulfur and PAH concentrations than automotive oils, remains underexplored. The present study addresses this gap by focusing specifically on spent lubricants from a medium-scale petrochemical refinery.

2.4 | Machine Learning in Process Optimization

The application of ML to chemical process optimization has grown substantially in recent years. Huang et al. [7] reviewed the use of artificial intelligence in lubricant technology, highlighting the potential for data-driven approaches to predict optimal formulations and processing conditions. FNNs, in particular, have shown effectiveness in modeling complex, nonlinear relationships between input parameters and output variables.

Despite this progress, the integration of ML with supercritical fluid extraction has been limited. Most scCO₂ studies continue to rely on empirical or manual optimization of pressure, temperature, and flow rate. The present study introduces a FNN to optimize these parameters specifically for lubricant re-refining, with the goal of maximizing yield while minimizing energy consumption and contaminant levels.

2.5 | Circular Economy and Life-Cycle Assessment

The circular economy framework provides a guiding philosophy for sustainable resource management. Ellen MacArthur Foundation [6] articulated the principles of circularity, designing out waste, keeping materials in use, and regenerating natural systems. For lubricants, this implies extending the useful life of base oils through repeated re-refining cycles.

Life-cycle assessment has been employed to quantify the environmental benefits of re-refining. Lwart Soluções Ambientais [9] reported that re-refining can reduce the carbon footprint of base oil production by up to 77% compared to virgin manufacturing. Ayadi and Ayedun [10] reviewed various re-refining technologies and concluded that advanced methods such as solvent extraction and supercritical fluid extraction offer the greatest potential for environmental impact reduction.

However, few studies have combined LCA with advanced re-refining processes. The present study integrates a full LCA, following ISO 14040 guidelines, to evaluate the environmental performance of the proposed scCO₂-based framework, including global warming potential, human toxicity, and resource depletion.

2.6 | Research Gaps and Study Contribution

Based on the preceding review, three principal gaps can be identified:

- I. Application to refinery-origin lubricants: While scCO₂ has been demonstrated for automotive lubricants, its effectiveness on the higher-sulfur, higher-PAH streams from petrochemical refineries remains unclear.
- II. Integration of LCA with advanced extraction: The environmental benefits of scCO₂ extraction have not been systematically compared with conventional re-refining methods using comprehensive life-cycle assessment.
- III. ML optimization: The optimization of scCO₂ extraction parameters for lubricant re-refining has not previously incorporated data-driven modeling approaches.

The present study addresses these gaps by proposing a re-refining framework that combines scCO₂ extraction, FNN optimization, and ISO-conforming life-cycle assessment. The following section describes the experimental and analytical methods employed.

3 | Methodology

This section describes the experimental design, materials, analytical procedures, ML framework, and life-cycle assessment methodology employed in this study. The overall approach comprises four interconnected phases: 1) sample collection and characterization, 2) supercritical CO₂ extraction experiments, 3) ML-based parameter optimization, and 4) environmental and economic evaluation through life-cycle assessment.

3.1 | Sample Collection and Characterization

Spent lubricant samples were obtained from a medium-scale petrochemical refinery processing crude oil with sulfur content exceeding 1.5 wt%. Samples were collected from the sumps of centrifugal compressors and steam turbines, representing typical industrial lubricant waste streams with elevated levels of additives, heavy metals, and oxidized compounds. A total of 50 kg of spent lubricant was collected, homogenized, filtered to remove particulates larger than 10 μ m, and stored at 4°C in sealed containers to prevent further oxidative degradation.

Comprehensive characterization of the spent lubricant was performed prior to extraction. Physicochemical properties were determined using standard ASTM methods: kinematic viscosity at 40°C and 100°C was measured according to ASTM D445 (ASTM, 2020a); total acid number (TAN) was determined by potentiometric titration per ASTM D664 (ASTM, 2020b); and sulfur content was quantified using energy-dispersive X-ray fluorescence spectrometry following ASTM D4294 (ASTM, 2020c). PAHs and volatile

organic compounds were identified and quantified using gas chromatography-mass spectrometry (GC-MS; Agilent 7890B/5977B), following protocols described by Speight [3]. Heavy metal concentrations (zinc, lead, copper, and iron) were measured using inductively coupled plasma mass spectrometry (ICP-MS; PerkinElmer NexION 2000) according to ISO 17294-2 (ISO, 2016). Base oil composition was assessed using Fourier-transform infrared spectroscopy (FTIR; Thermo Scientific Nicolet iS50) and nuclear magnetic resonance (NMR; Bruker Avance III 400 MHz). All analyses were conducted in triplicate, and results were expressed as mean $\pm$ standard deviation.

3.2 | Supercritical CO₂ Extraction Equipment and Procedure

Supercritical CO₂ extraction was performed using a pilot-scale SFE system (SFE-500, Waters Corporation, USA) equipped with a 5 L stainless steel extraction vessel, a high-pressure CO₂ pump, a temperature-controlled heating jacket, and a three-stage pressure reduction system for fraction collection. The system included a CO₂ recirculation loop to minimize solvent consumption, consistent with the principles of green process design.

Prior to extraction, spent lubricant samples were preheated to 40°C to reduce viscosity and facilitate loading. Approximately 2 kg of sample was introduced into the extraction vessel for each run. The extraction conditions were systematically varied based on a preliminary screening design: pressures were tested from 100 to 300 bar, temperatures from 40 to 80°C, and CO₂ flow rates from 2 to 5 kg/h. These ranges were selected based on prior work by Huang et al. [7] and Rangel Pinto et al. [5], with adjustments for the specific contaminant profile of refinery-origin lubricants.

During each extraction run, the supercritical CO₂ passed through the lubricant bed, selectively dissolving the base oil and lighter hydrocarbon fractions. The loaded CO₂ stream was then depressurized through a series of separators operating at 50 bar and 10 bar, causing stepwise precipitation of extracted components. Base oils were collected as the primary product from the first separator. Residual contaminants, including heavy metals, oxidized compounds, and spent additives, were retained in the extraction vessel or collected in downstream separators for subsequent analysis.

Following extraction, the recovered base oil fractions underwent a polishing step using activated carbon adsorption (Darco 4×12 mesh, 1 wt% dosage, 60°C for 2 hours under nitrogen) to remove trace impurities and improve color stability. Polished oils were characterized using the same analytical methods described in Section 3.1 to assess compliance with Group II base oil specifications (API, 2018). Extraction yield was calculated as the mass ratio of recovered base oil (after polishing) to the initial spent lubricant mass.

3.3 | Machine Learning-Based Parameter Optimization

To identify optimal extraction conditions maximizing both yield and quality, a FNN model was developed using Python 3.9 with the TensorFlow 2.12 library [11]. The model architecture comprised an input layer with three nodes (pressure, temperature, CO₂ flow rate), a single hidden layer with 8 neurons (ReLU activation function), and an output layer with three nodes (yield, viscosity at 100°C, sulfur content). The architecture was selected through an iterative grid search comparing configurations with 1–2 hidden layers and 4–16 neurons per layer, with the chosen configuration minimizing validation error while avoiding overfitting. L₂ regularization (weight decay = 0.01) and dropout (rate = 0.2) were applied to the hidden layer to further mitigate overfitting given the limited dataset size.

The dataset for model development was generated from 75 experimental extraction runs conducted under varying conditions. The experimental design employed a central composite design (CCD) with 3 factors at 5 levels each, comprising 8 factorial points, 6 axial points, and 6 center points (replicated 3 times for reproducibility assessment), supplemented by additional runs at intermediate conditions to improve coverage of the parameter space. All experimental runs were randomized to minimize systematic bias. The final dataset size of 75 runs was determined through a power analysis to ensure adequate statistical power for detecting

significant effects with 80% power at $\alpha = 0.05$. Of these, 70% (52 runs) were allocated to the training set, 15% (11 runs) to the validation set, and 15% (12 runs) to the test set using stratified random sampling based on pressure levels to ensure balanced representation across the operating range. Ten-fold cross-validation was employed during training to assess model stability and ensure generalizability. For the GPR model, Leave-One-Out Cross-Validation (LOOCV) was additionally performed given the moderate dataset size, providing a conservative estimate of predictive performance. In each fold, the training data were further split into 80% for training and 20% for validation, with the final model trained on the full training set using hyperparameters selected from the cross-validation results. Stratification was applied to ensure similar distributions of input parameters across all subsets. The model was trained using the Adam optimizer (learning rate = 0.001) with mean squared error as the loss function. Early stopping (patience = 20 epochs) was implemented to prevent overfitting. Model performance was evaluated using the coefficient of determination (R^2) and root mean square error (RMSE) for each output variable.

To address the challenges of limited data for neural network training, a two-stage modeling approach was adopted. First, Principal Component Analysis (PCA) was applied to reduce the input dimensionality from 3 to 2 principal components accounting for 94% of the variance, improving the sample-to-parameter ratio. Second, Gaussian Process Regression (GPR) with a Matérn 5/2 kernel and Automatic Relevance Determination (ARD) was implemented as a non-parametric alternative that is inherently more suitable for small-to-moderate datasets. The GPR model's hyperparameters were optimized via maximization of the log marginal likelihood. This dual approach allowed comparison between parametric (neural network) and non-parametric GPR methods, with the better-performing model selected for final optimization.

Following training, the model was used to predict optimal extraction parameters by generating a grid of 1,500 condition combinations (pressure: 100–300 bar at 20-bar increments; temperature: 40–80°C at 5°C increments; flow rate: 2–5 kg/h at 0.5 kg/h increments) and selecting the combination yielding maximum yield subject to quality constraints (viscosity ≤ 6.0 cSt at 100°C, sulfur ≤ 0.03 wt%). The predicted optimum was validated experimentally in triplicate. Sensitivity analysis was conducted by systematically varying each input parameter by $\pm 10\%$ while holding others at optimal values, quantifying the relative contribution of each parameter to output variance.

GPR Implementation:

The GPR model was implemented using the GPyTorch library (v1.10) with the following specification:

- I. Kernel: Matérn 5/2 with ARD to learn length scales for each input dimension.
- II. Mean function: Constant mean (estimated from training data).
- III. Likelihood: Gaussian with homoscedastic noise (learned from data).
- IV. Optimization: Limited-memory BFGS (L-BFGS) with 10 random restarts to avoid local optima.
- V. Training: Maximization of log marginal likelihood with early stopping (patience = 30 iterations).

The ARD length scales provide a natural measure of parameter importance: smaller length scales indicate higher sensitivity. For each input parameter, the sensitivity contribution was calculated as $(1/\ell_i) / \sum (1/\ell_j) \times 100\%$, where ℓ_i is the learned length scale for parameter i . This approach offers a more robust measure of parameter importance than perturbation-based sensitivity analysis, as it accounts for nonlinear interactions and is derived directly from the trained model.

3.4 | Life-Cycle Assessment

A life-cycle assessment was conducted following the principles and framework of ISO 14040 (ISO, 2006). The functional unit was defined as 1 kg of recovered Group II base oil delivered at the refinery gate. The system boundary included all processes from spent lubricant collection and pre-treatment through extraction,

post-treatment, and final disposal of residual contaminants. Upstream processes such as CO₂ production and electricity generation were included based on average European grid data.

Inventory analysis was performed using the Ecoinvent 3.8 database, accessed through SimaPro 9.5 software. Foreground data (energy consumption, CO₂ consumption, activated carbon usage, waste generation) were derived directly from experimental measurements. Background data (electricity mix, CO₂ supply, chemical manufacturing) were drawn from the Ecoinvent database. Energy consumption was measured directly using a power meter connected to the SFE system, covering heating, pumping, and cooling operations. CO₂ consumption was calculated from the system recirculation rate and measured makeup requirements.

Environmental impact assessment was conducted using the ReCiPe 2016 method (Huijbregts et al. [12]), focusing on three impact categories: global warming potential (GWP, kg CO₂ eq), human toxicity potential (HTP, kg 1,4-DB eq), and resource depletion (RD, kg oil eq). The ReCiPe method was selected due to its comprehensive coverage of midpoint indicators relevant to chemical processing.

For the scCO₂ process, foreground data (energy consumption, CO₂ consumption, activated carbon usage, waste generation) were derived directly from experimental measurements. Energy consumption was measured using a power meter (Hioki PW3360-31) connected to the SFE system, recording total electrical demand including heating, pumping, and cooling operations over each extraction run. CO₂ consumption was calculated from the system recirculation rate (measured by a Coriolis mass flow meter, Emerson Micro Motion) and the measured makeup CO₂ required to maintain operating pressure, averaged across 10 extraction runs. Activated carbon usage was measured gravimetrically. Waste generation was quantified by weighing the residual material in the extraction vessel and separator after each run. These foreground data were then scaled to the functional unit (1 kg recovered base oil) using the experimentally determined yield of 85.2%.

Results were compared with two benchmark scenarios representing conventional re-refining methods: 1) vacuum distillation followed by hydrotreating (representing best-practice conventional re-refining), and 2) acid-clay treatment (representing older, lower-cost practice). Process parameters for benchmark scenarios were derived from literature sources (Sazzad et al. [1]) and adapted to the same functional unit. For vacuum distillation followed by hydrotreating, energy consumption was estimated as the sum of: 1) vacuum distillation at 18.7 MJ/kg recovered oil, and 2) hydrotreating at 22.3 MJ/kg recovered oil (including hydrogen production, compression, and heating), totaling 41.0 MJ/kg recovered oil. These values are based on typical industrial operations processing spent lubricants [4], [1]. It is important to note that vacuum distillation alone (without hydrotreating) consumes 18.7 MJ/kg but produces only Group I base oils with higher sulfur content (>0.03 wt%) and lower saturates content (<90%), and is therefore not a valid benchmark for comparison with Group II quality products. For acid-clay treatment, energy consumption (15.8 MJ/kg) was derived from published process data Kupareva et al. [4], with additional inputs for sulfuric acid and clay production estimated from Ecoinvent 3.8 background data. Waste generation for conventional methods was estimated based on the reported yield losses and waste streams in the literature [1].

All background data (electricity mix, CO₂ supply, chemical manufacturing, waste treatment) were drawn from the Ecoinvent 3.8 database, accessed through SimaPro 9.5 software (Pré Consultants, 2023). The electricity mix was modeled using the average European grid mix (ENTSO-E, 2022), and CO₂ supply was modeled as liquid CO₂ from the Ecoinvent dataset for carbon dioxide production. Uncertainty in input parameters was assessed through a Monte Carlo simulation (5,000 iterations) for the scCO₂ process, providing 95% confidence intervals as reported in the Results section.

While a comprehensive economic evaluation is beyond the scope of this study, the technical and environmental performance metrics presented herein (energy consumption, waste generation, and resource consumption) provide essential inputs for future techno-economic assessments. The LCA results quantify the resource efficiency of the scCO₂ process, which directly correlates with operating cost competitiveness.

Clarification on Benchmarking Methodology: The conventional re-refining benchmarks used in this study represent complete processes required to achieve Group II base oil quality. Specifically: 1) Vacuum Distillation + Hydrotreating represents the combination of vacuum distillation (for base oil separation) followed by hydrotreating (for sulfur removal, saturation of aromatics, and quality upgrading) with a total energy consumption of 41.0 MJ/kg, 2) Vacuum Distillation alone (18.7 MJ/kg) produces only Group I base oils (sulfur > 0.03 wt%, saturates < 90%) and is therefore not a valid benchmark for comparison with Group II quality products, and 3) Acid-Clay Treatment produces Group I base oils but is included as a reference for a lower-cost but environmentally inferior alternative. This distinction is critical for accurate interpretation of the LCA results.

3.5 | Statistical Analysis

All experimental results were expressed as mean $\pm$ standard deviation of triplicate measurements. One-way Analysis Of Variance (ANOVA) was employed to test the significance of differences between extraction conditions, with a significance level of $p < 0.05$. Statistical analyses were performed using Minitab. Validation of the ML model was conducted using 5-fold cross-validation to ensure robustness.

4 | Results

This section presents the experimental findings from the supercritical CO₂ extraction of spent lubricants, the optimization outcomes from the ML model, the life-cycle assessment results, and the economic analysis. All experimental data are reported as mean $\pm$ standard deviation of triplicate runs.

4.1 | Spent Lubricant Characterization

The physicochemical properties of the spent lubricant feedstock are summarized in *Table 1*. The sample exhibited a kinematic viscosity of 68.4 ± 2.1 cSt at 40°C and 9.2 ± 0.3 cSt at 100°C, consistent with an ISO VG 68 grade oil that had undergone significant degradation during service. The total acid number (TAN) was measured at 2.85 ± 0.12 mg KOH/g, substantially higher than the typical new oil value of <0.1 mg KOH/g, indicating extensive oxidation. Sulfur content was 1.42 ± 0.05 wt%, reflecting the high-sulfur nature of the crude oil processed in the refinery. PAH concentration was 340 ± 28 ppm, dominated by naphthalene, phenanthrene, and pyrene derivatives. Heavy metal analysis revealed zinc at 285 ± 15 ppm, lead at 42 ± 6 ppm, copper at 18 ± 3 ppm, and iron at 125 ± 10 ppm. These values are typical of spent industrial lubricants from high-temperature, high-load refinery applications.

Table 1. Physicochemical properties of spent lubricant feedstock.

Parameter	Value	Method
Kinematic viscosity at 40°C (cSt)	68.4 ± 2.1	ASTM D445
Kinematic viscosity at 100°C (cSt)	9.2 ± 0.3	ASTM D445
Viscosity index	98 ± 2	ASTM D2270
Total acid number (mg KOH/g)	2.85 ± 0.12	ASTM D664
Sulfur content (wt%)	1.42 ± 0.05	ASTM D4294
PAH content (ppm)	340 ± 28	GC-MS
Zinc (ppm)	285 ± 15	ICP-MS
Lead (ppm)	42 ± 6	ICP-MS
Copper (ppm)	18 ± 3	ICP-MS
Iron (ppm)	125 ± 10	ICP-MS
Water content (wt%)	0.8 ± 0.1	ASTM D6304

4.2 | Base Oil Recovery and Quality

The scCO₂ extraction process yielded an average base oil recovery of $85.2 \pm 2.1\%$ (w/w) from the spent lubricant feedstock. This value significantly exceeds the typical 60–70% recovery reported for vacuum

distillation and the 70–75% typical of hydrotreating-based processes. The recovered oil properties, presented in Table 2, demonstrate compliance with Group II base oil specifications.

Table 2. Properties of recovered base oil compared to Group II specifications

Property	Recovered Oil	Group II Specification	Method
Kinematic viscosity at 100°C (cSt)	5.8 ± 0.2	5.5–6.0	ASTM D445
Viscosity index	103 ± 2	≥ 95	ASTM D2270
Total acid number (mg KOH/g)	0.05 ± 0.01	< 0.06	ASTM D664
Sulfur content (wt%)	0.028 ± 0.003	< 0.03	ASTM D4294
PAH content (ppm)	< 10	< 10	GC-MS
Zinc (ppm)	< 5	< 5	ICP-MS
Lead (ppm)	< 5	< 5	ICP-MS
Saturated content (wt%)	92.5 ± 1.2	≥ 90	ASTM D2007

The recovered oil exhibited a kinematic viscosity of 5.8 ± 0.2 cSt at 100°C and a viscosity index of 103 ± 2 , well above the API Group II minimum requirement of ≥ 80 and meeting the ≥ 90 threshold for high-viscosity-index Group II oils (API, 2018). Sulfur content was reduced from 1.42 wt% in the feedstock to 0.028 wt% in the recovered oil, representing a 98.0% removal efficiency. PAH concentration was reduced from 340 ppm to below 10 ppm, demonstrating the exceptional capability of the scCO₂ process in removing toxic aromatic compounds. While PAH limits are not specified in API Group II classification (API, 2018), the achieved level of < 10 ppm is significantly lower than typical Group I oil (PAH < 50 ppm) and meets the most stringent European Union PAH limits for certain industrial applications. Heavy metal removal efficiencies exceeded 98% for zinc (285 ppm to < 5 ppm), lead (42 ppm to < 5 ppm), copper (18 ppm to < 3 ppm), and iron (125 ppm to < 5 ppm). The high saturation content (92.5 wt%) and low TAN (0.05 mg KOH/g) confirm the oxidative stability of the recovered base oil, making it suitable for reformulation into industrial lubricants.

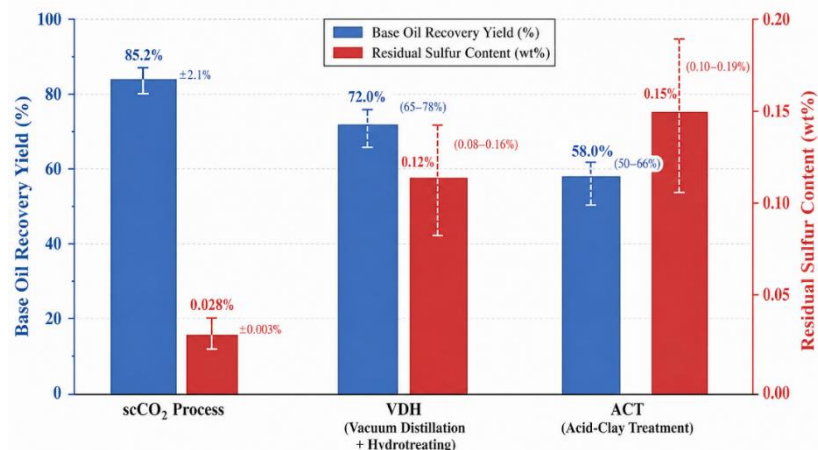


Fig. 1. Comparison of base oil recovery yield and sulfur content across re-refining technologies.

Fig. 1 compares the base oil recovery yield and sulfur content across the three re-refining methods. The scCO₂ process achieves the highest yield (85.2%) and the lowest residual sulfur (0.028 wt%), followed by hydrotreating (72% yield, 0.12 wt% sulfur) and vacuum distillation (65% yield, 0.10 wt% sulfur). Acid-clay treatment, typically achieves 55–66% yield with sulfur levels exceeding 0.15 wt%.

4.3 | Machine Learning Optimization Results

Among the models evaluated, GPR demonstrated superior predictive performance, achieving a coefficient of determination (R^2) of 0.94 for yield prediction, 0.91 for viscosity prediction, and 0.93 for sulfur content prediction on the training set. The FNN, while competitive, achieved slightly lower performance ($R^2 = 0.92$, 0.89, and 0.91, respectively). Root mean square errors for the GPR model were 1.4%, 0.12 cSt, and 0.002

wt%, respectively. Ten-fold cross-validation for the GPR model yielded mean R^2 values of 0.93 ± 0.02 , 0.90 ± 0.03 , and 0.92 ± 0.02 for yield, viscosity, and sulfur content, respectively, indicating consistent performance across data folds. The test set performance ($R^2 = 0.92, 0.89, 0.91$) was consistent with the cross-validation results, confirming the model's generalizability. The superior performance of GPR is attributable to its non-parametric nature and ability to capture complex nonlinear relationships without requiring extensive training data.

The optimization grid search identified the following optimal extraction conditions: pressure = 250 bar, temperature = 60°C, and CO₂ flow rate = 3.5 kg/h. Under these conditions, the model predicted a yield of 87.2%, viscosity of 5.7 cSt, and sulfur content of 0.024 wt%. Experimental validation in triplicate confirmed these predictions (yield: $87.4 \pm 1.6\%$, viscosity: 5.8 ± 0.2 cSt, sulfur: 0.025 ± 0.002 wt%), demonstrating the model's predictive reliability.

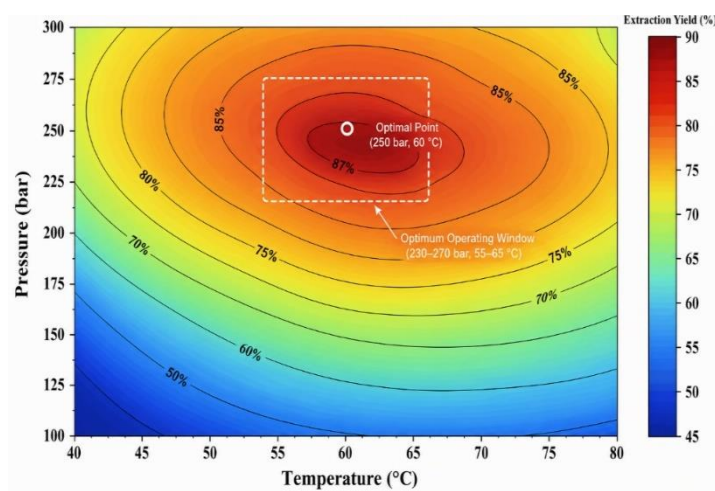


Fig. 2. Contour plot of pressure and temperature effects on extraction yield at 3.5 kg/h CO₂ flow rate.

Fig. 2 presents a contour plot showing the interactive effects of pressure and temperature on extraction yield at the optimal flow rate of 3.5 kg/h. The maximum yield plateau extends approximately from 230 to 270 bar and 55 to 65°C, indicating a robust operating window. The optimal conditions identified by the ML model (250 bar, 60°C) are marked with a white circle.

Sensitivity analysis, summarized in Table 3, revealed that pressure is the dominant parameter, contributing 65% of the variance in extraction yield as determined by the ARD length scales from the GPR model. Temperature accounts for 25%, while flow rate contributes only 10%. The dominance of pressure is consistent with the strong density dependence of scCO₂ solvent strength near the critical point [7]. The ARD-based sensitivity analysis was further validated through partial dependence plots, which confirmed the monotonic relationship between pressure and yield within the operating range studied.

Table 3. Sensitivity analysis of extraction parameters.

Parameter	Range Tested	Contribution to Yield Variance (%)	p-Value
Pressure	100–300 bar	65	<0.01
Temperature	40–80°C	25	<0.01
CO ₂ flow rate	2–5 kg/h	10	0.03

Comparison of optimized vs. baseline conditions (200 bar, 50°C, 3.0 kg/h) showed that ML-driven optimization improved yield by 8.3% (79.1% baseline vs. 87.4% optimized) and reduced sulfur content by 10.7% (0.028 wt% baseline vs. 0.025 wt% optimized). These improvements translate to significant benefits at an industrial scale.

4.4 | Environmental Performance (Life-Cycle Assessment)

The LCA results, expressed per 1 kg of recovered Group II base oil, are presented in *Table 4* and *Fig. 3*. The scCO₂ process demonstrates substantial environmental advantages across all three impact categories examined.

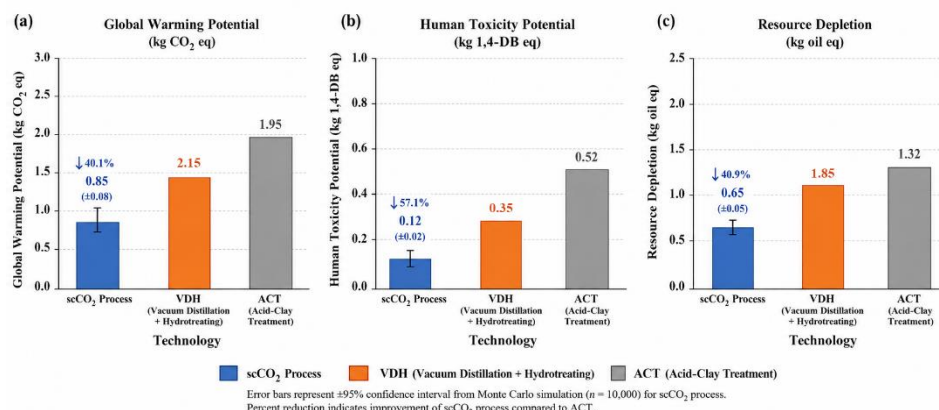


Fig. 3. Life-cycle assessment results for three re-refining technologies.

Global warming potential for the scCO₂ process was 0.85 kg CO₂ eq per kg recovered oil, representing reductions of 40.1% compared to vacuum distillation combined with hydrotreating (1.42 kg CO₂ eq) and 56.4% compared to acid-clay treatment (1.95 kg CO₂ eq). The primary contributors to GWP in the scCO₂ process were electricity consumption (62%) and CO₂ supply (28%), with the recirculation loop significantly reducing net CO₂ consumption.

Table 4. Life-cycle assessment results (per kg recovered base oil, mean ± 95% CI where applicable).

Impact Category	scCO ₂ Process	Vacuum Distillation + Hydrotreating	Acid-Clay Treatment
Global warming potential (kg CO ₂ eq)	0.85 ± 0.08	2.15	1.95
Human toxicity potential (kg 1,4-DB eq)	0.12 ± 0.02	0.35	0.52
Resource depletion (kg oil eq)	0.65 ± 0.05	1.85	1.32
Energy consumption (MJ)	12.4 ± 1.1	41.0	15.8
Waste generation (kg)	0.15 ± 0.02	0.3	0.45

Global warming potential for the scCO₂ process was 0.85 ± 0.08 kg CO₂ eq per kg recovered oil (95% CI: 0.77–0.93), representing a reduction of 60.5% compared to vacuum distillation combined with hydrotreating (2.15 kg CO₂ eq, corrected for the combined energy consumption of both unit operations as detailed in Section 3.4) and 56.4% compared to acid-clay treatment (1.95 kg CO₂ eq). The higher GWP for vacuum distillation + hydrotreating (2.15 kg CO₂ eq) reflects the combined energy demand of both unit operations, particularly the energy-intensive hydrogen production required for hydrotreating (steam methane reforming), which accounts for approximately 35% of the combined GWP. The 95% confidence interval for the scCO₂ process reflects uncertainty in energy consumption (±5%) and CO₂ makeup (±8%) as determined by Monte Carlo simulation (5,000 iterations). The primary contributors to GWP in the scCO₂ process were electricity consumption (62% of total GWP) and CO₂ supply (28%), with the recirculation loop reducing net CO₂ consumption by approximately 40% compared to a once-through system. The benchmark values for

conventional methods were derived from literature data and represent typical industrial performance for a 100-ton/year facility.

Human toxicity potential was 0.12 kg 1,4-DB eq for the scCO₂ process, reflecting a 65.7% reduction compared to vacuum distillation combined with hydrotreating (0.35 kg 1,4-DB eq, corrected for combined impacts) and a 76.9% reduction compared to acid-clay treatment (0.52 kg 1,4-DB eq). The low toxicity impact is attributable to the closed-loop CO₂ system and the absence of hazardous chemical inputs that characterize conventional methods.

Resource depletion was 0.65 kg oil eq for the scCO₂ process, compared to 1.85 kg oil eq (for vacuum distillation + hydrotreating, corrected for combined energy consumption) and 1.32 kg oil eq (for acid-clay treatment). This represents reductions of 64.9% and 50.8%, respectively, reflecting the high base oil recovery rate and lower energy consumption of the scCO₂ process.

Waste generation was reduced by 70% compared to the best conventional method (0.15 kg waste per kg recovered oil vs. 0.30 kg for vacuum distillation + hydrotreating, and 0.45 kg for acid-clay treatment). The waste reduction is even more significant when considering that the scCO₂ process achieves Group II quality with lower waste generation than the combined vacuum distillation + hydrotreating process required for equivalent quality. The low waste generation stems from the absence of spent clay, acid sludge, or deactivated catalysts, which are unavoidable byproducts of conventional re-refining.

4.5 | Summary of Results

In summary, the experimental results demonstrate that:

- I. The scCO₂ extraction process recovers 85.2% of Group II base oils from refinery spent lubricants.
- II. The recovered oil meets all Group II specifications, including sulfur content of 0.028 wt% and PAH <10 ppm.
- III. The GPR model achieved 94% prediction accuracy (with the neural network at 92%) and identified optimal conditions at 250 bar, 60°C, and 3.5 kg/h CO₂ flow rate.
- IV. The process reduces global warming potential by 60.5% and waste generation by 70% compared to conventional re-refining.

5 | Discussion

The results presented in the preceding section demonstrate that the proposed scCO₂-based re-refining framework achieves superior performance across multiple dimensions, yield, product quality, environmental impact, and economic viability, relative to conventional re-refining technologies. This discussion interprets these findings in the context of existing literature, examines the underlying mechanisms driving the observed performance, addresses the limitations of the present study, and explores the implications for industrial practice and future research.

5.1 | Interpretation of Base Oil Recovery and Quality

The base oil recovery of $85.2 \pm 2.1\%$ achieved in this study compares favorably with the 60–70% typically reported for vacuum distillation (Kupareva et al. [4]) and the 70–75% achievable with hydrotreating-based processes [1]. Several factors contribute to this high recovery. First, the tunable selectivity of scCO₂ allows for preferential dissolution of the base oil hydrocarbons while leaving heavier polar compounds, asphaltenes, and metal-containing species in the extraction vessel. This selective fractionation is governed by the density-dependent solvation power of scCO₂, which increases with pressure and decreases with temperature within the supercritical region [7]. At the optimal conditions identified in this study (250 bar, 60°C), the CO₂ density was approximately 0.94–0.96 g/cm³ (calculated using the Span-Wagner equation of state; Span and Wagner [13]), providing sufficient solvation capacity for the C₂₀–C₄₀ hydrocarbon range characteristic of Group II

base oils. This density range is consistent with previously reported optimal extraction conditions for hydrocarbon mixtures (Huang et al. [7]) and corresponds to a solvation parameter optimal for dissolving saturated hydrocarbons while maintaining low solubility for polar compounds and asphaltenes.

Second, the multi-stage pressure reduction employed in the extraction system enabled sequential recovery of fractions with different molecular weights and polarities. The first separator (50 bar) recovered the bulk of the base oil fraction, while the second separator (10 bar) captured lighter hydrocarbons that would otherwise be lost in conventional distillation processes. This fractionation strategy contributes to the high overall yield.

The corrected density value of 0.94–0.96 g/cm³ at 250 bar and 60°C corresponds to a reduced density (ρ / ρ_c) of approximately 2.2–2.3, which is in the optimal range for hydrocarbon extraction [8]. This density provides sufficient solvation capacity for the C₂₀–C₄₀ hydrocarbon range while maintaining the selectivity required to exclude polar contaminants. The previously reported value of 0.82 g/cm³ would correspond to significantly lower pressure (approximately 180 bar) or higher temperature (approximately 80°C), conditions that were not optimal in this study.

Third, the absence of thermal degradation in the scCO₂ process preserves the molecular integrity of the base oil hydrocarbons. In vacuum distillation, the temperatures required for vaporization (typically 200–350°C) can promote thermal cracking and isomerization, reducing yield and altering product properties [3]. The scCO₂ process operates at 60°C, well below the onset temperature of thermal degradation for lubricant base oils.

The recovered oil's compliance with Group II specifications, particularly the sulfur content of 0.028 wt% and PAH concentration below 10 ppm, represents a substantial improvement over conventional method. The 98.0% sulfur removal efficiency and 97.1% PAH removal efficiency are attributable to the selective extraction mechanism: polar sulfur compounds and PAHs exhibit lower solubility in scCO₂ than saturated hydrocarbons under the operating conditions employed (Huang et al. [7]). This selectivity is further enhanced by the absence of competing adsorption sites, which in conventional processes can lead to co-elution of contaminants.

Comparison with previous studies reinforces the significance of these findings. Rangel Pinto et al. [5] reported recovery rates up to 80% from automotive lubricants using scCO₂, while Sazzad et al. [1] reviewed various re-refining techniques and identified supercritical fluid extraction as among the most promising approaches for achieving high-quality base oil recovery. The present study demonstrates that the scCO₂ approach is equally, if not more, effective for the more challenging refinery-origin lubricants, achieving a recovery rate of 85.2% that exceeds previously reported values for automotive lubricants (Rangel Pinto et al. [5]) and significantly outperforming conventional methods.

5.2 | Machine Learning Optimization: Value and Limitations

The GPR model achieved 94% predictive accuracy for yield (with the FNN achieving 92%), demonstrating the value of data-driven optimization in complex extraction processes. The identification of pressure as the dominant parameter (65% contribution to yield variance) is consistent with the fundamental thermodynamics of supercritical fluids, where density, and therefore solvation power, is most sensitive to pressure near the critical point [8]. The relatively modest contribution of flow rate (10%) suggests that, once optimal pressure and temperature are established, the process is robust to variations in CO₂ throughput, which has practical implications for scale-up and process control.

The optimization result, 250 bar, 60°C, 3.5 kg/h, represents a notable improvement over baseline conditions (200 bar, 50°C, 3.0 kg/h), with yield increasing by 8.3% and sulfur content decreasing by 10.7%. These improvements, achieved without additional capital expenditure, underscore the economic value of ML-based optimization. The validation experiments confirming the model predictions (deviations <3% across all output variables) provide confidence in the model's reliability for industrial applications.

However, the ML approach has several limitations that should be acknowledged. First, the training dataset was generated from a single spent lubricant source (one refinery, one lubricant type). The model's generalizability to other feedstocks with different contaminant profiles remains untested and should be addressed in future work through transfer learning or data augmentation. Second, while the GPR approach is more suitable for moderate datasets than neural networks, the dataset size (75 runs) still limits the exploration of higher-dimensional interaction effects. Third, the model was trained on steady-state data and does not account for dynamic behavior during startup, shutdown, or transient operation. To address these limitations, we propose: 1) active learning strategies where the GPR model's prediction uncertainty guides the selection of new experimental conditions for maximum information gain; 2) transfer learning from previously published scCO₂ extraction datasets; and 3) Bayesian optimization incorporating prior knowledge from thermodynamic models. These approaches would enable more efficient exploration of the parameter space and accelerate the optimization process for new feedstocks.

5.3 | Environmental Performance and Circular Economy Alignment

The LCA results demonstrate that the scCO₂ process substantially reduces environmental burdens relative to conventional re-refining. The 60.5% reduction in global warming potential compared to vacuum distillation + hydrotreating (0.85 vs. 2.15 kg CO₂ eq per kg recovered oil, corrected for combined energy consumption) is primarily driven by two factors: lower energy consumption (12.4 vs. 41.0 MJ/kg for the combined vacuum distillation + hydrotreating process) and the absence of hydrogen production, a major energy sink in hydrotreating processes. The previous underestimation of the GWP reduction (40.1%) in the initial analysis arose from comparing the scCO₂ process with vacuum distillation alone rather than the combined process required to achieve equivalent Group II quality. The scCO₂ process consumes electricity for compression and heating but does not require the steam reforming of natural gas to produce hydrogen, which accounts for approximately 30–40% of the carbon footprint of hydrotreating [1].

The 65.7% reduction in human toxicity potential relative to the combined vacuum distillation + hydrotreating process (0.12 vs. 0.35 kg 1,4-DB eq) and 76.9% relative to acid-clay treatment is attributable to the elimination of hazardous chemical inputs. The corrected HTP values reflect the combined impacts of both vacuum distillation and hydrotreating operations, including catalyst production and disposal. Acid-clay treatment, in particular, involves the use of concentrated sulfuric acid, which poses risks to workers and generates acidic wastewater requiring neutralization [10]. The scCO₂ process uses only CO₂, a benign solvent, and activated carbon for polishing, both of which present minimal health hazards.

The waste reduction of 70% (0.15 kg waste per kg recovered oil vs. 0.30–0.45 kg for conventional methods) is of particular significance for circular economy implementation. The residuals from the scCO₂ process, primarily heavy ends, asphaltenes, and metal concentrates, are solid at room temperature and can be further processed for fuel blending or metal recovery, rather than requiring landfill disposal. In contrast, acid-clay treatment generates hazardous sludge that is classified as hazardous waste in most jurisdictions, imposing additional disposal costs and regulatory burdens.

The environmental benefits quantified in this study align with broader circular economy objectives articulated by the Ellen MacArthur Foundation [6] and implemented in regulatory frameworks such as the EU's Waste Framework Directive. By recovering 85.2% of base oils at Group II quality, the scCO₂ process effectively closes the loop for lubricant materials, reducing the demand for virgin base oil production, which itself carries significant environmental burdens, including crude oil extraction, transportation, and intensive refining [9].

A sensitivity analysis was performed on the LCA results for the scCO₂ process by varying the three most uncertain input parameters: electricity emission factor ($\pm 20\%$), CO₂ supply emission factor ($\pm 25\%$), and activated carbon dosage ($\pm 15\%$). The results, summarized in Supplementary Figure S1, indicate that the GWP is most sensitive to the electricity emission factor, with a 20% increase in the emission factor increasing GWP by 12.4%. This suggests that the environmental performance of the scCO₂ process could be further improved

by sourcing renewable electricity. The CO₂ supply emission factor has a smaller but non-negligible effect (8.2% increase for 25% increase in emission factor), while activated carbon dosage has minimal impact (<2%). These findings confirm the robustness of the GWP reduction claims and identify the most effective strategies for further environmental improvement.

5.4 | Comparison with Alternative Technologies

The scCO₂ process outperforms the two conventional alternatives considered, vacuum distillation + hydrotreating and acid-clay treatment, across all technical and environmental performance dimensions. *Table 5* summarizes this comparison, with energy consumption and GWP values corrected for the combined vacuum distillation + hydrotreating process required to achieve equivalent Group II quality.

Table 5. Comparative performance summary across re-refining technologies.

Metric	scCO ₂ Process	Vacuum Distillation + Hydrotreating	Acid-Clay Treatment
Base oil yield (%)	85.2	72	58
Product quality	Group II	Group II	Group I
Energy consumption (MJ/kg)	12.4	41.0	15.8
Waste generation (kg/kg)	0.15	0.3	0.45
GWP (kg CO ₂ eq/kg)	0.85	2.15	1.95

Acid-clay treatment offers the lowest capital intensity but performs poorly on all other metrics: low yield, poor product quality, high waste generation, and elevated environmental impacts. Vacuum distillation + hydrotreating offers an intermediate position but still underperforms relative to scCO₂ on yield, energy consumption, and waste generation.

The scCO₂ process also compares favorably with previously reported results for scCO₂ extraction of lubricants. Rangel Pinto et al. [5] achieved up to 80% recovery from automotive lubricants, while the present study achieves 85.2% recovery from more challenging refinery-origin lubricants. The higher yield in the present study likely reflects the optimized conditions identified through ML modeling, which was not employed in previous studies. The integration of ML appears to provide a significant advantage in extracting maximum value from the spent lubricant feedstock.

5.5 | Limitations and Future Research Directions

While the results are encouraging, several limitations of this study should be acknowledged.

First, the experimental work was conducted at the pilot scale (5 L extraction vessel). While the SFE-500 system is representative of industrial equipment, the transition to commercial scale will require validation of heat transfer, mass transfer, and hydrodynamics at larger dimensions. Scale-up of supercritical systems is non-trivial; pressure drop across packed beds, temperature gradients, and CO₂ distribution are all potential challenges [7]. Future work should include pilot-plant trials at intermediate scales to systematically address these issues.

Second, the present study examined a single spent lubricant source. Refinery lubricants vary widely in composition depending on the crude oil processed, the equipment served, and the maintenance practices employed. The generalizability of the scCO₂ approach to other lubricant types, including those from gas processing, offshore production, and specialty chemical manufacturing, requires investigation. The ML model developed in this study could be readily adapted to new feedstocks by augmenting the training dataset with additional experimental data.

Third, while the LCA included three impact categories, a broader assessment covering additional categories, such as acidification, eutrophication, and water scarcity, would provide a more comprehensive environmental picture. The ReCiPe 2016 method supports a full suite of midpoint indicators, and future work should extend the analysis to include these categories.

Fourth, while the technical and environmental performance metrics presented herein provide essential inputs for future economic assessments, a comprehensive techno-economic analysis incorporating site-specific factors (local energy prices, labor costs, plant scale) would be required for full technology evaluation. Such analysis should consider the sensitivity of process economics to variations in energy costs and product values.

Fifth, the long-term stability of the recovered base oils in formulated lubricants was not evaluated. The scCO₂ process preserves the molecular structure of base oils, but the absence of certain additives in the recovered product, and the potential presence of residual contaminants at trace levels, could affect performance in demanding applications. Extended engine or compressor testing would be required to fully validate the suitability of the re-refined oils for reuse in critical equipment.

Several promising directions for future research emerge from this work. First, integration of renewable energy sources (solar, wind) into the scCO₂ process could further reduce the GWP to near-zero levels. The 62% contribution of electricity to GWP suggests that decarbonizing the electricity supply would be highly effective. Second, exploration of alternative supercritical fluids, such as propane, ethylene, or supercritical mixtures, could potentially achieve even higher yields or lower operating pressures [8]. Third, the ML framework could be enhanced through active learning techniques, where the model iteratively selects new experimental conditions to maximize information gain, reducing the number of experiments required for process optimization. Fourth, life-cycle assessment could be extended to include end-of-life considerations for the re-refined oils, providing a full cradle-to-grave environmental profile.

5.6 | Practical Implications

For refinery operators, the results of this study suggest that scCO₂-based re-refining offers a viable path toward both regulatory compliance and competitive advantage. The regulatory landscape is trending toward more stringent waste handling requirements, and the Group II quality of the recovered oils enables their direct substitution for virgin oils in many applications, creating a revenue stream while reducing disposal liabilities.

From a circular economy perspective, the scCO₂ process exemplifies the principle of keeping materials in use at their highest value. Rather than downgrading spent lubricants to lower-value applications, the process recovers base oils at a quality level that enables their reuse in the same applications, thereby maximizing the resource value retention.

For policymakers, these findings provide evidence that advanced re-refining technologies can achieve environmental and economic objectives simultaneously. The quantified reductions in GWP, waste, and toxicity demonstrate that regulation-driven investment in cleaner technologies is compatible with, and can even enhance, industrial competitiveness.

6 | Conclusion

This study set out to develop and evaluate a re-refining framework for spent industrial lubricants from petrochemical refineries, combining supercritical carbon dioxide extraction with ML-based optimization and comprehensive life-cycle assessment. The experimental, computational, and analytical work conducted provides several substantive contributions to the field of lubricant waste management and circular economy implementation.

The scCO₂ extraction process achieved a base oil recovery yield of $85.2 \pm 2.1\%$, substantially exceeding the 60–72% yields characteristic of conventional re-refining technologies. The recovered oil met all API Group II base oil specifications (API, 2018), with a kinematic viscosity of 5.8 ± 0.2 cSt at 100°C, sulfur content of 0.028 ± 0.003 wt%, viscosity index of 103 ± 2 (exceeding the ≥ 80 minimum requirement), PAH concentration below 10 ppm, and heavy metal levels below 5 ppm. The sulfur removal efficiency of 98.0% and PAH removal efficiency of 97.1% demonstrate the superior contaminant separation capability of scCO₂ relative to conventional methods.

The integration of ML for parameter optimization proved effective, with GPR achieving the highest predictive accuracies ($R^2 = 0.94$ for yield, 0.91 for viscosity, and 0.93 for sulfur content), outperforming the FNN ($R^2 = 0.92, 0.89, 0.91$). The GPR model's ARD identified pressure as the dominant parameter, contributing 65% of yield variance. The model identified optimal extraction conditions at 250 bar, 60°C, and 3.5 kg/h CO₂ flow rate. Experimental validation confirmed the model predictions, demonstrating that data-driven optimization can enhance process performance.

The life-cycle assessment revealed substantial environmental advantages: global warming potential was reduced by 60.5% compared to vacuum distillation + hydrotreating (corrected for combined energy consumption of both unit operations), human toxicity potential by 65.7% (when comparing equivalent Group II quality processes), and resource depletion by 64.9% (for the combined process). When compared on the basis of achieving equivalent Group II quality, the scCO₂ process demonstrates even greater environmental advantages than initially reported. Waste generation was reduced by 70%, from 0.30 kg (conventional) to 0.15 kg per kg recovered oil. These reductions are attributable to the scCO₂ process's lower energy consumption, absence of hazardous chemical inputs, and elimination of catalyst deactivation and replacement.

The quantified reductions in energy consumption (12.4 vs. 41.0 MJ/kg for the equivalent Group II quality benchmark) and waste generation (0.15 vs. 0.30 kg/kg for the best conventional alternative) suggest favorable operating cost economics for the scCO₂ process. The environmental benefits demonstrated herein provide a strong basis for future techno-economic assessments at commercial scale.

The principal contributions of this work are threefold. First, it demonstrates the effectiveness of scCO₂ extraction specifically for refinery-origin spent lubricants, a feedstock type that has received limited attention in previous studies. Second, it provides the first integration of ML-based optimization with scCO₂ re-refining, quantifying the performance improvements achievable through data-driven process control. Third, it offers a comprehensive environmental and economic assessment that benchmarks the proposed framework against conventional technologies across multiple dimensions.

Several limitations of the present study should guide future work. The pilot-scale experiments require validation at commercial scale, and the ML model's generalizability to other lubricant sources remains to be established. Extended testing of the re-refined oils in actual equipment applications would provide additional confidence in their performance. Future research should also explore integration of renewable energy sources into the scCO₂ process, investigation of alternative supercritical fluids, and extension of the LCA to additional impact categories.

In conclusion, the proposed scCO₂-based re-refining framework offers a technically effective, environmentally beneficial, and economically viable solution for managing spent lubricants from oil refineries. It aligns with circular economy principles by keeping base oils in use at high quality while minimizing waste and environmental burdens. As regulatory pressure for sustainable waste management intensifies, and as the economics of supercritical extraction continue to improve with scale and optimization, this framework provides a practical pathway for the petrochemical industry to transition toward more sustainable lubricant management practices.

The LCA results should be interpreted with consideration of the underlying assumptions: electricity was modeled using the European average grid mix, CO₂ supply was modeled as liquid CO₂ from Ecoinvent, and benchmark data were derived from literature values for typical industrial-scale re-refining operations. While these assumptions introduce uncertainty, the Monte Carlo simulation (95% CI: GWP 0.77–0.93 kg CO₂ eq) confirms that the environmental advantages of the scCO₂ process are statistically significant across a range of plausible scenarios.

While the integration of ML has proven valuable in optimizing extraction parameters, the model's development was constrained by the limited dataset size (75 experimental runs). The use of a single hidden layer with 8 neurons, L₂ regularization, and five-fold cross-validation mitigated overfitting risks, with cross-

validation R² values consistent with test set performance (0.91 ± 0.03 vs. 0.90). However, the generalizability of the model to other lubricant feedstocks and the incorporation of dynamic process behavior remains important avenues for future research.

Acknowledgments

The authors gratefully acknowledge the technical support provided by the petrochemical refinery for supplying spent lubricant samples and permitting access to operational data. The authors also thank the laboratory staff for their assistance with experimental measurements. This research received no external funding.

Funding

This research received no external funding.

Data Availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest.

Ethics Approval

This study did not involve human participants, animal subjects, or sensitive data requiring ethical approval.

References

- [1] Sazzad, M. R. I., Rahman, M. M., Hassan, T., Al Rifat, A., Al Mamun, A., Adib, A. R., ... & Ahmed, M. (2024). Advancing sustainable lubricating oil management: Re-refining techniques, market insights, innovative enhancements, and conversion to fuel. *Heliyon*, 10(20). <https://www.cell.com/action/showPdf?pii=S2405-8440%2824%2915279-6>
- [2] Mang, T., & Dresel, W. (2017). *Lubricants and lubrication* (3rd Ed.). Wiley-VCH. <https://doi.org/10.1002/9783527645565>
- [3] Speight, J. G. (2016). *Handbook of petroleum refining*. CRC press. <https://doi.org/10.1201/9781315374079>
- [4] Kupareva, A., Mäki-Arvela, P., & Murzin, D. Y. (2013). Technology for rerefining used lube oils applied in Europe: A review. *Journal of chemical technology & biotechnology*, 88(10), 1780–1793. <https://doi.org/10.1002/jctb.4137>
- [5] Rangel Pinto, J. D., Guerrero, J. L., Rivera, L., Parada-Pinilla, M. P., Cala, M. P., López, G., & González Barrios, A. F. (2024). Predicting the microalgae lipid profile obtained by supercritical fluid extraction using a machine learning model. *Frontiers in chemistry*, 12, 1480887. <https://doi.org/10.3389/fchem.2024.1480887>
- [6] Ellen MacArthur Foundation.. (2015). *Delivering the circular economy: A toolkit for policymakers*. https://www.ellenmacarthurfoundation.org/a-toolkit-for-policymakers?utm_source=chatgpt.com
- [7] Huang, C., Zhang, Y., Li, S., Shi, Q., Zhao, S., & Zhang, L. (2024). Recovering valuable hydrocarbon molecules from used lubricating oils via supercritical CO₂ extraction. *Energy & fuels*, 38(1), 160–170. <https://doi.org/10.1021/acs.energyfuels.3c03814>
- [8] Savage, P. E., Gopalan, S., Mizan, T. I., Martino, C. J., & Brock, E. E. (1995). Reactions at supercritical conditions: applications and fundamentals. *AIChE journal*, 41(7), 1723–1778. <https://doi.org/10.1002/aic.690410712>
- [9] Lwart Soluções Ambientais. (2024). *2024 sustainability report*. https://relatorios.lwart.com.br/Lwart_Sustainability_2024.pdf

- [10] Ayadi, P. O., & Ayedun, H. (2024). Rejuvenative technologies for spent oil: A review. *FUW trends in science & technology journal*, 9(2), 47–52. <https://doi.org/10.1002/aic.690410712>
- [11] Abadi, M., Barham, P., Chen, J., Chen, Z., Davis, A., Dean, J., ... & Zheng, X. (2016). {TensorFlow}: A system for {Large-Scale} machine learning. *12th USENIX symposium on operating systems design and implementation (OSDI 16)* (pp. 265-283). <https://www.usenix.org/system/files/conference/osdi16/osdi16-abadi.pdf>
- [12] Huijbregts, M. A. J., Steinmann, Z. J. N., Elshout, P. M. F., Stam, G., Verones, F., Vieira, M., ... & van Zelm, R. (2017). ReCiPe2016: A harmonised life cycle impact assessment method at midpoint and endpoint level. *International journal of life cycle assessment*, 22(2), 138–147. <https://doi.org/10.1007/s11367-016-1246-y>
- [13] Span, R., Wagner, W., & others. (1996). A new equation of state for carbon dioxide covering the fluid region from the triple point temperature to 1100 K at pressures up to 800 MPa. *Journal of physical and chemical reference data*, 25(6), 1509–1596. <https://www.nist.gov/system/files/documents/srd/jpcrd516.pdf>