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Research article

Advancing gasoline desulfurization: Multi-objective fuzzy optimization in systems technology

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ABSTRACT

Ultrasonic-assisted oxidative desulfurization (UAOD) is utilized to lessen environmental problems due to sulfur emissions. The process uses immiscible polar solvents and ultrasonic waves to enhance desulfurization efficiency. Prior research focused on comparing the effectiveness of UAOD for gasoline using response surface methodology. This study evaluates the desulfurization efficiency and operating costs, including ultrasonic power, irradiation time, and oxidant amount to determine optimal conditions. The study used a multi-objective fuzzy optimization (MOFO) approach to evaluate the economic viability of UAOD for gasoline. It identified upper and lower boundaries and then optimized the desulfurization efficiency and operating costs while considering uncertainty errors. The fuzzy model employed max-min aggregation to optimize the degree of satisfaction on a scale from 0 (unsatisfied) to 1 (satisfied). Optimal conditions for gasoline UAOD were found at 445.43 W ultrasonic power, 4.74 min irradiation time, and 6.73 mL oxidant, resulting in a 66.79 % satisfaction level. This yielded a 78.64 % desulfurization efficiency (Y_A) at an operating cost of 13.49 USD/L. Compared to existing literature, gasoline desulfurization was less efficient and less costly. The solutions provided by MOFO demonstrate not only economic viability through decreased overall operating costs and simplified process conditions, but also offer valuable insights for optimizing prospective future industrial-scale UAOD processes.

1. Introduction

Majority of the energy consumed in the power and transportation sectors is produced by fossil fuels, which have had substantial influence on humanity and the environment. Fossil fuels, containing hydrocarbons, release a substantial amount of energy upon combustion. However, their combustion releases significant amounts of carbon dioxide and other pollutants, including sulfur dioxide, contributing greatly to global warming and climate change.

Petroleum products like gasoline are fundamental components of contemporary economies and are required for daily life, industry, and transportation. Gasoline is produced by a complex refining process from crude oil, and it is a crucial hydrocarbon. The composition of gasoline often includes sulfur, and the combustion of sulfur generates undesirable sulfur oxides, which also harm catalysts during chemical refining [1]. Refining crude oil into gasoline presents a significant challenge, particularly when dealing with heavy and extra-heavy oils, which make up approximately 70 % of the global oil resources. These oils possess distinct characteristics, including

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high viscosity, low API gravity, and high sulfur and asphaltene content. These properties render them unsuitable as refinery feedstocks for gasoline production without further upgrading or blending with conventional crude oils [2]. The presence of sulfur compounds in raw fuel oil contributes to major environmental problems that generate the combustion product of sulfur oxides (SOx), such as sulfur dioxide. The pollution generated by SOx also leads to acid deposition, adversely affecting man-made structures, vegetation, and water bodies. Regarding public health, exposure to SOx can result in various acute and chronic respiratory issues and contribute to cardiac or pulmonary conditions [3]. SOx can damage vehicular catalytic converters responsible for oxidizing carbon monoxide, nitrogen oxide, and volatile organic compounds. If left untreated, releasing SOx from fuel oil combustion exacerbates these issues, posing a significant threat to future generations and further deteriorating the environment. The process of diminishing the sulfur content in fuels is called desulfurization. This is crucial in fuel utilization because it allows for advanced emission controls and reduces the environmental impact of sulfur. Varied methods have been implemented to reduce the sulfur content in fuels through desulfurization such as hydrodesulfurization (HDS) [4], oxidative desulfurization (ODS) [5], extractive desulfurization [6], biodesulfurization [7], and desulfurization through alkylation, chlorinolysis, and supercritical water [8]. The significance of these procedures is that they provide a large portion of the sulfur used in the industry, enable the use of sulfur-free compounds in several catalytic processes, and reduce the emission of hazardous sulfur compounds [1].

HDS is the primary industry method for sulfur removal from hydrocarbon fuels. Catalysts containing molybdenum, tungsten, and nickel or cobalt promoters were first developed in Germany in the 1920s and 1930s [9,10] and are commonly known as "promoted Mo or W catalysts," which consist of a mixture of MoS_2 and Co_9S_8 supported on alumina (Al_2O_3) [9,11]. Polyoxometalate (POM) catalysts also demonstrate significant catalytic activity in oxidative desulfurization, with the ability to convert into peroxo-POM compounds that exhibit high activity in the presence of hydrogen peroxide (POMs include phosphomolybdic acid (PMo), phosphotungstic acid (PW), silicotungstic acid (SiW) and sodium phosphotungstate (NaPW) [12]. It is costly due to its high temperature and pressure requirements [13]. Ionic liquids (ILs) serve dual functions in ODS, acting as both solvents for sulfur compound removal from fuel and catalysts for their oxidation. The effectiveness of ODS is influenced by various factors including the quantities of IL and oxidant used, reaction parameters, and the recyclability of IL. ILs that are immiscible in water are especially efficient in this process due to their high polarity, enabling their repeated use without compromising the yield of desulfurization [14]. The surface area of an ionic liquid in water varies due to factors like concentration, specific ions present, and temperature and pressure. Generally, ionic liquids tend to create micelles or aggregates when introduced into aqueous solutions, thereby impacting their surface area [15]. ODS is a method that employs an oxidizing agent to eliminate sulfur, contrasting with the use of hydrogen in the HDS process. Common oxidants in ODS include oxygen from the air and hydrogen peroxide (H_2O_2), offering an alternative to using hydrogen gas generated in refineries, which incurs high capital and operational expenses [16]. ODS presents promising industrial applications for desulfurizing real fuels like gasoline, gas oil, and diesel. Choi et al. [5] have demonstrated its significant advantage over HDS as it can be carried out at lower temperatures and pressures without the need for hydrogen. The sulfur compounds that are most difficult to eliminate through HDS are the most reactive in ODS. Choi et al. [5] showcased the efficacy of ODS in desulfurizing real fuels, such as diesel, with efficiency rates reaching 96.6 %. Their findings underscore the versatility of ODS beyond gasoline, highlighting its potential application to a wide range of fuel types. ODS offers a promising alternative to traditional desulfurization methods, mitigating the need for costly catalysts, elevated pressure conditions, and a significant amount of hydrogen associated with HDS. Recent studies have explored novel catalysts and solvents to enhance ODS efficiency, with successful applications in real diesel fuels suggesting potential applicability to other fuel types [17]. Variations in oxidant performance can lead to undesired side reactions, impacting the quality of the desulfurized gasoline. The choice of solvent and catalyst plays a crucial role in extracting sulfur compounds while preserving desired fuel components [18].

Ultrasonic-assisted oxidative desulfurization (UAOD) is an ODS technique that uses ultrasonic waves that can increase heterogeneous reactions, boost the activity of oxidants, and hasten the breakdown of macromolecular molecules. The study of Lin et al. [19] suggests that ultrasonic treatment enhances desulfurization efficiency through ultrasonic waves, cavitation, and activated oxygen, aligning with cleaner gasoline production. It provides an economically feasible solution to combat rising fossil fuel consumption, lowering energy costs with lower operating temperatures and pressures compared to traditional methods like HDS [20]. UAOD primarily aims to reduce sulfur in gasoline, potentially affecting other properties. However, the study by Barilla et al. [21] reveals a negligible decrease in density post-desulfurization, with a 1.62 % difference. The concentration of cycloparaffins usually stays stable, while UAOD boosts paraffin content and reduces aromatics, possibly enhancing fuel characteristics. Additionally, the calorific value increases slightly by 0.55 %, while viscosity remains relatively stable, with a 3.51 % difference post-treatment Barilla et al. [21]. Previous studies have used response surface methodology (RSM) to optimize the objective functions of UAOD in gasoline by considering variables such as ultrasonic power, irradiation time, and oxidant amount [22]. RSM is a mathematical and statistical approach commonly employed for constructing empirical models. It seeks to establish relationships between responses and various input variables while minimizing the number of experiments [23]. However, it is important to note that RSM is limited to fitting data to a second-level polynomial model, which may not accommodate all systems with curvature [24].

Multi-objective fuzzy optimization (MOFO) is a methodology that combines multi-objective optimization with fuzzy set theory to handle uncertainty and vagueness in objective functions or constraints. In MOFO, the goal is to maximize all membership functions simultaneously, and the objectives and constraint functions are treated as modified constraints [25]. MOFO enables the inclusion of fuzzy constraints and objectives, allowing for the handling of imprecise information and decision-making in a more realistic way [26]. MOFO is an effective approach in optimizing UAOD process. The MOFO compound demonstrates adaptability across various desulfurization technologies that have differing goals. Nevertheless, this research places particular emphasis on the UAOD process as the central focus. Since MOFO is utilized, a generation of a Pareto front is performed to obtain the optimal solutions that simultaneously optimize multiple objectives such as the optimal amounts of ultrasonic power, irradiation time and oxidants for the desulfurization of gasoline. Since it is an optimization technique that has multiple objectives, the study uses MOFO to evaluate the efficiency of gasoline and the operating costs to address the limitations in terms of the economic feasibility of applying UAOD. The study applies a designing software called LINGO to model and generate the mathematical equations of the Pareto front generation and fuzzy optimization process of the variables included in the desulfurization of gasoline.

While the study by Zhou et al. [22] significantly advanced gasoline desulfurization through RSM by optimizing UAOD efficiency, it only focused on maximizing the desulfurization rate of the process. MOFO takes the economic aspect of the process into account, a critical consideration in real-world applications. Achieving the highest desulfurization efficiency often comes at a significant cost in real-world applications. The novelty of the study lies in its application of MOFO to achieve a fuzzy optimal solution that balances both operational cost and UAOD efficiency of gasoline desulfurization. Multiple objectives are simultaneously optimized unlike RSM which only focuses on the optimization of a single objective function. This optimization model is essential for practical applications where cost-effectiveness is important. RSM has traditionally been applied across experiments for uni-objective of experimental parameters without accounting for the total operating cost of the process conditions. Neglecting cost analysis creates a gap in knowledge and hinders real-world application. This study bridges this gap by incorporating a cost analysis. Cost minimization and process optimization are often conflicting objectives. The study employs MOFO to address this multi-objective optimization challenge, enabling the simultaneous optimization of both response variables. The MOFO approach incorporates operational cost as an additional objective alongside UAOD efficiency. A wide range of optimal solutions are found through MOFO which represent the best trade-off between desulfurization efficiency and cost. This is all accomplished within a defined level of cumulative uncertainty. The study focuses on optimizing multiple process parameters using the optimized objective function identified by Zhou et al. [22] using RSM. Ultrasonic power, irradiation duration, and oxidant amount are the variables that will be optimized using MOFO and Pareto identification will be employed to establish the trade-offs between the variables and operational costs. A balance is achieved between maximizing desulfurization efficiency and minimizing total operating costs within the multi-objective fuzzy optimization model. This study also addresses the inherent limitations of scaling up laboratory-scale processes and emphasizes the importance of developing a solution for optimized performance in future industrial applications. The variables to be optimized include ultrasonic power, duration of irradiation, and oxidant amount. Pareto identification is also applied to establish the boundary limits of the variables associated with operating costs to obtain the maximum desulfurization efficiency of gasoline and minimum total operating costs in the fuzzy optimization model. The study also recognizes practical constraints associated with scaling up the process and ensuring consistency in performance, particularly emphasizing the laboratory scale and its potential as a reference for future industrial applications. The objectives of this study are to integrate total operating costs into experimental parameter optimization to enhance the development of cost-effective gasoline refining. Additionally, the fuzzy optimal solution for operational costs and desulfurization efficiencies using MOFO was determined. This entails establishing correlations between ultrasonic power, irradiation time, and oxidant amount with desulfurization costs. Finally, the identification of the Pareto frontier for gasoline based on operating costs and optimizing parameters to maximize desulfurization efficiency while minimizing total operating costs within a fuzzy optimization model was investigated.

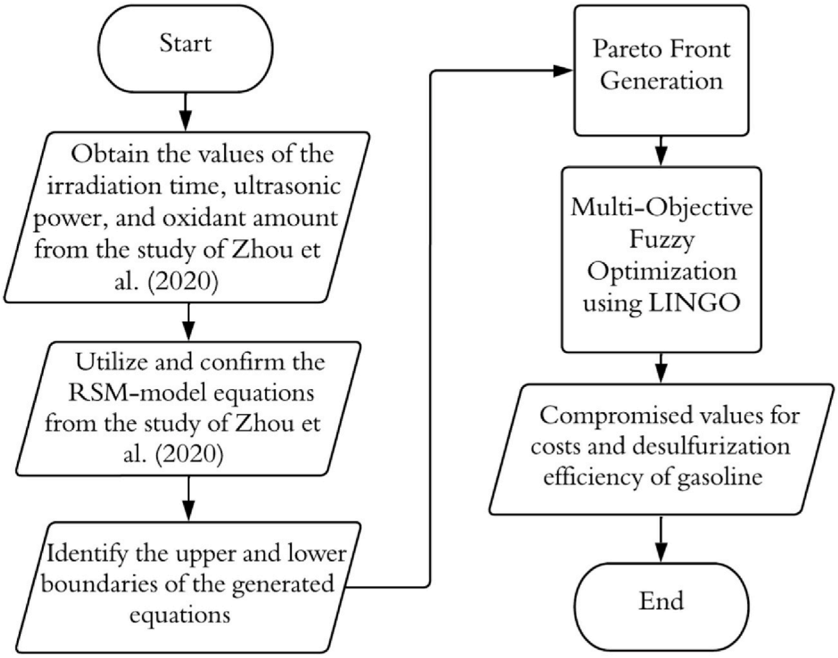

Fig. 1. Flowchart for multi-objective fuzzy optimization of UAOD.

2. Methodology

2.1. Methodological overview

The process of fuzzy optimization is illustrated in Fig. 1. The RSM equations employed in the MOFO framework were derived from controlled laboratory-scale experiments conducted by Zhou et al. [22]. The present study analyzed the efficiency and cost of UAOD by utilizing the results obtained from the study of Zhou et al. (2020) as a reference point with an initial concentration of 2.862 sulfur wt. %. The objective is to determine the compromise solution that simultaneously maximizes the desulfurization efficiency based on the removal of the initial concentration of gasoline while minimizing the total cost of electric and material consumption. The lower boundary is established by minimizing operating costs, while the upper boundary for sulfur removal is determined by maximizing the operating variables. The cost equation was determined to identify the global maximum of the fuzzy method, which is included in the objective of the study. The study is based on Zhou et al. [22] which enables the computation of the cost involved in the production of gasoline. The results were analyzed using appropriate optimization techniques to assess whether the Pareto front generation followed by fuzzy optimization offers a significant improvement in terms of performance and its associated cost in the UAOD. Generating the Pareto front is accomplished using the ε-constraint method. Among the range of desulfurization efficiency values, the operation expenses are minimized for the 10 equally divided parts, within which a graph is produced depicting the Pareto optimal solution. The MOFO method is subsequently employed in obtaining the optimal solution based on the initial research objective outlined by Zhou et al. [22]. Within the method of fuzzy optimization, a membership function is established to acquire the optimal solution by considering fuzzy limits.

2.2. Pareto front generation for the determination of boundary limits

The Pareto front generation using the MOFO approach was employed to establish boundary limits for Y_A represent the response variables of gasoline indicating the efficiency of sulfur compound removal and C_T represents the total operating cost of the desulfurization process, which includes the material and energy cost. Using the ε-constraint method, a single-objective is optimized while all the other objectives are transformed into constraints [27]. The objective functions of the desulfurization process are to maximize the Y_A based on response equation generated in RSM shown in **Eq. (1)** and minimize the C_T as seen at **Eq. (2)**. The data for the desulfurization efficiencies of gasoline versus its corresponding minimum total operating cost were then used to generate a Pareto curve that contains the Pareto optimal solutions.

The determination of the lower boundary limits for Y_A, and C_T is brought by the minimization of C_T in **Eq. (3)** whilst **Eqs. (4)–(9)** describes the constraints in which the objectives can be attained. **Eq. (4)** provides the sum Y_A by combining the Y_1 and the cumulative uncertainty error. **Eq. (5)** is the general response surface equation, wherein Y_1 is affected with respect to the operating variables: irradiation time, oxidant amount, and ultrasonic power. **Eq. (6)** illustrates the cumulative uncertainty of the response for the gasoline desulfurization. **Eq. (7)** solves for the total operating cost and **Eqs. (8) and (9)** guarantees the control variables and response to be in the feasibility range.

$$Y_A = 80.46 + 1.14X_1 - 1.27X_2 - 0.098X_3 - 1.00X_1X_2 - 1.35X_1X_3 - 1.65X_2X_3 - 3.43X_1^2 - 2.02X_2^2 - 4.18X_3^2 \tag{1}$$

Objective function:

$$\text{Maximize } Y_A \tag{2}$$

$$\text{Minimize } C_T \tag{3}$$

Constraints:

$$Y_A = Y_1 + W_{Y1} \tag{4}$$

$$Y = \beta_0 + \sum_i \beta_i X_i + \sum\sum_{i<j} \beta_{ij} X_i X_j + \sum_i \beta_{ii} X_i^2 \tag{5}$$

$$W_{Y1} = \left(\sum_{n=i}^{n} \frac{\partial Y_1}{\partial X_n} W_{X_n} \right)^{\frac{1}{2}} \tag{6}$$

$$C_T = \sum C_i \tag{7}$$

$$X_i' < X_i < X_i'' \tag{8}$$

$$Y \geq 0 \tag{9}$$

The variables in the formulated equations are the representation of Y_1 as the response of the gasoline desulfurization. $\beta_i, \beta_{ij}, and\ \beta_{ii}$ are the regression coefficients wherein i is the single factor index and j is the interacting factor index. The coded variables X_i and X_j and their bounds X_i' and X_i'' were introduced, along with the individual cost of the operating variables C_i. The -1 and 1 are the range that determines the low and high settings of the experiment.

2.3. Multi-objective fuzzy optimization for desulfurization variables

The identification of a compromise solution can be accomplished using MOFO. LINGO 20.0 is the software utilized to perform and solve the optimization model for this study [28]. MOFO can be attained by determining the fuzzy boundary limits derived from the generation of the Pareto front [29]. The objective is to convert the multi-objective function, which encompasses desulfurization efficiency and total operating costs, into a singular objective function. The linear membership function is obtained which characterizes the fuzzy goals. It linearly converts the input values to a scale ranging from 0 to 1, where the lowest input value is assigned a value of 0 and the highest input value is assigned a value of 1. A max-min aggregation was applied to maximize the satisfaction level of the linear membership function simultaneously (**Eq. (10)**). The linear membership functions (**Eqs. (11) and (12)**) are applicable in optimizing UAOD due to the trend that the objective function values are being linearly formed. In **Eq. (13)**, the upper and lower limits of the degree of satisfaction should vary between 0 and 1, making the expected outcome a fraction that represents partial satisfaction. When the quantified values of satisfaction levels meet the conditions of the objective function and fuzzy constraints, then the degrees of satisfaction of the uncertainty error and total variable costs are acquired.

$$max\ \lambda_o \tag{10}$$

$$\lambda_A = \frac{Y_A - 74.24}{6.58} \tag{11}$$

$$\lambda_C = \frac{15.08 - C_{T1}}{2.39} \tag{12}$$

$$0 \le \lambda_o, \lambda_A, \lambda_C \le 1 \tag{13}$$

λ_o is the overall degree of satisfaction, while λ_A and λ_C are the satisfaction levels for gasoline desulfurization and variable costs, respectively. The efficiency of gasoline desulfurization is expected to have an increasing trend approaching the highest possible rates, while the desired linearity of the operating costs is in a decreasing trend in which both objective functions are calculated simultaneously.

2.4. Case analysis of the study of Zhou et al. [22]

The analysis is derived from the data collected in a study conducted by Zhou et al. [22] regarding the UAOD in gasoline. The study focused on optimizing the UAOD process of gasoline to reduce sulfur content. The experimental methodology involved using actual fluid catalytic cracking (FCC) with specific sulfur contents, along with H_2O_2, formic acid, and ultrasonic energy for desulfurization. Various instruments such as an ultrasonic system, FTIR spectrometer, and ultraviolet fluorescent sulfur analyzer were used to assess alterations in functional groups and sulfur quantity in the samples. A control variable approach was implemented in a single-factor experiment to evaluate the ultrasonic conditions, varying parameters like ultrasonic power, irradiation time, and oxidant amount. Sulfur extraction and mass fraction determination were carried out using specific methods and instruments. A verification experiment under optimized conditions showed that UAOD was more efficient than oxidative desulfurization alone for reducing sulfur content. The specific operating variables examined in the study include ultrasonic power levels ranging from 100 W to 700 W, irradiation time ranging from 3 min to 11 min, and oxidant amount ranging from 5 mL to 10 mL, as reported by Zhou et al. [22]. Additionally, the simulation analysis incorporates an energy consumption cost of 0.185 USD/kWh and a material cost of 11.10 USD/L for H_2O_2, sourced from Zürich [30]. These data sets are crucial for the accurate integration of a comprehensive cost analysis, which is essential for attaining a compromise solution using MOFO to maximize the sulfur removal efficiency while minimizing expenses.

3. Results and discussion

3.1. Parametric analysis

The desulfurization process involves the utilization of the variables ultrasonic power, irradiation time, and the oxidant amount. The determination of optimal ultrasonic power levels (W), irradiation time (min), and oxidant amount (mL) while balancing desulfurization efficiency and operating costs, can be achieved through the application of multi-objective fuzzy optimization. The results for gasoline were obtained from LINGO 20.0 and MS Excel was used to present and examine the effects of adjusting the variables on both desulfurization efficiency and operational costs. The analysis focuses on the relationship between the three (3) variables and their impact on the desulfurization process, providing valuable insights for the optimization of UAOD.

3.2. Evaluating the effect of ultrasonic power on the desulfurization efficiency and operating costs of gasoline

Asakura and Yasuda [31] determined that at frequencies below 129 kHz, the strength of ultrasonic power rises as the effective electric power increases. The ultrasonic frequency used in the experiment was 19 kHz–21 kHz. Thus, it is expected for the electric consumption to increase with rising ultrasonic power. The response of ultrasonic power on the desulfurization efficiency and total cost of gasoline UAOD were determined by increasing the amount of ultrasonic power from 100 W to 700 W. At constant irradiation time and oxidant amount, the relationship between the ultrasonic power and the corresponding objective functions is displayed in Fig. 2a. The graph shows an observable increase in conversion as the power increases from 100 W to 400 W, with the maximum conversion of 80.46 %. Increasing the power from 400 W to 700 W caused a decline in the conversion for gasoline UAOD. The optimal ultrasonic power level for treating gasoline with UAOD is 400 W, and power levels that are too high or too low do does not produce the optimum results. High ultrasonic power can rapidly consume the available oxidants. Ultrasonic waves can generate significant cavitation that can produce reactive species that consume the oxidants in the system. The total efficiency of the oxidation process in UAOD may be constrained by the depletion of oxidants due to excessive ultrasonic power [32]. For the ultrasonic power-operating cost relationship, the total operating costs constantly increased from 13.40 USD/L to 16.58 USD/L. There is direct proportionality observed since increasing ultrasonic power results in higher energy input, thus increasing the total operating cost.

Fig. 2b exhibits the specific costs for the material used and the electric consumption as the ultrasonic power was increased. The material cost slightly decreased from 12.75 USD/L to 11.91 USD/L when the ultrasonic power increased from 100 W to 400 W, while it increased to 12.13 USD/L as the ultrasonic power continued to increase to 700 W. With increasing ultrasonic power, there is a visible trend with the conversion and the material cost. When the conversion was increasing to 400 W of power, the material cost decreased. Meanwhile, when the conversion was decreasing as the power increased to 700 W, the material cost increased due to the oxidant gasification loss from increasing the ultrasonic power more than the optimum, resulting to more oxidant amount to be consumed. The cost for electric consumption continuously increased from 0.65 USD/L to 4.36 USD/L at 100 W–700 W. Higher ultrasonic power levels require more energy to produce the needed vibrations and cavitation effects. The operational costs also tend to increase due to the higher electricity consumption caused by the increased energy demand. Additionally, the equipment required to produce and deliver higher ultrasonic power levels might need more electricity, which would result in higher energy costs and usage [33]. The increase in total operating costs observed in Fig. 2a is consistent with the breakdown of costs displayed in Fig. 2b. It displays that the total cost heavily depends on the electric consumption since changing the ultrasonic power mostly required energy for production. The residual plot in Fig. 2c indicates a good-fit with the model for ultrasonic power and sulfur conversion. This is shown by the residuals being randomly scattered around the zero line on the x-axis, where it varies from −1.72 % to 0.86 %. Notably, there is an absence of discernible patterns or distinct data points significantly deviating from zero, signifying that the model effectively captures the underlying data patterns without the presence of outliers that could potentially distort the model fit [34]. A three-dimensional (3D) surface plot depicting the effect of ultrasonic power on both cost and conversion efficiency is presented in Fig. 2d. This plot shows that

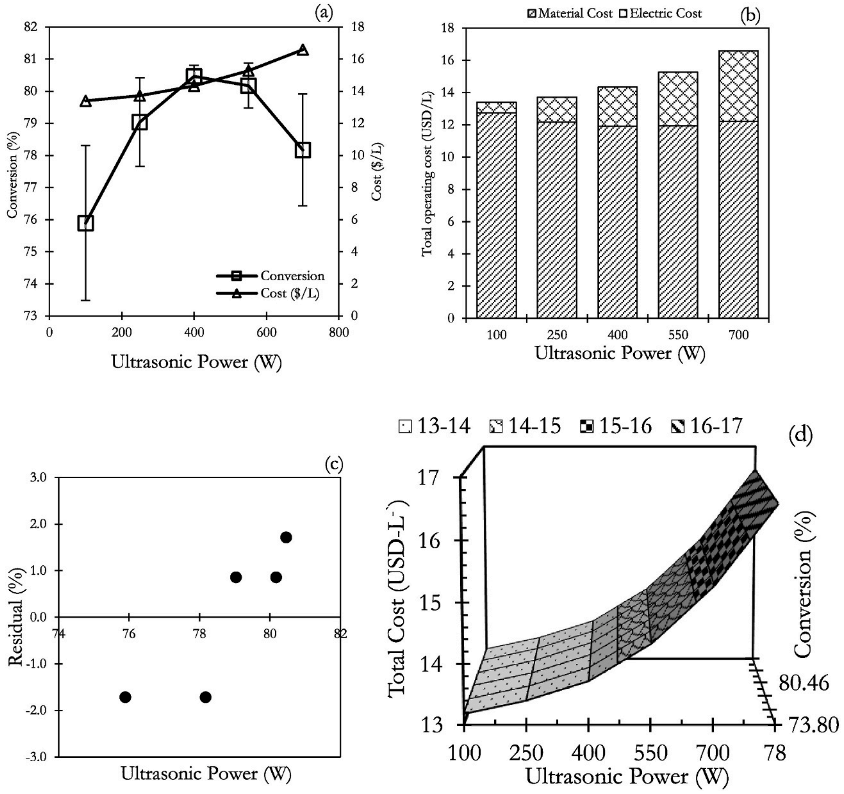

Fig. 2. Ultrasonic Power on Desulfurization Conversion and Gasoline Cost: (a) Ultrasonic Power vs. Objective Functions, (b) Ultrasonic Power vs. Operating Costs, (c) Residual Plot, and (d) 3D Surface Plot of Ultrasonic Power vs. Cost and Conversion.

both the cost and conversion efficiency increase with increasing ultrasonic power.

3.3. Evaluating the effect of irradiation time on the desulfurization efficiency and operating costs of gasoline

In the UAOD of gasoline, the value of irradiation time was set from 3 min to 11 min and the desulfurization efficiency at each condition was measured while keeping the other variables constant. The relationship of the irradiation time against conversion and cost is shown in Fig. 3a. The conversion increases from 3 min to 5 min, with values of 72.86 %–73.42 % respectively. This is due to the increase in the effective local concentration of the reacting species as the irradiation time increases which leads to a higher conversion [12]. The maximum desulfurization efficiency is found at the 5 min mark with a percent conversion of 73.42 %. Further increasing the irradiation time after 5 min resulted in a decrease in the conversion. This implies that all the oxidant has been used up which results in the termination of the decomposition reaction of the oxidant, leading to a stunted oxidation reaction [22]. Other factors that led to the decrease of conversion with increasing irradiation time are attributed to the instability of H_2O_2 in the UAOD system which is caused by insufficient catalyst dosage [19].

The analysis of the relationship of the total operating costs, which include the material and electric costs in accordance with irradiation time, is illustrated in Fig. 3b. An increase in the irradiation time from 3 min to 5 min showed a decrease in the material cost at the maximum conversion. The decrease in material cost per liter of sulfur removed may be attributed to the increasing trend of sulfur conversion from the 3 min–5 min, optimal irradiation time. The total operating costs at the maximum conversion (72.96 %) was 18.36 USD/L with the material cost and electric cost amounting up to 18.01 USD/L and 0.35 USD/L respectively. Further increasing the irradiation time led to increasing electricity costs. This is because of the increasing amount of power being consumed in the process. As the variables ultrasonic power and oxidant amount are held constant at 400 W and 8 mL, the amount of electricity being used in the process would only increase with increasing irradiation time, therefore leading to higher electric costs. The increase in the material cost in the process could also be attributed to the decreasing amount of sulfur converted as the irradiation time increases beyond the 5 min mark. Fig. 3c displays the residual plot for irradiation time with its corresponding desulfurization efficiency. Since the residuals are dispersed randomly around the zero line on the x-axis from −1.0 % to 0.51 %, then it indicates a good fit with the model. There are also no clear patterns or notable outliers, suggesting the model accurately captures the data without distortion. A three-dimensional (3D) surface plot that shows the effect of irradiation time on both cost and conversion efficiency is presented in Fig. 3d. The total cost and conversion both increase with increasing irradiation time.

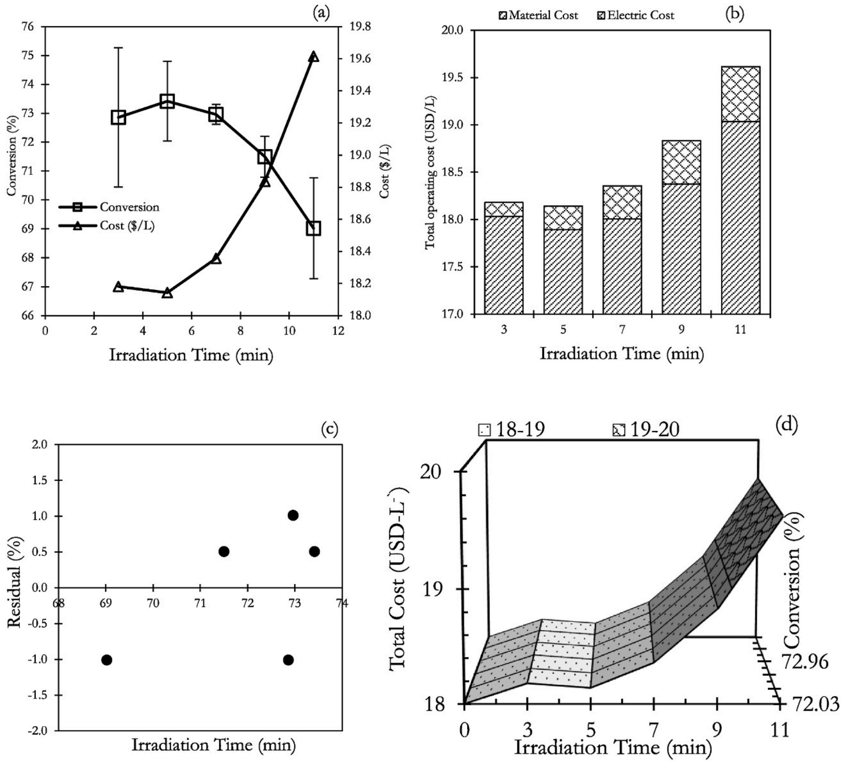

Fig. 3. Irradiation Time on Desulfurization Conversion and Gasoline Cost: (a) Irradiation Time vs. Objective Functions, (b) Irradiation Time vs. Operating Costs, (c) Residual Plot, and (d) 3D Surface Plot of Irradiation Time vs. Cost and Conversion.

3.4. Evaluating the effect of oxidant amount on the desulfurization efficiency and operating costs of gasoline

The effect of oxidant amount on the conversion of gasoline, as well as its associated cost, was investigated by varying the oxidant values from 5 to 10 mL, while keeping other variables constant. The goal of this study was to determine the optimal oxidant amount for achieving maximum conversion efficiency in the UAOD process. The conversion data obtained from the experiments are presented in Fig. 4a. It was observed that the conversion of gasoline increased from 74.04 % to 78.42 % when the oxidant level was set at 8 mL. However, further increasing the oxidant amount to 10 mL resulted in a decline in the conversion of gasoline. The findings indicate that there exists an optimal oxidant amount of 8 mL for the efficient treatment of gasoline using UAOD, beyond which excessive oxidant does not yield optimum results. In the study of Shakirullah et al. (2010) their research showcased the desulfurization capabilities of the formic acid and H_2O_2 combination, forming peroxyformic acid, which achieved a gasoline desulfurization rate of 57.66 %. This combination showed high selectivity in targeting sulfur compounds, enhancing overall desulfurization efficiency. These findings underscore the critical role of the oxidant agent in enhancing gasoline desulfurization efficiency. The observed findings can be attributed to several factors that influence the efficiency of the UAOD process at different oxidant amounts. At lower oxidant amounts, the reaction exhibits suboptimal progress, leading to incomplete conversion of sulfur-containing compounds in gasoline. The reaction rate is improved as the oxidant amount is increased to 8 mL, resulting in a higher conversion rate of sulfur compounds. However, when the oxidant amount is further increased to 10 mL, the excessive amount of oxidant leads to overoxidation [22]. This overoxidation can cause undesired side reactions, such as the degradation of the hydrocarbon matrix, resulting in a decline in the overall conversion efficiency.

Fig. 4b presents a valuable insight into the relationship between the oxidant amount and the total operating cost in the UAOD process. The material cost exhibits an increasing trend from 12.02 to 13.02 per USD/L of gasoline converted as the oxidant amount increases from 6 to 10 mL. This observation is expected since a higher oxidant amount leads to the consumption of a greater quantity of oxidants, consequently raising the material cost. The direct relationship between oxidant amount and material cost highlights the importance of optimizing the oxidant dosage to strike a balance between conversion efficiency and cost-effectiveness. The electricity cost remains constant at 1.08 USD/L of gasoline converted throughout the examined range of oxidant amounts. The constant electric cost can be attributed to the fixed variables of ultrasonic power (400 W) and irradiation time (7 min). By maintaining these parameters at a constant level, the energy consumption related to ultrasound irradiation remains unchanged, thereby ensuring a consistent contribution of electricity cost to the total operating cost. The increasing trend observed in the total operating cost in Fig. 4b is primarily driven by the material cost associated with the oxidant usage. As the oxidant amount is increased from 6 to 10 mL, the material

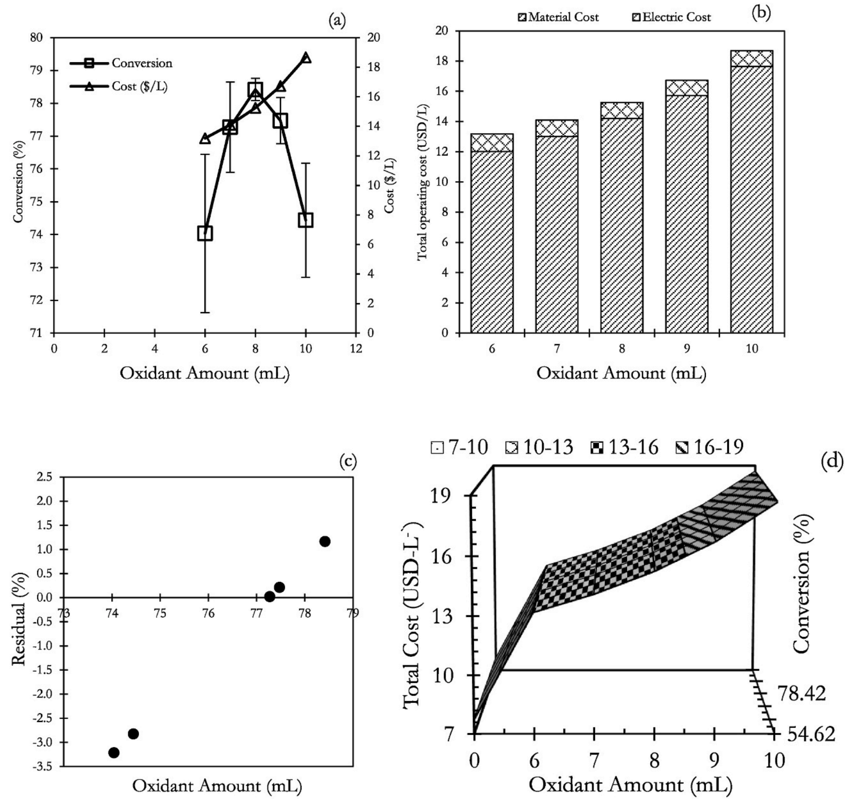

Fig. 4. Oxidant Amount on Desulfurization Conversion and Gasoline Cost: (a) Oxidant Amount vs. Objective Functions, (b) Oxidant Amount vs. Operating Costs, (c) Residual Plot, and (d) 3D Surface Plot of Oxidant Amount vs. Cost and Conversion.

cost escalates accordingly, resulting in a higher total operating cost. The constant electricity cost further reinforces this trend, as it remains unaffected by variations in the total operating cost. In Fig. 4c, the residual plot of oxidant amount against desulfurization efficiency shows random dispersion from −3.21 % to 1.16 %, indicating a good model fit. Moreover, absence of patterns or outliers suggests accurate data capture by the model without distortion. Fig. 3d presents a 3D surface plot depicting the effect of irradiation time on both cost and conversion efficiency. Conversion and cost generally increase with increasing oxidant amount.

3.5. Pareto front generation for the identification of the fuzzy limits of desulfurization efficiency and cost

Using the ε-constraint method, the C_T of gasoline was minimized. This was done by incrementally varying the sulfur conversion rate from a lower limit of 74.24 % up to the upper boundary limit of 80.92 %. By minimizing the C_T, the pre-determined points of Y_A and Y_B were identified to determine their respective minimized cost values. The global maximum Y_A of 80.92 % was attained and resulted in a C_T of 15.08 USD/L. The values of the operating variables were set to 464.77 W ultrasonic power, 5.48 min irradiation time, 8.06 mL oxidant amount. The minimum conversion of 74.24 % has a cumulative uncertainty error of 2.15 % and at a total operating cost of 12.69 USD/L. This condition was achieved with operating variables set at 424.88 W ultrasonic power, 3 min irradiation time, and 6 mL oxidant amount.

Fig. 5 illustrates the Pareto front for gasoline, which includes the boundary limits of Y_A, Y_B, C_T of the desulfurization process. The Pareto curve obtained in Fig. 5 serves as a critical criterion for selecting the optimal solution. The generated curve clearly illustrates that when conversion increases, the cost also increases. This relationship implies that achieving higher conversion necessitates the use of higher values of the operating variables such as ultrasonic power, irradiation time, and oxidant amount. Consequently, this will lead to an increased total cost. Increasing irradiation time and ultrasonic power during gasoline desulfurization enhances conversion but incurs additional electricity cost [22]. After a certain point, further increases in the operational variables result in increased costs while reducing the conversion rate. This study aims to maximize conversion while determining the most cost-effective option. Each point on this curve represents a Pareto front condition, indicating that any attempt to improve the performance of one variable would compromise that of the other.

3.6. Multi-objective fuzzy optimization

The objective of MOFO is to maximize the overall degree of satisfaction of the system, which is achieved by maximizing the objective function. The determination of the upper and lower boundary limits was based on the outcomes as included the generation of the Pareto front. The optimal boundary limits for the desulfurization efficiency and total costs for gasoline are shown in Fig. 6. The linear membership functions are shown in the figure and were used to maximize the desulfurization efficiency of gasoline while simultaneously minimizing the operational costs. The lower and upper limits for Y_A were found to be 74.24 % and 80.82 %. If the desulfurization efficiency falls below 74.24 %, it is considered unsatisfactory, while it becomes satisfactory if the desulfurization efficiency reaches 80.82 % or higher. Meanwhile, if the efficiency falls within the range of 74.24 %–80.82 %, it resides within the boundaries of the satisfaction threshold. In the case of maximizing Y_A, the upper limit for C_T was 15.08 USD/L, whereas the lower limit for minimizing C_T was determined to be 12.69 USD/L. The UAOD costs exceeding 15.08 USD/L and falling below 12.69 USD/L would yield satisfactory and unsatisfactory outcomes, respectively. Costs within the range of 12.69 USD/L to 15.08 USD/L would result in partial satisfaction. The overall degree of satisfaction obtained from the LINGO software is 0.6679, indicating partial satisfaction. At this satisfaction level, the desulfurization efficiency is 78.64 % at a cost of 13.49 USD/L, which falls within the range for partial satisfaction.

Table 1 shows the summary of the results of multi-objective fuzzy optimization for gasoline. This is done through the use of LINGO software. The results of fuzzy optimization for gasoline showed degrees of satisfaction of 0.6679 for λ_A, λ_C, and λ_{overall}. When the

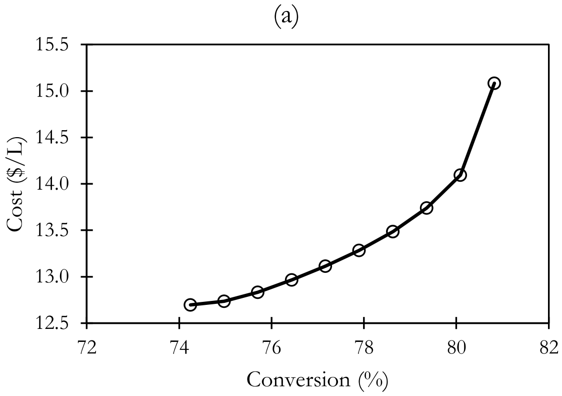

Fig. 5. Pareto front curve of the conversion of gasoline vs total operating cost.

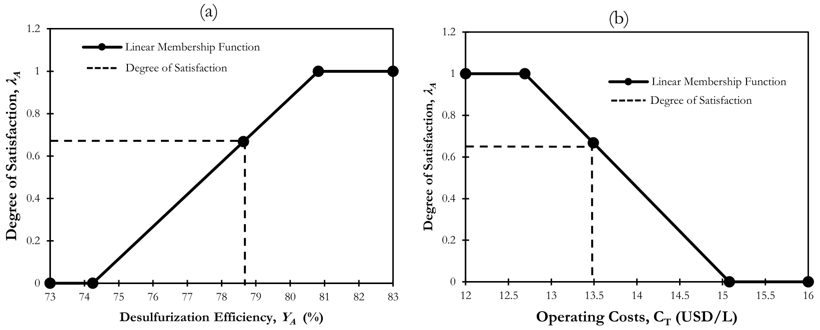

Fig. 6. Linear membership function for desulfurization efficiency and cost of gasoline.

Table 1
Fuzzy optimal solutions for the UAOD of gasoline.

Parameters	Feasible at Y_A
$\lambda_{overall}$	0.6679
λ_A	0.6679
λ_C	0.6679
Y	78.64 %
C_T	13.49 USD/L

degree of satisfaction is 0.6697, the fuzzy optimum conversion for the desulfurization efficiency of gasoline (Y_A) is 78.64 % which falls between the lower and upper boundaries: 74.24 % and 80.82 %. The corresponding total operating cost (C_T) is 13.49 USD/L of sulfur removed where the range for costs is from 12.69 USD/L to 15.08 USD/L. Both the desulfurization efficiency and operating costs for gasoline UAOD achieved partial satisfactory results as optimized using MOFO. The conditions associated with the optimum conversion and cost were an ultrasonic power of 445.43 W, an irradiation time of 4.74 min, and an oxidant amount of 6.73 mL.

The optimization process displayed the optimal values of ultrasonic power, irradiation time, and oxidant amount of gasoline desulfurization. High levels of ultrasonic power have the ability to rapidly deplete the oxidants present in a system. The generation of intense cavitation through ultrasonic waves leads to the production of reactive species that consume these oxidants. Consequently, the overall efficiency of the oxidation process in UAOD may be limited by the excessive ultrasonic power causing the depletion of oxidants [32]. In relation to the irradiation time, an increase in irradiation time results in a higher effective local concentration of the reacting species, thereby leading to an enhanced conversion. However, once all the oxidant has been consumed, the decomposition reaction of the oxidant ceases, resulting in a limited oxidation reaction. An excessive irradiation time can lead to a decrease in conversion due to the instability of H_2O_2 in the UAOD system, which is attributed to an inadequate dosage of catalyst [19]. Lastly, the oxidants can facilitate the conversion of aromatic sulfur compounds, which are typically difficult to remove. The oxidized sulfur compounds such as sulfones and sulfoxides become more polar which makes them easier to eliminate. However, excessive levels of oxidants can lead to over-oxidation of the oil sample, resulting in the destruction of its components and a decrease in the effectiveness of the desulfurization process. It is not advisable to continuously increase the operating variables without proper optimization, as this would also lead to increased costs [22]. The primary objective of the process should be to achieve the highest possible desulfurization efficiency while minimizing the associated operating costs.

Table 2
Comparison of RSM and fuzzy optimization solutions for the UAOD of gasoline.

Variables		RSM Optimization	Fuzzy Optimization	Blank Condition		
				1	2	3
Desulfurization Efficiency (%)	Y_A	80.82	78.64	55.00	72.03	73.80
Total Cost (USD L^{-1})	C_{T1}	15.08	13.49	13.18	18.00	7.72
Ultrasonic Power (W)	z_1	465.05	445.43	0	4.74	6.73
Irradiation Time (min)	z_2	5.48	4.74	445.43	0	6.73
Oxidant Amount (mL)	z_3	8.06	6.73	445.43	4.74	0

3.7. Summary and comparison of results

The upper and lower limits needed for multi objective fuzzy optimization were determined by conducting a single objective optimization of cost and desulfurization efficiency. The upper and lower limits found were then used in multi-objective fuzzy optimization. Linear membership functions were then used to obtain the maximum degrees of satisfaction under the objectives of obtaining the maximum desulfurization efficiency and the minimum cost for the UAOD of gasoline. In multi-objective fuzzy optimization, the upper and lower limits were assigned degrees of satisfaction of 0 and 1, respectively. The upper limit is referred to as the desired condition while the lower limit is referred to as the undesired condition.

The values of the cost and desulfurization efficiency of the UAOD of gasoline for the upper limit were 80.82 % and 15.08 USD/L. The conditions associated with the optimum values of the RSM optimization of gasoline are 465.05 W ultrasonic power, 5.48 min irradiation time, 8.06 mL oxidant amount. The overall satisfaction for the fuzzy optimization results of the UAOD of gasoline is 66.79 %. The costs for the optimum conditions obtained through RSM optimization were calculated through Pareto front generation.

Table 2 shows a summary of the result of MOFO in the UAOD of gasoline. The fuzzy optimal results correspond to a total cost of 13.49 USD/L with a desulfurization efficiency of 78.64 %. The conditions associated with the fuzzy optimal results of gasoline are 445.43 W ultrasonic power, 4.74 min irradiation time, 6.73 mL oxidant amount. The cost obtained through RSM optimization was initially 15.08 USD/L and was reduced by 1.59 USD/L through fuzzy optimization, resulting in a cost reduction of 10.54 %. This is because all of the process conditions at the fuzzy optimum are observed to have decreased when compared to the process conditions derived from RSM optimization. The calculated irradiation time at the fuzzy optimum (4.74 min) was 13.50 % faster than the calculated irradiation time (5.48 min) using RSM. The values of ultrasonic power and oxidant amount at the fuzzy optimum also decreased when compared to RSM conditions, resulting in reductions of 4.22 % and 16.50 %, respectively. The results reach a good compromise solution with an overall satisfaction of 66.79 % as the desulfurization efficiency at the fuzzy optimum resulted only in a moderate decrease of 2.70 % when compared to RSM conditions. The yield is acceptable given the relatively large reduction in the cost and process conditions at the fuzzy optimum.

Table 2 also depicts that the blank conditions serve as significant reference points for both RSM and Fuzzy Optimization analyses. Specifically, the first, second, and third columns represent the blank conditions related to ultrasonic power, irradiation time, and oxidant amount, respectively. These conditions establish fundamental values that serve as a basis for evaluating the optimization methods, offering essential benchmarks for assessing the effectiveness of the respective optimization strategies. It is important to note that the presence of zero values for a variable lead to lower costs and blank conditions. In contrast, fuzzy optimization demonstrates superior desulfurization efficiency, underscoring its effectiveness in the optimization process.

The fuzzy optimal solutions for the UAOD of gasoline are preferable when compared to their RSM optimal solution counterparts due to the relatively large decrease in the cost and process conditions with a minimal reduction in efficiency. The conditions required for operation in the fuzzy optimum conditions correspond to cost reductions of 10.57 %. This comes with a trade-off that amounts to minimal reductions of the desulfurization efficiency by 2.18 %. The effectiveness and sustainability of the process conditions at the fuzzy optimum are justifiable due to the relatively large cost reduction and reduced process requirements in the fuzzy optimum.

Table 3 presents a comparison of studies that utilize various desulfurization processes. UAOD operates under less demanding process conditions when compared to other desulfurization methods. For real-world, non-synthetic fuels, UAOD is a viable option considering its efficiency (78.64 %) when compared to both mixing-assisted oxidative desulfurization (62.37 %) and activated carbon-based ODS (55.10 %). PW and H_2O_2 are favorable choices compared to other catalyst and oxidant systems listed in Table 3 such as the carbon nanotube and molecular oxygen system or the iron-molybdenum and cerium (IV) oxide system. This is because PW and H_2O_2 are relatively cheaper, easier to handle, and are readily available than other catalysts and oxidants. The production of other catalysts and oxidants requires synthesis from precursor chemicals. Additional chemical synthesis results in higher costs and causes an increase in the total operating cost of the desulfurization process. Other desulfurization techniques in Table 3 such as adsorption desulfurization, carbon nanotube catalyst ODS, and photocatalytic ODS have relatively high efficiencies. These are caused by synthetic model fuel which only contains dibenzothiophene (DBT) derivatives with minimal impurities. Other sulfur compounds present in actual fuel oil are absent in synthetic model fuel. Thus, there is no competition between different sulfur compounds, thereby increasing the

Table 3
Summary and comparison of various desulfurization techniques.

Desulfurization Technology	Catalyst	Oxidant	Efficiency	Type of Fuel Oil	Cost	Reference
Ultrasonic-assisted oxidative desulfurization	Phosphotungstic acid	Hydrogen peroxide	78.64 %	Gasoline	13.49 USD/L	This study
Mixing assisted oxidative desulfurization	Phosphotungstic acid	Hydrogen peroxide	62.37 %	Diesel	–	Barilla et al. (2022) [21]
Activated carbon ODS	Phosphotungstic acid	Hydrogen peroxide	55.10 %	Diesel	–	Barilla et al. (2023) [35]
Adsorption desulfurization	Sulfuric acid	Bentonite clay	97.22 %	Synthetic Model Fuel (DBT)	–	Ullah et al. (2021) [36]
Carbon nanotube catalyst ODS	Carbon nanotubes	Molecular oxygen	100 %	Synthetic Model Fuel (DBT)	–	Zhang et al. (2013) [37]
Photocatalytic ODS	Iron-molybdenum nanocatalyst	Cerium (IV) oxide	100 %	Synthetic Model Fuel (DBT)	–	Beshtar et al. (2024) [38]

desulfurization efficiency. It is also important to note that cost analyses are not included in the studies on other desulfurization methods, resulting in a potential gap in past literature.

4. Conclusion

The study demonstrates the effectiveness of MOFO in optimizing the UAOD of gasoline, focusing on maximizing desulfurization efficiency while minimizing costs. MOFO successfully identified optimal operating conditions by focusing on achieving the highest desulfurization efficiency at the lowest possible cost, resulting in a 10.54 % reduction in total operating cost compared to the RSM optimal solution. Despite a moderate decrease in desulfurization efficiency, the solutions provided by MOFO are deemed acceptable due to their cost-effectiveness and less demanding process conditions. Moreover, the study highlights the potential for further improvements and applications, suggesting upscaling the process, incorporating fuzzy optimization, and expanding the range of variables studied. Additionally, incorporating total operating cost alongside experimental parameter optimization bridges the gap between traditional research and industrial practicalities, paving the way for widespread implementation of UAOD as a cost-effective method for sulfur removal in gasoline refining. Further research is suggested to expound the function of ILs in aqueous solutions within the context of MOFO. Empirical investigations should aim to determine whether ILs predominantly act as extraction agents or catalysts. Utilizing computational modeling and comparative analyses can offer significant understanding of IL properties. Through thorough examinations, the optimization of MOFO for industrial purposes can be improved.

CRediT authorship contribution statement

Stephen S. Correa: Writing – review & editing, Writing – original draft, Software, Investigation. **Kate Andre T. Alviar:** Writing – review & editing, Writing – original draft, Visualization, Investigation. **Angel Nicole V. Arbilo:** Writing – review & editing, Writing – original draft, Methodology, Investigation. **Angelo Earvin Sy Choi:** Visualization, Supervision, Software, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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