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Patent analysis in Biodiesel Research: A Scope for Mathematical Methods

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Abstract- Biodiesel, a renewable fuel derived from both edible and non-edible sources, poses a potential challenge to traditional diesel. This research examines biodiesel-related patents from 2003 to 2018, sourced from the International Patent Database. The study comprises five sections: biodiesel overview, feedstock-based generation, catalyst development, recent production advances, and reactor technology. 2nd gen biodiesel, derived from trash due to its cost-effectiveness, garners attention. Large-scale biodiesel production utilizes reactor technology, with the continuous stirred tank reactor deemed a simple and viable option. Various parameters impact biodiesel yield, engine performance, and emission characteristics. Mathematical tools like Design of Experiments, Artificial Neural Networks, and Metaheuristic algorithms aid in experimentation, analysis, predictions, and optimization. A robust mathematical framework could enhance biodiesel research, ensuring cost-effectiveness, sustainability, and quality.

Keywords- Biodiesel, production method, reactor technology.

I. INTRODUCTION

Paragraph The widespread usage of fossil fuels is the main cause of global climate change, which presents a serious threat to the entire planet. The primary source of global warming is the combustion of fossil fuels, which results in the atmospheric release of carbon dioxide (CO2) [1]. The International Energy Agency reports that energy consumption has increased consistently, with oil demand rising from 3,232,737 kiloton in 1990 to 4,449,499 kiloton in 2017. In addition to creating environmental issues, this increasing demand is using the limited supply of non-renewable fossil fuels, which is expected to run out by 2050 [2-4].

Biofuels show promise as a replacement for fossil fuels in order to meet the world's energy demands and address these urgent challenges. Biofuels are liquid or gaseous fuels used in the transportation industry that are created by biological and chemical processes or that are obtained from biomass such as microalgae, plants, and bacteria [3]. A popular biofuel in Europe, biodiesel is made by chemically transforming fats or oils from plant and animal sources into esters that have characteristics similar to those of mineral diesel. In terms of biodiesel production, the United States is the leader with 6.9 billion liters [5] produced, while Indonesia is the top Asian nation with 4.0 billion liters produced in 2018, placing it third internationally.es content here.

II. GENERATION OF BIODIESEL

Based on the feedstock used—edible, non-edible, or waste oil—biodiesel is classified into distinct generations. This categorization is based on the feedstock's primary sources as well as the attributes of the final product. Refined vegetable oils, such as canola and soybean oil, are frequently used as feedstock in the United States. Recycled cooking oil and animal fats are also utilized to lower production costs [5]. The first generation of biodiesel manufacturing is covered by patents, with the fourth generation still in the early stages of development.

For the manufacturing of biodiesel, the first generation uses food oils like soybean and rapeseed. While palm and coconut oil are preferred in Asian nations like Malaysia and Indonesia, rapeseed and soybean oil are popular in the U.S. and the E.U. Variations in feedstock have an effect on production methods, costs, and quality since the kind of fatty acids bonded to triglyceride molecules determine the type of biodiesel that is generated [6]. In order to expedite and streamline the purification process, Saka and Kusdiana (2001) suggested transesterifying rapeseed oil in supercritical methanol without the need for a catalyst [7]. Second-generation biodiesel, which uses waste oil and non-edible oils instead of edible oils, was developed as a result of the rivalry for edible oils, which drives up production prices. This kind lowers production costs and deals with the rivalry in the food supply [8].

The growth of second-generation biodiesel still requires land, which puts food and fiber production in competition, restricts the availability of nutrients, and lowers oil quality [9]. Algae-based biodiesel, which is the third generation, aims to get over earlier restrictions. It provides reduced greenhouse gas emissions, increased production and growth, less difficulty with land use, and higher oil outputs. The necessity for large-scale production, substantial sunshine requirements, increased production costs, and challenging oil extraction are some of the obstacles.

III. METHOD PRODUCING BIODIESEL

The current method for production of glycerin and biodiesel from feedstock includes pre-treatment, FFA esterification, and triglyceride transesterification. Biodiesel feedstock is separated from contaminants such as sulfur, phosphorus, phosphatides, gums, sterols, metals, and other color bodies during the pre-treatment process [10]. The esterification process produces water and biodiesel when the FFA component of biodiesel feedstock mixes with methanol and a catalyst. Transesterification is a process that produces both biodiesel and glycerine simultaneously by reacting triglycerides in the feedstock for biodiesel with methanol under the help of a catalyst [10]. In this context, other techniques for producing biodiesel will also be covered, such as pyrolysis and the direct usage and blending of oils.

1. Pre-Treatment Process

A technique described in U.S. Patent [11] permits the use of low-quality feedstock as a source of free fatty acids (FFA) during the pre-treatment stage in order to produce high-quality biodiesel. Filtration and distillation are the two steps in this process; the feedstock is heated to 43°C before being filtered to create filtrate. In order to acquire pure feedstock for the synthesis of biodiesel, it is imperative that contaminants be removed through repetitive, successive distillation. Before using feedstock for the manufacturing of biodiesel, this pre-treatment is necessary to remove any contaminants, especially when using waste such leftover cooking oil, brown greases, and crude maize oil. By providing a more workable and affordable option for biodiesel production, these waste materials meet the demand for affordable biodiesel that can rival diesel derived from petroleum. Nevertheless, the pre-treatment procedure is required due to the high FFA level in these wastes [12].

2. Direct Use and Blending of Oils

Because of their qualities in freezing temperatures, it is impractical to use vegetable oils directly as biodiesel; therefore, blending with biodiesel is a typical technique. The percentages of biodiesel blended with other fuels, including B20 (6%–20%) and B5 (5%) differ by nation. Four blending concepts are introduced by U.S. Patent No. 20180223202: actual biodiesel content measurement, target biodiesel content provision, automated processes with distillation, and separation [13].

A technique of heating biodiesel and diesel over the cloud point to create a second blend of biodiesel is proposed in U.S. Patent No. 7458998. This mixture is kept in a hot setting appropriate for fuel use [14]. It is necessary to guarantee adherence to diesel regulations concerning kinematic viscosity, density, and flashpoint. In a 2017 study, Arabi et al. examined a combination of palm oil, biodiesel, and diesel and discovered no appreciable variations in the fuel's characteristics up to 30% biodiesel volume [15].

3. Pyrolysis

Pyrolysis is a thermochemical process that produces solid (biochar), gaseous (biogas), and liquid (bio-oil) products at high temperatures (280°C–850°C) without the need for an oxidizing agent [16]. Because transesterification produces biodiesel with a high oxygen content, there is a risk of corrosion. On the other hand, by removing oxygen from the process and producing hydrocarbon diesel, biodiesel produced through pyrolysis offers a solution [17]. U.S. Patent Application 20070144060 describes a process for thermally cracking or rapidly pyrolyzing feedstock triglycerides as a pretreatment. By using this method, pollutants in biodiesel are efficiently removed, resulting in a distillate fraction that has a high free fatty acid (FFA) content [18]. In Figure 1, the general pyrolysis reaction is shown.

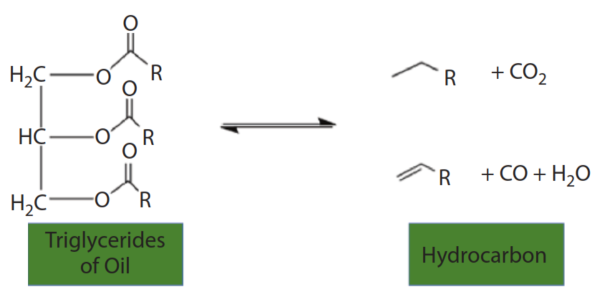

Figure 1: General reaction for pyrolysis

Table 1: Comparison of product yield and operational parameters for the pyrolysis technique [19].

Pyrolysis method	Temperature (°C)	Residence time (s)	Heating rate (°C/s)	Product yield (%)		
				Oil	Char	Gas
Slow	Medium-High (400–500)	Long (450–550)	Low (10)	30	35	35
Fast	Medium-High (400–650)	Short (0.5–10)	High (100)	50	20	30
Flash	High (700–1,000)	Very short (<0.5 s)	Very high (>500)	75	12	13

The simultaneous maintenance of two essential reaction conditions—temperature and a non-reactive atmosphere—makes the pyrolysis method known as a complicated process. During this process, biomass's long chains of carbon, hydrogen, and oxygen components break down into smaller molecules. There are three primary forms of pyrolysis: flash, fast, and slow. Of these techniques, quick and flash pyrolysis are thought to be the best for producing biodiesel because they can produce large yields—50% and 75%, respectively—at high reaction temperatures in a relatively short amount of time—as Table 1 [19] shows. Takuya (2012) states that at 390°C, triglycerides in oils break down into fatty acid chains and then into hydrocarbon chains. The yield of the oil product can be improved by raising the reaction temperature and heating rate [20].

IV. REACTOR'S TECHNOLOGY FOR BIODIESEL PRODUCTION

Batch reactors, semi-continuous flow reactors, and continuous flow reactors are common biodiesel production reactors that are essential to the large-scale, profitably producing biodiesel industry. A biodiesel reactor with four components is described in U.S. Patent No. 20080282606: an inlet for the inflow of raw materials, an outlet for the reaction mixture, a baffle for chamber segmentation, and a stir bar for internal stirring. Choosing feedstock oil, calculating the amount of alcohol and catalyst needed, combining these in the reactor to create a mixture, removing products and byproducts, and distilling the mixture to purify the biodiesel product are all steps in the production process of biodiesel [21]. Different types of reactors are now used to improve the production of biodiesel; this subject is covered in more detail later in this talk.

1. Continuous Stirred Tank Reactor

The same basic processing mechanism used in batch reactors is also used in Continuous Stirred Tank Reactors (CTSRs), which lower production costs by enabling continuous biodiesel production in a single step. CTSR consists of two crucial parts: the reactor and phase separator. The first step produces biodiesel from alcohol and triglycerides; the second stage removes the glycerol. The phase separator in the second stage enhances transesterification through chemical equilibrium shifting, resulting in a high biodiesel yield, approximately 97.3% [22]. CTSR, widely used on an industrial scale, offers simplicity and deep operational understanding.

A modified CTSR is used in U.S. Patent No. 2005052103 to propose an improved biodiesel preparation technique [23]. The feedstock oil is introduced into the modified reactor along with a feeding funnel, condenser, thermometer, and alcohol recovery/recycle system. After letting the mixture rest for four hours, the CTSR forms two layers, with the top layer undergoing additional processing [23].

Fixed Bed Reactor

As shown in Figure 2, a Fixed Bed Reactor (FBR) converts biodiesel by forcing oil and solvent through a cylindrical tube containing catalyst pellets organized in a static bed. For simple recovery, FBR uses a heterogeneous catalyst, which removes the requirement for catalyst and product separation. By guaranteeing gradual deactivation and extended lifespan, this kind of catalyst improves the reaction and eventually lowers manufacturing costs. However, the existence of residual glycerol (a by-product) at the bottom of the reactor requires a larger molar ratio, which affects catalyst effectiveness and calls for extra removal processes [24].

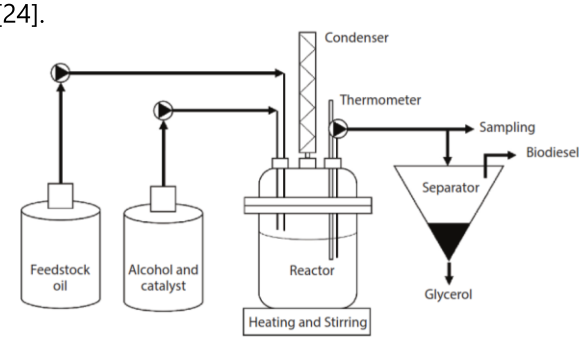

Figure 2: Graphical representation of a single reactor CTRS. Reproduced with permission from [22].

A hydro-processing technique for an acidic biomass feedstock in a guard bed to prevent unwanted polymerization is introduced in U.S. Patent No. 2011/0245551A1. Low temperature operation of the guard bed reactor saturates olefins with hydrogen, preventing olefins and other chemicals from polymerizing. The product can be recycled back into the reactor by using a noble metal or non-metal catalyst [25].

Micro-Mixer Reactor

Micro-mixer reactors, sometimes referred to as micro-reactors or micro-structured reactors, are widely employed in the manufacturing of biodiesel. These reactors, which have many millimeter-range channels, allow for improved oil separation by having high surface-to-volume ratios and short mass transfer distances between heat and diffusion. A novel approach to the generation of biodiesel is presented in U.S. Patent No. 8,404,005, which uses an enhanced catalytic transesterification process inside the micro-channel of the micro reactor. The first and second reactants are dispersed in this process, creating a laminar slug flow pattern that promotes immiscibility, effective mixing, and the start of the reaction that yields glycerol and biodiesel [26].

V. DEVELOPMENT OF CATALYST

In order to generate mesoporous calcium, barium, and magnesium silicate—which are used as catalysts in the production of biodiesel—Lin et al. [27] employed a co-condensation approach. A co-precipitation approach was used to manufacture a CaO-ZnO mixed metal oxide catalyst in a European Patent EP 2 522 713 A2 [28]. First, two aqueous solutions were made. Deionized water was used to dissolve the initial 1 M aqueous metallic cation solution of $Ca(NO_3)_2 \cdot 4H_2O$ and $Zn(NO_3)_2 . 6H_2O$. Both metal nitrate salt amounts were set so that the finished product's Ca to Zn ratios may be equivalent to 0.25:1, 2:1, 4:1, 6:1, 8:1, and 10:1. The process for producing biodiesel from plant oil in one step, with concomitant glycerol conversion into its derivatives, is suggested in EP2862915B1 [29]. Methyl acetate is employed as a reagent, while sodium methoxide solution in methanol is utilized as a catalyst. It was suggested in EP3674384 [30] to use tert-butoxide 1M solution as a catalyst in tetrahydrofuran (BuOK/THF) or tert-butanol (BuOK/BuOH). This catalyst prevents the transesterification reaction with the catalyst solvent and the production of glycerol during the transesterification process. The two-step procedure used to create biodiesel from waste oil or low-grade feedstock is detailed in a US Patent [31]. The two-step method consists of a pre-treatment stage wherein FFAs are removed using an acidic catalyst (such as HCl or H2SO4), and a second step whereby transesterification is catalyzed by an alkaline catalyst (such as KOH or NaOH). The US Patent US007906665B2 [32] presents a new catalyst system for the economical and environmentally beneficial manufacture of biodiesel. By using kiln dust, such as animal fats and vegetable oils, the catalysts facilitate effective esterification and transesterification reactions. Examples of these dusts are cement kiln dust (CKD) and lime kiln dust (LKD). The reusable nature of the catalyst systems offers a sustainable approach to the production of biodiesel.

VI. TRANSESTERIFICATION

The invention described in an international patent [33] relates to a transesterification reaction that produces biodiesel when vegetable oil and/or animal fat feed is atomized before the reaction. The method works well for producing biodiesel continuously. a process for transesterifying triglycerides using methanol, particularly those that contain free fatty acids. A catalyst made from an acidic ion exchange resin is used in this process. Under transesterification-friendly conditions, a reaction mixture comprising triglyceride and methanol comes into contact with the catalyst [34].
Stern et al. [35] describes a method for producing a fatty acid ester composition. One limitation of this invention is that a basic transesterification step needs to come after an acid transesterification step. Holmberg et al. [36] reveal a method for transesterifying triglycerides. One limitation of this invention is that it requires the presence of the enzyme lipase. Bam et al. [37] reveal a purification technique for alcohol esters. One limitation of this invention is that triglycerides must be used as the starting material. Furthermore, glycerin recycling occurs during the process, but it does so after the transesterification reactor. Assmann et al. [38] reveal a continuous process for reduced alkyl ester synthesis. One limitation of this innovation is that it requires the transesterification process to be done in a tube reactor in at least two phases. Furthermore, according to the reference, "no glycerol (reaction product) come into contact with starting oil is particularly important to the transesterification reaction" (col. 3, lines 4-7).

A method for creating fatty acid esters of short-chain monohydric alcohols is disclosed by Wimmer [39]. This invention is limited in that the fatty acid ester that is created is reconstituted with the glycerin that was previously split off during the transesterification process.

Ergun et al. [40] describes a process for making fatty acid methyl ester. The requirement for crack emulsification of the transesterification reactor's contents limits the use of this technology. A continuous transesterification technique is disclosed by Fleisher [41]. One limitation of this invention is that the transesterification reaction necessitates the use of a plug-flow reactor.

VII. MICROWAVE ASSISTED BIO-DIESEL PRODUCTION

The Chinese invention [42] is related to the utilization of microwave energy for ester exchange. The procedure involves several stages: positioning the reactor within a microwave generator; maintaining the temperature between 45 and 85 oC; applying heat reflux for a duration of 2 to 10 minutes; then cooling and performing centrifugal separation to acquire the crude biological diesel oil. This is followed by a cleaning process involving water washing, drying, and finally, continuous vacuum distillation at temperatures ranging from 220 to 250 oC to produce the refined biological diesel oil. All of these steps must be done in accordance with quantity relative ratios. The technique's benefits include low energy usage, quick reaction times, safe and dependable operation, and great manufacturing efficiency.

Studies conducted in a batch reactor at atmospheric pressure revealed that the yield barely climbed above 20%. The trials were expanded to include continuous flow under different circumstances, but the yield remained below 35%. Other researchers also obtained nearly comparable results [43]. The reason for the decreased yield was probably TBA's preferential dehydration to IB, a highly volatile molecule that was easily able to escape from the reaction zone.

Tokyo Electric Co. has also applied for two patents [43-44]. utilizing microwave irradiation for ETBE manufacture. Nevertheless, the stated maximum conversion—roughly 28%—at air circumstances is excessively low. Our most recent study, which used the same methodologies, produced findings that were comparable. On the other hand, improved conversion close to 90% was achieved by applying a continuous microwave power and letting the reaction temperature in a sealed vessel reach solvo thermal conditions. In a similar vein, several Japanese researchers have attempted to produce biodiesel using solid and microwave catalysts, as well as a method and apparatus for the process [45-46]. Mahlia et al. [47] analyze a total of 1,660 patents in biodiesel production from 1999 to 2018, with a focus on advancements in feedstock materials (nonfood crop, oil bearing crop grown in nonarable land, and reduced greenhouse gas emission), pre-treatment (avoid corrosion of reactors), catalysts (speedup the reaction rate and cost-effective), reactors, and testing methods (ASTM or EN). In their patent analysis the transesterification process is cost-effective method towards biodiesel production. Faba et al. [48] conducted a patent survey on catalyst role in biodiesel conversion and high lightened the importance of optimization of catalyst for different feedstock. In addition, highlighted the importance of solid catalysts in chemical processes, such as easier separation post-reaction, the potential for reuse, and compatibility with optimally designed chemical reactors. These benefits contribute to the sustainability of the processes by reducing both economic costs and environmental impacts. However, recent literature concluded that optimization of parameters (power, reaction time, temperature, catalyst type and concentration, and alcohol-to-oil ratio) of microwave-assisted transesterification is crucial in converting oils into high yield biodiesel [49]. Emphasis on use of nano catalyst and their concentration for biodiesel conversion for better recycling and reusability need intense research with a focus on optimization [50, 51].

VIII. OPTIMIZATION STUDY OF PROCESS PARAMETERS

In order to extract oil from an oleaginous organism, a solvent that does so must be mixed with a portion of the organism's culture in order to form a solvent-organism mixture. After that, this combination is run through a partitioning chamber to produce an extracted aqueous fraction that has an extractable organism that is viable as well as a solvent-oil fraction. Ultimately, the viable organism that was recovered is recycled into a system for cultivating [52]. One way to increase lipid production in an alga species is to provide an oleaginous alga and feed it a growth medium containing an effective amount of glycerol. This increases the oleaginous alga's lipid production compared to a corresponding oleaginous alga feeding on a growth medium without glycerol [53]. A second-order polynomial regression equation, incorporating three independent variables, models the yield of methyl ester as biodiesel fuel from these variables. The equation can be defined as follows [54].

$$X_{me} = b_0 + \sum_{t=1} b_t x_t + \sum_{t=1} b_{tt} x_t^2 + \sum_{t\neq u=1} b_{tu} x_t$$

On rare occasions, certain kinds of wax oil may exhibit poor flow properties in cold weather. In the context of biodiesel, Infineum Corporation's European patent [55] is significant for introducing additives that improve the flow properties of high-wax-content oils. These additives are esters, made from a C10 to C40 aliphatic monocarboxylic acid and an alkoxylated aliphatic monohydric alcohol with over C18 atoms before alkoxylation. The alkoxylation degree is defined as five to thirty moles of alkylene oxide per mole of alcohol.

The US patent [56] tackles this challenge by either esterifying a specific blend of fatty acids or fractionating the methyl esters derived from fatty acid mixtures obtained from palm oil, palm kernel oil, or their derivatives. Remarkably, transesterification wasn't necessary. By fat splitting, fatty acids from the previously described oil and oil products were utilized as the starting 18:1 point. The esterification was carried out using standard acid catalysis.

The patent notes that methyl oleate and methyl linoleate have pour points of -18ºC and 39ºC, respectively. Thus, biodiesel enhanced with these esters will exhibit better pour point properties. For instance, a blend with C 14 (0.5%), C 16 (5%), C 18:1 18 (83.6%), and C 18 (11%) achieves a pour point of -21ºC.

The patent offers a rather thorough explanation of how fractional crystallization is used to separate specific fatty acid methyl or ethyl esters. Additionally, the patent contrasts the various fuel quality of Malaysian palm diesel, biodiesel composed of C18, C18:1, and C methyl esters, and diesel. The finished biofuel has a perfect pour point of -15ºC yet the same viscosity as biodiesel. Costs associated with fractionation may pose an issue for economic viability Fatty acids in certain oils possess hydroxyl groups and most oils contain unsaturated bond fatty acids like oleic acid, raising major concerns about the oxidative stability of their alkyl esters.

This patent [57] states that these alkyl esters can be combined with pour depressant, petrochemical motor gasoline, and other fuel additive requirements to provide a fuel with enhanced oxidative stability, higher oxygen content, and superior low-temperature properties. The recommended blend compositions involve mixing methyl esters of hydroxyl-rich fatty acids and oleic acid-based fatty acids in ratios ranging from 10:90 to 90:10, along with about 90% diesel and essential additives.

A procedure for producing biofuel that entails the following steps: The process of storing oils, fats, and carbohydrates in the cells of photosynthetic microorganisms involves culturing the microorganisms in a culture solution, converting the stored carbohydrates into oils and fats in the microorganisms' cells, extracting the oils and fats from the microorganisms' cells, and reforming the extracted oils and fats [58-59].

IX. PARAMETER OPTIMIZATION BIODIESEL PRODUCTION

The process of turning crude oil into biodiesel became more difficult when the wide range of fatty acid and free fatty acid compositions in the crude oil were taken into account. The conversion was further complicated by the range of parameter settings and unpredictable catalytic systems. As a result, before the synthesis of biodiesel could be commercialized, a great deal of laboratory work and scaling were needed. Numerous parameters for the esterification and transesterification processes required to be tuned in order to attain a high conversion rate. Using an acid catalyst to facilitate esterification, the free fatty acid level was first lowered to less than 1% in order to get a high methyl ester yield. A triglyceride was transformed into three methyl ester by base-catalyzed transesterification, whereas a new ester was produced from free fatty acids during the esterification process [60-61]

Homogeneous catalysts such H2SO4, HCl, NaOH, and KOH were needed for the esterification and transesterification processes. For methyl ester conversion, alkaline homogeneous catalysts are preferred because they typically speed up the process and are more cost-effective; however, an excess of alkaline catalyst might produce saponification, which lowers methyl ester conversion [62-63]. C. Manghas oil's strong acidity exceeded the safe threshold for a direct transesterification reaction with an alkaline catalyst. Therefore, in order to obtain a high amount of methyl ester from this oil, both esterification and transesterification operations were needed. Researchers [64-66] performed numerous parameter improvements of biodiesel products utilizing esterification and transesterification. Process optimization has been done in a variety of ways, often using mathematical models [67-68]. In order to acquire the ideal methyl ester, this study employed the mathematical connection between process parameters that was created using the Box–Behnken design of experiments. Design of experiments based on response surface methodology was adopted to optimize the microwave-assisted transesterification parameters (catalyst, flow rate, methanol, power, reaction time, power, methanol-to-oil molar ration and so on) for high yield biodiesel conversion from waste cotton seed [69], waste cooking oil [70], Jatropha curcas [71], camelina oil [72], Argemone Mexicana oil [73], waste cat fish [74], chinaberry seed oil [75], and so on. The advantage of DOE and RSM limit the practical experiments (which reduces cost and energy saving) and derive mathematical model by establishing relationship between input-output. However, the major drawback of DOE and RSM model is analysis and prediction carried out one response at a time. This model fails to capture the detailed insights of dependencies among the outputs. In addition, online monitoring or multiple output control with inputs cannot be fully solved with DOE and RSM. However, taking into account the parameter range constraints, the Artificial Intelligence based on ANN model can be employed for prediction and optimization for biodiesel production [76-78].

1. Artificial Neural Networks and Metaheuristic algorithm-based Optimization

Artificial neural networks, or ANNs, are one technique used to model the process parameters for the manufacture of biodiesel. The process parameters for producing biodiesel and bioethanol have been widely modeled using artificial neural networks (ANNs), which minimizes the trial and error involved in traditional experiments. In this work, ANN and ACO were combined to optimize the relevant parameters and generate high methyl ester yields. ANN, sometimes referred to as a "black box modeling method," draws its inspiration from the biological nervous system of humans. Since many people thought—and then demonstrated—that artificial neural networks (ANN) could answer queries about science and engineering, ANN—which are just a class of non-linear computer algorithms—have grown in popularity [79-80]. Researchers have reported on the optimization of biodiesel production by the application of ANN modeling. Sivamani et al., 2018 [81]. used an artificial neural network genetic algorithm to optimize the production of biodiesel from Simarouba glauca. They discovered that the mean square error (MSE) of 0.00458, which was close to the MSE's acceptable range, was the value acquired from the ANNs. The biodiesel made from leftover groundnut oil was optimized by Ayoola et al., 2019 [82] using artificial neural networks (ANN). Regression coefficients (R) of 0.9241 and correlation coefficients (R2) of 0.8539 were discovered. Therefore, it was concluded that the ANN could accurately predict biological systems. A huge number of data processing components, referred to as nodes or neurons, make up an ANN model. These neurons are linked to one other and grouped in layers. The beauty of ANN is that it can construct a correlation without requiring the user to know in advance how the data processing pieces relate to one another [83-84]. Neurons and their connections with one another modify the input data at each stage of the process to produce an output [78]. However, ANNs trapped at local minima while solving multi-objective cost functions [85]. In addition, time-consuming to compute the gradient information with numerous iterations. Meta-heuristic (MH) algorithms overcome the said limitations that could optimize neural network parameters (weights, hidden neurons and their respective layers, learning rate, momentum constant, bias values) that accurately predict the outputs [86, 87]. In addition, MA optimize the convergence trend, exploration, exploitation, and avoid local minima [85]. There are more than 500 MA has been developed and among them 350+ algorithms were developed in the last decade [88]. These algorithms were classified into majorly four categories and are described in Table 1. MH algorithms trained ANNs showed better prediction accuracy for different applications (classification, prediction, optimization, system identification, monitoring process) of diverse engineering domains (mechanical, civil, chemical, medical and so on) [85]. MH algorithms predict several stages (fuel properties, biodiesel yield, performance and emission characteristics) of biodiesel production techniques and optimize parameters that could minimize the cost [89]. In addition, the real time process control and monitoring that could improve the process efficiency in biodiesel production that ensure enhanced performance in terms of efficiency, economic, and sustainability [90]. Therefore, MH algorithms has greater potential to apply for better prediction, and optimization.

ANN possess better generalizability to solve complex nonlinear real-world problems, and hybridizing with MH algorithms led extended benefits in solving optimization problems in biodiesel research [90]. ANN-GA-RSM is applied to enhance the performance and emission characteristics in propylene glycol and biodiesel–diesel blends [91]. ACO can solve complex process parameters, combining it with ANN is a clever approach [92-93]. Therefore, in order to take advantage of mathematical models' advantages in optimizing the process parameters for both esterification and transesterification for C. manghas biodiesel, the ANN-ACO strategy was applied in this work. ANN combined with Salp Swarm algorithm to optimize the engine parameters for better performance and emission characteristics in linseed oil biodiesel with graphene nanoparticles [94]. The success of above research work led ANN, and MH algorithms can be applied for biodiesel research for enhanced performance in terms of efficiency, economic, and sustainability.

2. Optimization Methods in Biodiesel Research

Optimization methods involving experimentation, prediction, and optimization, such as ANN, DOE-RSM, and MH algorithms, are crucial in solving complex real-world problems. Table 2 present the details of biodiesel research conducted by distinguished literature.

In Biodiesel research the mathematical modelling and optimization tools widely used are DOE, ANN and MH algorithms. DOE is a systematic method to determine the relationship between factors (transesterification, and engine) affecting output (yield, performance and emission characteristics) of a process. The advantages of DOE are [106, 107]: a) reduced experimental trials resulted in cost-effective technique, b) helps in systematically conducting experiments to explore the effect of several variables simultaneously, and c) analyze individual and factors interaction between variables resulting in detailed process insights. The limitations of DOE are [106]: a) complexity in designing experimental plan and understanding the process, b) conclusions drawn are valid only within the range of experimental conditions. c) analyze and establish relationship between factors with individual output at once. ANNs are computational mathematical models inspired by the human brain [108]. They consist of interconnected groups of artificial neurons and are used to approximate functions that depend on a large number of inputs [109].

ANNs are capable of modelling the complex nonlinear relationships, learn from data and improve over time, and predict the unseen data after extensive training [110-112]. In addition, predict multiple outputs and inputs simultaneously could help to capture interdependencies among output patterns and real-time monitoring of process [113, 114]. Need of huge training data sets, black box model due to difficulty in understanding and interpreting how the decisions or predictions are made, and training is computationally intensive are the limitations of ANNs [115, 116]. MH algorithms are problem-independent algorithmic frameworks that offer a set of strategies to progress heuristic optimization algorithms [117].

Table 2. Metaheuristic algorithms

Metaheuristic algorithms [85, 88, 95]			
Evolutionary (inspired by biological behaviour)	**Human (human behaviour in communities and human cooperation)**	**Physics (physics law in real life)**	**Swarm (animal's behaviour in movement and hunting groups)**
Genetic algorithm	Election Based Optimization Algorithm	Big Bang-Big Crunch	Particle swarm optimization
Differential evolution	Driving Training-Based Optimization	Gravitational search algorithm	Cuckoo search
Lightning search algorithm	Growth Optimizer	Black hole	Grey wolf optimization
Memetic Algorithm	Teacher learner-based optimization	Multi-Verse Optimizer	Symbiotic Organisms Search
Differential Search Algorithm	Queuing search	Sine Cosine Algorithm	Elephant Search Algorithm
Bull Optimization Algorithm	human behavior-based optimization	Gradient-based optimizer	Butterfly Optimization Algorithm
Evolutionary Strategies	Passing Vehicle Search	Intelligent Water Drops	Artificial Bee Colony
Evolutionary Programming	Forensic-Based Investigation	Ray Optimization	Bat Algorithm
Biogeography-Based Optimizer	Soccer League Optimization	Colliding Bodies Optimization	Whale Optimization Algorithm
Stochastic Fractal Search	World Cup Optimization	Kinetic Gas Molecule Optimization	Firefly Algorithm
Scatter search	Most Valuable Player Algorithm	Electromagnetic field optimization	Symbiotic Organisms Search
Lightning search algorithm	Volleyball Premier League	Heat transfer search	Dolphin echolocation
Golden Tortoise Beetle Optimizer	Team game algorithm	Atom search optimization	Grasshopper Optimization Algorithm
	Dice Game Optimizer	Spring Search Algorithm	Butterfly Optimization Algorithm
	Shell Game Optimization	Momentum search algorithm	Sandpiper Optimization Algorithm
	Squid Game Optimizer Algorithm	Solar System Algorithm	Sailfish Optimizer
	Ring Toss Game-Based Optimization	Energy Valley Optimizer	Golden eagle optimizer

MH algorithms can be applied to wide range of practical problems, and ability to overcome the local optima and determine global maxima or minima solutions [85]. MH algorithm solutions are approximations rather than exact solutions, performance of algorithms are highly sensitive to algorithm parameters, and time-consuming approach to solve complex problems [85, 118-121]. Applying DOE is essential for experimentation and they derive mathematical expression useful for prediction [122, 123]. ANNs predicts multiple outputs and inputs which cannot be done by DOE method, and can be implemented to a process for real-time process monitoring [115, 121]. MH algorithms optimizes the ANNs parameters to enhance the prediction and optimization capability of any process [121].

Table 3. Summary of Experimental, prediction and optimization methods applied for biodiesel

Feedstock	Analyzed performance	Experiment method	Prediction and optimization methods
Allamanda seed oil [96]	Yield	BBD	DFA
Jatropha [97]	HC, NOx, CO_2, CO	CCD	DFA
Mahua oil [98]	Yield	BBD	GA and DFA
Canola, safflower and waste vegetable oil [99]	BTE, EGT, NOx, CO_2	CCD	DFA
Waste cooking oil [100]	Yield	CCD	DFA
Algal oil [101]	Yield	BBD	ANN
Chrysophyllum albidum seed oil [102]	Yield	CCD	ANN-GA
Okra seed oil [103]	SO, HC, NOx, heat release rate	CCD	GA
Rubber seed oil [104]	Yield	CCD	ANN
Waste lard [105]	Yield	CCD	PSO, GA, FA

X. CONCLUSION

The study examines biodiesel-related patents and draws several findings. A more environmentally friendly and sustainable fuel source is thought to be biodiesel, especially in light of the growing demand for energy. In order to prevent food and fuel from competing with each other, indigestible materials should be used to produce biodiesel. Because of its affordability and advantages in waste management, second-generation biodiesel made from trash is currently preferred. The manufacture of biodiesel requires catalysts, and heterogeneous catalysts are favored due to their ease of separation and reusability. The transesterification process is the subject of the majority of discussed patents; it is widely used since it is easy to use and reasonably priced. Because of their practical operation methods, reactors—especially Continuous Stirred Tank Reactors (CTSR)—play a crucial role in the manufacture of biodiesel on an industrial scale. Mathematical modeling and optimization tools predominantly utilized are Design of Experiments (DOE), Artificial Neural Networks (ANN), and Metaheuristic (MH) algorithms, each offering unique advantages and facing specific limitations.

Each of these methods has its own set of advantages and drawbacks. DOE is crucial for experimentation and generating mathematical expressions vital for prediction. ANNs offer the unique ability to predict multiple outputs and inputs, a feature not provided by DOE, and are useful for real-time process monitoring. Meanwhile, MH algorithms enhance the predictive and optimization capabilities of processes by optimizing ANN parameters.

The integration of DOE, ANNs, and MH algorithms in biodiesel research represents a comprehensive approach, where each method's strengths effectively complement the others. While DOE establishes a foundational understanding of experimental variables, ANNs bring advanced predictive capabilities, and MH algorithms optimize these predictions for more effective and efficient solutions. This synergy is pivotal in advancing biodiesel research, leading to more sustainable and efficient energy solutions.

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