Trends in Sonochemical and Hydrodynamic Reactor Strategies for Catalytic Production of Biodiesel: Effects of the Influencing Process Parameters and Kinetics

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ABSTRACT
Biodiesel researchers need to understand the optimal conditions involved in the production of biodiesel from readily available biological sources, as several research works have reported on biodiesel production. Therefore, this paper emphasizes specifically, the process parameters involved in biodiesel production and how they affect biodiesel yields. These parameters include, but not limited to the feedstock selection, catalyst type to use, free fatty acid, temperature, kinetics, hydrodynamics and reactor conditions. In biodiesel synthesis, a high fatty acid methyl ester yield of up to 100%w/w at 60 °C has been reported, which occurred with a methanol to oil molar ratio of 3.75, and 60 min reaction time. Homogeneous catalysts seems promising for the production of biodiesel, although they possess disposal challenges and reusability issues. In addition, carbon-based catalysts from natural sources have been used to resolve the presence of free fatty acids in biodiesel synthesis that results in the formation of soap. These carbon-derived catalysts prove their efficiency when modified with acids. The reactor suitable for biodiesel reaction, assume several configurations, like the batch, fixed bed and semi-batch configurations, with their respective reaction conditions. Furthermore, in the design of a hydrodynamic cavitation reactor operating on the rotor-stator mechanism, research has shown that the ratio of rotor to stator diameter Dr/Ds is maintained at 0.73 for efficient operation. Hence, a proper understanding of the process chemistry and techniques involved in biodiesel synthesis would ensure a high desired yield and sustainable process route.

Keywords: biodiesel, sonochemistry, catalysts, biomass, kinetics, FFA, reactor, hydrodynamic

INTRODUCTION

The inevitable industrial paradigm shift has revolutionized researchers’ attention from the conventional fossil fuel approach to produce fuels and chemicals, to the novel use of biomass, ranging from lipids to grease or wastes, to generate fuel-like substances. From the industrial oil boom era, the transportation sector became a huge consumer of fossil fuel and oil reserves available, which as a result, attention is currently being shifted to readily available biological sources. Gashaw and Teshita (2014) affirm that biodiesels poses to be a very promising alternative to diesel oil, this being that they have similar properties and are renewable. Biodiesel can be generated from animal fats or vegetable oils (Lipids), through a trans-esterification reaction (Fadhil et al, 2012). Majority of the world’s energy comes from petroleum-based fuels, but the environmental ruin from the use of petroleum-based fuels and from human activities is great, and this includes the release of toxic pollutants and greenhouse gases from the use of coals etc. (Anusi et al., 2018a). Non-edible oil became the focus for biodiesel researchers, since the use of edible lipids for biodiesel production has been of great concern lately, because they contend with food materials. Therefore, the attention on first generation biofuel sources cannot be recommended for the production of biodiesels. Leo et al. (2019) used waste free fatty acid (FFA)-based feeds—which include brown grease (BG) to produce diesel-like fuels. This shows that biodiesel production extend beyond the use of first-generation sources, as it cuts across Fats, Oils and Grease (FOG). The benefits of using biodiesel, go beyond its view as being biodegradable and...
non-toxic. In comparison with petroleum-based diesel, biodiesel emits low volume of carbon monoxide and particulates (Zhang et al., 2003). Despite these merits of using biodiesels over petroleum diesel, taking into account, its attendant difficulties in the production is relevant. Umenweke (2020) reveals that understanding the proper chemistry and (transesterification) reaction involved in biomass valorization process, would help the researchers obtain maximum results that aims towards environmental sustainability. Within the literatures reviewed by the authors, there hasn’t been much work or literatures that assess design parameters of biodiesel production, incorporating the relevance of sonochemistry and hydrodynamics in its study. Hence, this is the knowledge gap this review paper tends to address.

Therefore, this paper assesses the trends of how process parameters such as feedstock selection, catalysts selection, temperature, hydrodynamics (in relation to sonochemistry), the reaction kinetic approach, and their individual effects on the biodiesel yields. The review aims at proposing ‘optimum condition(s)’ biodiesel researchers can adopt to help increase biodiesel yields.

**OVERVIEW OF DRAWBACKS IN BIODIESEL PRODUCTION**

Understanding the design schemes for biodiesel production is highly essential, as it is vital to optimum yields of biodiesel products. Apart from the reaction chemistry, the process parameters and designs are paramount, as these design parameters includes Temperature, hydrodynamics, free fatty Acid (FFA), reactor type/design and reaction time. The most challenge involved in biodiesel production is greatly faced during the pre-processing stage, in the course of the processing and post-processing of the feedstock. The general notion by non-scientific or - government organization is that the constant production of biodiesel, will result into acute shortage of food (Anuar, 2016). Although, this complain has drawn the research focus on biodiesel away from both first- and second-generation biofuel sources. The cost ineffectiveness of the biodiesel production process is alarming, as the feedstock pretreatment plant alone, is enough to affect the return on investment of the entire process. According to Balat (2006), other technical drawbacks of biodiesel and petroleum-based diesel include fuel freezing in cold weather, storage issues, i.e., degradation of fuel under storage and decreased energy density. Vegetable oils needs to be transesterified to biodiesel because of its high viscosity and cold flow properties by reacting it with alcohol in the presence of basic or acid catalyst for biodiesel production (Bello et al., 2013). The main process variables that had effect on transesterification reaction are as follows; catalyst type and amount, reaction temperature, reaction time, contents of FFA (free fatty acids), water, and molar ratio of alcohol to vegetable oil, and intensity of mixing during chemical reaction (Pahola and Urmila, 2012).

Biodiesel has been largely accepted as a renewable energy option with advantages over petroleum diesel and as substitute energy source especially as blazing of biodiesel makes relatively very low contribution to the carbon footprint and greenhouse gas emissions. A major limiting factor in biodiesel production is that it is expensive to produce which stems from the cost of raw materials and energy consumption of the manufacturing process. The cost of raw material typically contributes to about 70-80% of the overall cost of production for biodiesel (Gole and Gogate, 2012). The production of biodiesel is made possible by different processes. However, Garnesh et al. (2018) observed that the transesterification reaction between a triglyceride (usually present in vegetable oils) and an alcohol using a strong catalyst (acid, base, or enzymatic) is the easiest method used and hence the most explored.

**PROCESS PARAMETERS AND EFFECTS ON YIELDS**

**Biomass/Feedstock Selection**

The quality of feedstock selected for biodiesel production, affects not just the yield of biodiesel, but is largely dependent on cost. The high-quality feedstock, such as edible oils (like vegetable oils), can really consume up to an average of 80% of the biodiesel production cost. Although, using edible oil would result in an FFA of about less than 1% (Van Gerpen et al. 2004). Biodiesel is a renewable fuel that can sustain economic, development, energy security, and reduce to the barest minimum the inherent environmental effect of greenhouse gases emissions pose by fossil fuel. Therefore, there is a need for an alternative economic approach to reduce biodiesel cost of production. This has shifted researchers’ attention to the use of low-quality feedstock, for example, spoiled soybeans and waste cooking oils etc. It should be noted that Fatty Acid Methyl Esters (FAME) and Hydrotreated Vegetable oil (HVO) are from same organic biomass, but differ in production process and physicochemical properties. Although biodiesel feedstock varies greatly, it is paramount to consider cost and product yields in the feedstock selection. Figure 1 shows the sources of several biomasses selected for biodiesel production.
Lipids

Lipids are fats and oils. Generally, there are two types of oils, which can be obtained from both edible and non-edible plants. Previously, edible oils are the most used sources to produce biodiesel fuel. Examples are Palm oils, soybeans and sunflower etc. Therefore, since edible plant sources competes with food and in addition to their higher prices, waste vegetable oils (or waste cooking oil) and non-edible crude vegetable oils like the avocado oil, orange oil, mango oil and beniseed oil etc. are now prevalent (Anusi et al., 2018b). According to Johnston and Holloway (2007), lipids (both edible and non-edicibles) are very much reliable for the production of vegetable oils (or waste cooking oil) and non-edible crude vegetable oils (or waste cooking oil) and non-edible crude vegetable oils and used cooking oils contain a high content of free fatty acids (FFAs), which increases the overall biodiesel production rate. The interference of water and free fatty acids (FFA) in the alkali catalyzed process of producing biodiesel has resulted into several process and economic limitations. While feeding the low-grade oil constituents, it is proposed that an acid catalyst be introduced in the process. However, the major challenge with using acids as catalysts for transesterification is that the rate of reaction is much slower than that catalyzed by alkaline – based catalysts with a tougher reaction condition.

Microalgae oil is another very promising feedstock for biodiesel production. In fact, biodiesel production from microalgae oil is more promising and a sustainable alternative to previously mentioned feedstocks (Hanifa et al., 2011). Compared to plants, algae do not compete with food crops and have higher energy yields per area than terrestrial crops. Main advantage of using microalgae as feedstock is their rapid growth potential with short biomass doubling time (3.5 hours) during exponential growth and oil content ranging from 20 to 50% dry weight of biomass for numerous microalgae species. Hanifa et al. (2011) suggests that to use microalgae for the production of biodiesel, several processes have to be carried out. These consist of strain selection, cultivation, harvesting, extraction of the oil, and production of biodiesel from extracted oil, in which each step can be accomplished with various technologies.

Sewages sludge

Municipal sewage sludge is another promising feedstock for biodiesel production. Sewage sludge (also called 'biosolids') is a by-product generated in wastewater treatment facilities after primary and secondary treatment processes, normally entailing anaerobic digestion which produces both biogas and a semi-solid residue (sludge). Activated sludge wastewater treatment processes produce two main types of sludge: a primary sludge, normally a combination of floating grease and solids and a secondary sludge, mainly composed of microbial cells and suspended solids produced during aerobic biological wastewater treatment. New research indicates that the lipids contained in sewage sludge (primary and secondary sludge) may be a potential feedstock for biodiesel production (Capodaglio and Callegari, 2018; Kargbo, 2020). In particular, the drying of sewage sludge and the extraction process for recovery of the lipids fraction seem to be the most important challenges to overcome when developing a process for biodiesel production using sewage sludge as feedstock. Biodiesel production from sewage sludge poses huge challenges to overcome if commercial opportunities are to be realized. Some of these challenges are not unique to biodiesel production from waste sludge but to the biodiesel industry as a whole. They include challenges from collecting the sludge, optimum production of biodiesel, maintaining product quality, soap formation and product separation, bioreactor design, pharmaceutical chemicals in sludge and economics of biodiesel production (Kargbo, 2020).

Millions of tons of waste pile up in landfills, and these wastes could generate enormous amount of energy. Energy generated from household refuse incineration plants in the EU only increased 0.7% in 2015 and attained 8.7 million tons, this thereby increases garbage export and storage prices (Pieta et al., 2018). Sewage sludge refers to residual (slurry) material from the treatment of urban wastewater. The treatment of...
sewage produces sewage sludge, which is generated in large amount. It is observed that the sludge contains considerable lipid, which is capable of undergoing transesterification process. According to Kargbo (2010), there are obvious challenges in the process of generating biodiesel from sewage sludge, and to enable the commercial production of biodiesel, these challenges must be overcome. These challenges range from collecting the sludge, optimum production, FFA concerns, and bioreactor design, and pharmaceutical chemical sludge, economic and regulatory concerns.

The sludge that is produced during wastewater treatment from sewage is further subjected to treatment before disposal leading to major cost in a sewage treatment plant (STP) operation to about 50%-60% (Capodaglio and Callegari, 2018). The illegal disposal of sludge result into several environmental challenges. Therefore, recommended techniques such as landfilling, incineration etc. Wu et al. (2016) prepared KOH/activated carbon as solid base catalysts, using wet impregnation method to convert sewage sludge to biodiesel via in-situ transesterification reaction. This shows how biodiesel research has turned towards the use of heterogeneous catalyst. The yield generated by the use of fatty acid methyl esters (FAME) when catalysts are introduced is above 90% (Jiang et al., 2011). Usman (2018) formulated several treatment steps to handle sewage sludge, and converting it to biodiesel through transesterification reaction. The steps followed filtration, drying, fluid extraction, transesterification and finally separation from glycerol. The generation of biodiesel from sewage sludge treatment will be cost effective if only it would be operated on continuous operation. Therefore, there is a need to scale up the batch process involved in transesterification. More so, the biodiesel generated from wastewater sewage treatment would be an amazing source of power, if properly harnessed (Usman, 2018). Several solvents can be utilized to extract lipids from sewages, and they are ethanol, methanol, methane, hexane, chloroform and toluene, with preference given to toluene and chloroform due to high volume of extract. The solvent selection is based on polarity, boiling point, cost and miscibility (Clarke et al., 2018).

**Effects of Reaction Temperature and Catalysts on Biodiesel Production/Yield**

Catalyst is very vital in transesterification of vegetable oils to produce biodiesel (BD). Recently, biological and chemical catalysts, their challenges and prospects are being investigated. For favored industrial application, these catalysts are expected to be low-cost and environmentally benign. The chemical catalysts include homogeneous, heterogeneous, and supercritical fluids catalysts. Studies have shown that these catalysts are very effective in tailoring the transesterification reaction to high biodiesel yield; however, homogeneous catalysts are corrosive to equipment and require energy-intensive purification and separation steps. Besides, catalyst recovery and reuse are problematic. These limitations favor the use of heterogeneous catalysts, which could be reused, and separated from the reaction mixture via centrifugation. Also, they have comparable catalytic performance and could be derived from natural sources and sludge. Figure 3 shows the catalytic processes to produce biodiesel. The use of supercritical fluid technique to promote BD production is a notable research effort. Compared to the conventional catalytic process, the supercritical fluid offers easier separation, faster reaction and catalyst is not required to achieve comparably high biodiesel yield. Also, the bi-phasic problems of oil and methanol are mitigated. Biological catalysts have been employed to tailor the transesterification of oil. Notably, enzymes of lipase and immobilized lipase have received great attention. The use of enzymes to tailor the production BD offers several environmental advantages and the milder reaction conditions. However, their reaction takes longer time and some of the applied enzymes are easily
non-catalytic supercritical methanol processes are presented. Also, heterogeneous catalysts of alkali metal, alkaline earth metal, and ionic liquids. Also, deactivation if the oil contains impurity. Here we focus on homogeneous and heterogeneous catalysis, with emphasis on heterogeneous catalysts of alkali metal, alkaline earth metal, and ionic liquids. Also, non-catalytic supercritical methanol processes are presented.

Chemical catalyst either homogeneous (alkali or acid) or heterogeneous (solid acid or solid alkali, nano-catalysts etc.), are effective in bringing the process to completeness, although the reaction demands high energy and a cumbersome purification process to obtain the purified end product (Baskar et al., 2019). Lipases are the most common and feasible biological catalyst for biodiesel production. They have the advantage of having low operating conditions and high product purity. Enzymatic transesterification can be carried out at 35 to 45°C (Hanifa et al., 2011). Contrary to chemical catalysts, enzymes do not form soaps and can catalyze Oils with high FFA by also catalyzing esterification of the FFA to produce biodiesel (Shereena and Thangaraj, 2020). Shereena and Thangaraj (2020) highlighted important variables that influence the trans-esterification reaction. These include: reaction temperature, molar ratio of alcohol to oil, catalysts, residence time, and stirring/mixing intensity. Since triglycerides and alcohols do not mix at room temperature, the chemical reaction is usually carried out at a higher temperature and under continuous stirring, to increase the mass transfer between the phases (Romano and Sorichetti, 2011). In recent years various intensification technologies have been developed to overcome this drawback such as mechanical mixing, simple stirred reactors that can dramatically increase the reaction rate and time needed for complete conversion of the reactants to products. Biodiesel

### Table 1. Optimum temperature for biodiesel production from various substrates

| Substrate (Source) | Lipase/Catalyst | Optimum temperature (°C) | References |
|--------------------|-----------------|--------------------------|------------|
| Castor oil         | Immobilized bacillus aerius | 55 | Narwal et al. (2015) |
| Waste edible oil   | Rhizopus orizea PTCCS174 | 35 | Pasek et al. (2011) |
| Soybean oil        | Rhizopus chinensis CCTCC M201021 | 30 - 40 | Qin et al. (2008) |
| Waste cooking oil  | Immobilized candida | 40 - 50 | Chen et al. (2009) |
| Palm oil and palm kernel oil | Aspergillus niger | 40 | Kareem et al. (2017) |
| Soybean oil        | Novozym 435 | 40 - 50 | Du et al. (2003) |
| Sweet basil seed oil | Novozyme 435 | 40 - 60 | Amini et al. (2017) |
| Jatropha oil       | Immobilized Enterobacter aerogenes | 55 | Kumari et al. (2009) |
| Waste cooking oil  | lipase          | 55 | Istininigrum et al. (2017) |
| Neem seed oil      | Potassium hydroxide | 55 | Abbah et al. (2016) |
| Palm oil | K2O on MCM – 41 synthesized from rice husk | 100 | Hanumanth and Navindgi (2012) |
| Chlorella vulgaris | CuO/Zeolite | 60 | Dianursanti et al. (2016) |
| Palm kernel oil    | KOH             | 60 | Alamu et al. (2009) |
| Waste groundnut oil, waste soybean oil, waste palm kernel oil | KOH | 60 - 62 | Ayoola et al. (2017) |
| Waste cooking oil  | CaO nano - catalyst | 50 | Degfie et al. (2019) |
| Soybean oil        | NaOH            | 50 | Bashiri and Poubeiram (2016) |
| Rapeseed oil       | NaOH            | 55 – 65 | Wu et al. (2018) |
| Jatropha oil       | KOH             | 60 | Syam et al. (2015) |
| Canola oil         | NaOH            | 65 | Bala and Chidambaram, (2016) |
| Linseed oil        | NaOH            | 40 - 60 | Kumar et al. (2013) |
| Sunflower oil      | KOH             | 50 | Klofutar et al. (2010) |
| Sunflower oil      | NaOH            | 60 | Bambase et al. (2007) |
| Euphorbia lathyris | NaOH            | 65 | Adeniyi et al. (2019) |
| Jatropha curcas    | -              | 65 | Adeniyi et al. (2017) |

deactivated if the oil contains impurity. Here we focus on homogeneous and heterogeneous catalysis, with emphasis on heterogeneous catalysts of alkali metal, alkaline earth metal, hydrotalcites, carbon-based catalyst, and ionic liquids. Also, non-catalytic supercritical methanol processes are presented.

The process of transesterification can take place at different reaction temperatures based on the nature of oil used because temperature is a crucial process variable that influences the rate of reaction and yield of methyl esters. Higher temperatures improve the transesterification process by reduction of the mass transfer effect and the higher energy state to the molecules (Noureddini and Zhu, 1997). The required residence time is directly dependent on other parameters that enhance the rate of transesterification reaction, such as mixing intensity, residence time distribution (RTD), and temperature (Tabatabaei et al., 2019). Generally, the reaction is carried out close to the boiling point of methanol (60°C-70°C) at atmospheric pressure at molar ratio (alcohol to oil) of 6:1 (Huaping et al., 2006; Pramanik, 2003; Srivastava and Prasad, 2000). It was found out that higher reaction temperature increased reaction rate and shortened reaction time due to reduction in viscosity of oils (Masterton et al., 2011). However, it was discovered that there is a decrease in biodiesel yield once the reaction temperature is increased above the optimal level. This is because an increase in temperature favours saponification of the triglycerides (Mathiyazhagan et al., 2011). The effect of temperature on the biodiesel yield cannot be overemphasized, as Table 1 shows the optimum temperature required for the reaction to occur with different feedstock.

**Effects of Hydrodynamics and Sonochrome**

Shereena and Thangaraj (2020) highlighted important variables that influence the trans-esterification reaction. These include: reaction temperature, molar ratio of alcohol to oil, catalysts, residence time, and stirring/mixing intensity.
synthesis is limited by mass transfer, these technologies enhance reaction rate, reduces required molar ratio of alcohol to oil, reduces energy input by intensification of mass transfer and makes product separation easier.

Literatures have presented two main approaches in reducing the cost of biodiesel production process. The first approach is the use of waste cooking oil (WCO) as explored by Chuah et al. (2015), Ghayal et al. (2012), Kolhe et al. (2017), and Maddikeri et al. (2014) or non-edible oil resources such as rubber seed oil (Gupta et al., 2015) and Cannabis Sativa oil (Khan et al., 2019). The use of WCO and non-edible oil is an environmentally friendly solution and also solves the problem of the food for energy dispute. The second approach is the use/development of novel technologies to reduce raw material, energy consumption as well as increase rate of reaction (Abbaszadeh-mayvan et al., 2018). Energy consumption of the reactor is a function of the rate of the reaction in the process intensification synthesis for biodiesel production as opposed to conventional methods. Chipuricci et al. (2019) defined process intensification as the development of methods and/or equipment which provide higher yields/conversion with greater benefits compared to existing procedures. Process intensification processes include sonochemical reactors, microwave irradiated-assisted reactors (Milano, 2018) and hydrodynamic cavitation reactors amongst others. These processes are an economically viable approach/option in the synthesis of biodiesel production over conventional methods like mechanical stirring. In recent years to overcome mass transfer limitations in trans-esterification reaction systems, a novel technology gaining relevance is the cavitation technology which increases the rate of the chemical reactions otherwise limited by mass transfers. Cavitation as it affects rates of reaction finds application in ultrasound (sonochemistry) and hydrodynamic cavitation (effect) on reactors. Cavitation is a phenomenon that occur when a liquid in motion forms vapor-filled cavities or bubbles which grows, then experience implosion or a sudden collapse as a result of being subjected to a sudden rapid decrease and subsequent increase in pressure. Cavitation allows for more intense mixing. When the bubbles in the liquid are formed as a consequence of sound irradiation, acoustic cavitation occurs. This is the basis of sonochemistry. Cavitation as applied to sonochemical reactors is the passage of high intensity ultrasound through a liquid medium. Ultrasound applied to chemical reactions (sonochemistry) promotes the acoustic cavitation of fluids. Sonochemistry has aided many reactions. Hilares (2017) explained that, in lignocellulosic biomass conversion, it provides a severe physio-chemical environment for recalcitrant, multi component and heterogeneous lignocellulosic biomass that is difficult to obtain with other engineering methods. The introduction of ultrasound plays a positive influence essentially on the pretreatment and extraction of biomass.

**Hydrodynamic Cavitation**

Hydrodynamics is a phenomena associated with flow of liquid to produce energy usually by passing the liquid at high flow through a constriction. Hydrodynamic reactors work with the principle of cavitation hence it is popularly referred to as hydrodynamic cavitation (HC). Hydrodynamic Cavitation technology has been deployed as an intensification process for trans-esterification reactions using different feedstock. However, Maddikeri et al. (2014) was the first to apply this technology in the intensification of inter-esterification reaction in the synthesis of biodiesel from waste cooking oil.

Hydrodynamics cavitation influences the rate of reaction thereby increasing oil yield by the cavitation effect generated. This is brought about by the reactor configuration. The reactor configuration is hinged on the operating mechanism (rotor-stator mechanism) and/or the geometry of the constriction (slit venturi, a circular venturi or an orifice plate) of the reactor. When HC effect is generated by passing fluid through a constriction for example an orifice plate or venturi, the high turbulence is created as a result of the sudden implosion of the bubbles caused by an increase in the kinetic energy which occurs simultaneously with the decrease in pressure to a critical point as the fluid converges to pass through the constriction which has smaller cross-sectional area in the reactor (based on Bernoulli principle) assists in mass transfer between the immiscible reactants. The Hydrodynamic cavitation effect is also created by impeller design of the reactor. This is a process whereby the liquid flows through a rotor-stator type generator to form shockwaves which also leads to intense stirring of the reactants.

From literature HC has proved to have a better mixing capacity than mechanical stirring (Chuah et al., 2015) and sonochemical reactors (Gole et al., 2015). Even though sonochemical reactors yield good result, there is great difficulty in scaling it up. However, HC is one of the energy efficient, simple and cheapest methods of generating cavitation and scale up of this process is relatively easy. The use of hydrodynamic cavitation has enabled scientists overcome the drawbacks of conventional methods. Chuah et al. (2015) and Gole et al. (2015) in a comparative performance of HC and mechanical stirring and ultrasound cavitation, used the same reactor capacity and conditions to synthesize biodiesel and arrived at 98.1% yield of biodiesel while mechanical stirring was 19% in 15 minutes. Gole et al., (2013) reported that using ultrasound cavitation achieved 93% biodiesel yield was achieved after 40 minutes of reaction which is 2.7 fold longer reaction time compared to HC. It can be concluded that HC reactor is the most effective intensification process for biodiesel production. This finding supports the work of Kelker et al. (2008) which reports that hydrodynamic mode of cavitation was found to be more energy efficient as compared to the acoustic mode of cavitation. Literature findings revealed that cavitation reactors have the advantage of being simpler and do not need any external equipment such as the ultrasound generator. They are easier to control in a continuous mode and hydrodynamics has been used for processes such as cleaning of water, wastewater treatment and ethanol production (Dular et al., 2016; Ramirez-cadavid et al., 2016).

Hydrodynamic cavitation is a novel technology although it has been applied many times in literature for increased yield of biodiesel. In all instances where hydrodynamic cavitation technology has been used, it resulted in over 90% oil yields. However, these different instances have optimized different process operating parameters and mechanisms. Where some of the researchers worked on geometric and operational parameters, others worked on chemical process operating
conditions in the hydrodynamic cavitation reaction as shown in Table 2.

Abbaszadeh-mayvan et al., (2018), in the pioneer study of the intensification of continuous biodiesel production from waste cooking oil using shockwave power reactor, the process was evaluated and optimized through Response Surface Methodology (RSM). The shockwave power reactor is a hydrodynamic cavitation reactor which works with a rotor-stator mechanism, i.e., running on a rotor/stator type generator to generate energy (heat) for the reaction. Though the experiment was designed with RSM based on Central Composite Design (CCD) and five operating parameters were optimized namely: ratio of rotor diameter to stator diameter (Dr/Dr), ratio of cavity diameter to rotor diameter (Dc/Dr), ratio of cavity depth to gap between rotor and stator (dc/Dr), rotational speed of rotor (N), and residence time (Tr). Regarding the results obtained the most important parameter affecting the methyl ester conversion is the molar ratio of methanol to oil (1:6), 1%wt catalyst, 60°C reaction temperature and 5 bar of upstream pressure as effected on different plate geometries (varying number of holes and hole diameter), 95% biodiesel yield was produced. In their experiment the significance of upstream pressure as affected on different plate geometries (varying number of holes and hole diameter), 95% biodiesel yield was recorded at optimum parameters of the geometry of orifice plate at 25 holes with diameter measuring 2mm (this was after using orifice plate with varying and different geometries). This is owing to the fact that small hole sizes as compared to the diameter of the orifice plate results in smaller hole diameters which ultimately results in higher number of cavities resulting in higher number of cavitation events hence better emulsification leading to a better mass transfer. This greatly improves the miscibility of oil and alcohols leading invariably to greater biodiesel yield achieved by cavity oscillation hence increasing the rate of the transesterification reaction.

Chuah et al. (2015) obtained 98.1% yield of biodiesel using the following optimum reaction parameters, with molar ratio of oil to methanol (1:6), 1%wt catalyst and 60°C reaction temperature. It has been established that an important process parameter affecting the methyl ester conversion is the molar ratio of oil to methanol. In this study an increase in oil to methanol ratio from 1:4 to 1:6 resulted in an increase in oil yield from 45.2% to 98.1% in 15mins reaction time. Although from stoichiometry, 3 moles of alcohol are required in the transesterification reaction. A higher molar ratio in favor of alcohol is preferred due to the fact that the transesterification reaction is reversible. This could be attributed to the increase in the amount of methanol. More cavities are generated due to hydrodynamic cavitation in methanol compared to oil (Khan et al., 2019).

The molar ratio of triglyceride to methanol should not exceed (1:6). Beyond this, the excess methanol decreased the biodiesel yield from 98.1% to 95% (1:7). This is attributed to dilution of oil in methanol. On the one hand, increasing KOH catalyst concentration from 0.5wt % to 1wt% increased the methyl ester conversion as reaction rate was enhanced. On the other hand, an increase in catalyst concentration from 1wt% up to 1.25wt% the conversion decreased marginally from 98.1 to 91%. Beyond the optimum level, soap formation was observed. It could be attributed to the excess of KOH. Similar result was also reported by Gole et al. (2015).

Ghayal et al. (2012) established that flow geometry of the orifice plate of the HC reactor plays a crucial role in the intensification process of biodiesel and subsequently the yield produced. In their experiment the significance of upstream pressure as effected on different plate geometries (varying number of holes and hole diameter), 95% biodiesel yield was recorded at optimum parameters of the geometry of orifice plate at 25 holes with diameter measuring 2mm (this was after using orifice plate with varying and different geometries). This is owing to the fact that small hole sizes as compared to the diameter of the orifice plate results in smaller hole diameters which ultimately results in higher number of cavities resulting in higher number of cavitation events hence better emulsification leading to a better mass transfer. This greatly improves the miscibility of oil and alcohols leading invariably to greater biodiesel yield achieved by cavity oscillation hence increasing the rate of the transesterification reaction.

Kolhe et al. (2017) established optimum conditions for maximum biodiesel yield, though working on geometrical parameter of an orifice plate of 16 holes of 3mm diameter his main focus was the process conditions required. The optimum molar ratio of methanol to oil is 4.5:1. Catalyst concentration, reaction temperature and reaction time are the main factors affecting the reaction cost. Hydrodynamic cavitation requires mild reaction condition to carry out reactions. Hydrodynamic cavitation reactors not only increase the conversion and yield in short period of time but also reduces the quantity of alkali (KOH) requirement which further helps to reduce the separation time. This condition was further buttressed by Khan et al. (2019) when he established that the optimal operating parameters for transesterification reaction for optimum biodiesel yield were found to be as 1:6 molar ratio of oil to methanol, 1 wt. % of catalyst, 60°C of reaction temperature and 3 bar of upstream operating pressure.

### Table 2. Biodiesel yield using HC technology for different process optimized parameters on various feedstocks

| Feedstock                     | Catalyst | Time (sec) | Optimized process parameter | Yield (%) | Reference                      |
|-------------------------------|----------|------------|-----------------------------|-----------|--------------------------------|
| Waste cooking oil (WCO)       | KOH      | 30.10      | Rotor stator diameter (Dr/Dr) of shockwave reactor | 96.62     | Abbaszadeh-mayvan et al. (2018) |
| Palm oleinofi (WCO)           | KOH      | 900        | Molar ratio of oil to methanol (1:6), 1% wt catalyst, 60°C rxn temp. | 98.1      | Chuah et al. (2015)           |
| Used frying oil (UFO)         | KOH      | 900        | Geometry of orifice plate- 25 holes of 2mm diameter | >95       | Ghayal et al. (2012)          |
| Crude frying oil & RSO        | KOH      | 900        | Molar ratio of methanol to oil (4.5:1) | 95.6      | Kolhe et al. (2017)          |
| Cannabis sativa L. oil        | KOH      | 1200       | Geometry of orifice plate with 7 holes of 3mm diameter, 6:1 molar ratio of oil to methanol, 1wt% catalyst, 60°C, upstream operating pressure of 5bar | 97.5      | Khan et al., 2019            |
| WCO                           | Slit venturi | 90        |                               |           | Maddikeri et al. (2014)       |
Maddikeri et al. (2014) experiment explored the geometric parameter of the HC reactor for the intensified synthesis of biodiesel using HC, he focused on the use of different cavitation devices such as the orifice plate, circular and slit venturi on the hydrodynamic cavitation reactor on varying process operating parameters such as inlet pressure, molar ratio of oil to methyl acetate and catalyst loading. Maximum oil yield of 90% was obtained at optimum conditions of oil to methyl acetate ratio of (1:12), catalyst loading of 1% using slit venturi at an inlet pressure of 3 bar-fixed conditions. The power dissipated into the system for the slit venturi is 1.1 times higher for circular venturi and 1.5 times higher than orifice plate. The observed results can be explained on the basis of higher volumetric flow rate for given pressure drop and lower cavitation number obtained in slit venturi as compared to circular venturi and orifice plate.

**Influence of Reaction Mechanism on Biodiesel Production**

Generally, reaction kinetics give a measurement of reaction rates, factors that affect how the chemical reaction proceeds, and knowledge about the reaction mechanisms. Therefore, significance of this kinetic study is to determine the most effective way transesterification reaction can proceed, and how fast the lipids can convert into biodiesel.

The hypothetical stoichiometry of the reaction of transesterification in biodiesel production is as stated below;

\[
\begin{align*}
\text{CH}_2\text{O} - \text{C} - \text{R} & \rightarrow \text{CH}_2\text{OH} \quad \text{RCOOCH}_3 \\
\text{CH} - \text{O} - \text{C} - \text{R} + 3\text{CH}_2\text{OH} & \rightarrow \text{CH}_2\text{OH} + \text{RCOOCH}_3 \\
\text{CH}_2\text{O} - \text{C} - \text{R} & \rightarrow \text{Glycerol} \quad \text{Ethyl ester (Biodiesel)}
\end{align*}
\]

The overall transesterification reaction is summarized thus;

\[
\text{Tiglyceride} + 3\text{R'OH} \rightleftharpoons 3\text{R'COOR}_3 + \text{Glycerol}
\]

(1)

The chemical kinetics that describes the reaction pathway for transesterification reaction is shown as follows;

\[
\begin{align*}
\text{DG} + \text{ROH} & \rightleftharpoons \text{MG} + \text{E} \\
\text{MG} + \text{ROH} & \rightleftharpoons \text{GL} + \text{E}
\end{align*}
\]

(2)\(\text{3)\}

The overall reaction is now given as;

\[
\text{TG} + 3\text{ROH} \rightleftharpoons \text{GL} + 3\text{E}
\]

(4)

Where TG is glyceride, DG is diglyceride, MG is monoglyceride, ROH is an alcohol (methanol), and E is ethyl ester.

Previous works that had been carried out by different researchers (Anusi et al., 2018; Amira et al., 2014; Ahiejpor and Kuwornoo, 2010; Igboke and Nwafor, 2014; and Igbum et al., 2012) on the transesterification of triglyceride reaction proved its second order rate of reaction constant with its governing equations characterizing the stepwise reaction mechanisms as follows;

\[
\frac{d\text{TG}}{dt} = -K_1\text{TG} \cdot A + K_2\text{DG} \cdot E - K_7\text{TG} \cdot A^3 + K_6\text{GL} \cdot E^3
\]

(6)

\[
\frac{d\text{DG}}{dt} = K_1\text{TG} \cdot A - K_2\text{DG} \cdot E - K_3\text{DG} \cdot A + K_4\text{MG} \cdot E
\]

(7)

\[
\frac{d\text{MG}}{dt} = K_3\text{DG} \cdot A - K_4\text{MG} \cdot E - K_5\text{MG} \cdot A + K_6\text{MG} \cdot E
\]

(8)

\[
\frac{d\text{GL}}{dt} = K_5\text{MG} \cdot A - K_6\text{GL} \cdot E + K_7\text{TG} \cdot A^3 - K_8\text{GL} \cdot E^3
\]

(9)

\[
\frac{d\text{E}}{dt} = K_1\text{TG} \cdot A - K_2\text{DG} \cdot E + K_3\text{DG} \cdot A - K_4\text{MG} \cdot E + K_5\text{MG} \cdot A - K_6\text{GL} \cdot E + K_7\text{TG} \cdot A^3 - K_8\text{GL} \cdot E^3
\]

(10)

\[
\frac{da}{dt} = \frac{da}{dt}
\]

(11)

Where \(K_1\) to \(K_8\) are rate constants of reaction rate, \(TG\), DG, MG, GL, A and E are the concentrations in weight percent of triglyceride, triglyceride, monoglyceride, glyceride, alcohol and ester respectively. The kinetic analysis of equations (1-11) was summarized in the following matrix transformation.

\[
\begin{pmatrix}
\alpha_{11} & \alpha_{12} & \alpha_{13} & \alpha_{14} & \alpha_{15} & \alpha_{16} & \alpha_{17} & \alpha_{18} \\
\alpha_{21} & \alpha_{22} & \alpha_{23} & \alpha_{24} & \alpha_{25} & \alpha_{26} & \alpha_{27} & \alpha_{28} \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\
\alpha_{n1} & \alpha_{n2} & \alpha_{n3} & \alpha_{n4} & \alpha_{n5} & \alpha_{n6} & \alpha_{n7} & \alpha_{n8} \\
\end{pmatrix} =
\begin{pmatrix}
K_1 \\
K_2 \\
K_3 \\
K_4 \\
K_5 \\
K_6 \\
K_7 \\
K_8 \\
\end{pmatrix}
\]

(12)

The unified time of reaction and disappearance rate equations for various industrial reactor types’ designs was developed by Abowei et al., (2020). Their work also looked at the mathematical modeling of reaction time at equilibrium, non-equilibrium and disappearance rate that can be used in design of industrial base reactor. The basis of their work is that the reaction of transesterification reaction is reversible bimolecular reaction of second order. The kinetic rate was mathematically defined by Abowei et al., (2020) as follows;

\[
A + 3B \rightleftharpoons R + S
\]

(13)

Where \(A\) is the vegetable oil (PKO), \(B\) is sodium hydroxide, \(R\) is glycerol and \(S\) is methyl ester.

Hence, the modified rate expression is mathematically written as;

\[
-A = -\frac{d\alpha_A}{dt} = K_1\alpha_A\alpha_B - K_2\alpha_B\alpha_S
\]

(14)
Where \(-R_A\) is the rate of disappearance, \(K_1\) and \(K_2\) are the reaction rate constants, \(C_A\), \(C_B\), \(C_R\), and \(C_S\) are the concentrations of vegetable oil, NaOH, glycerol and methyl ester respectively. The disappearance rate model is as follows;

\[
-R_A = K_1(C_{AO} - C_{AO}X_A)(C_{BO} - C_{AO}X_A)
- K_2(C_{BO} + C_{AO}X_A)(C_{SO} - C_{AO}X_A) \tag{15}
\]

\[
-R_A = K_1\left[C_{AO}(1 - X_A)\left(C_{BO} - X_A\right)\right]
- K_2\left[\frac{C_{BO}}{C_{AO}} + X_A\right]C_{AO}\left(C_{SO} + X_A\right) \tag{16}
\]

Equation (16) can be summarized below;

\[
-R_A = K_1\left[C_{AO}^2(1 - X_A)\left(C_{BO} - X_A\right)\right]
- K_2\left[\frac{C_{BO}}{C_{AO}} + X_A\right]C_{AO}\left(C_{SO} + X_A\right) \tag{17}
\]

Calculating voidage of this reaction shown in equation (13) using equation 18

\[
\varepsilon = \frac{SP - \xi R}{\xi R} \tag{18}
\]

It can be shown that the reversible bimolecular reaction of transesterification reaction is exothermic inculcating this concept of negative voidage into the kinetic rate model of equation (17). Hence, the new kinetic rate model is;

\[
-R_A = \frac{C_{AO} - C_{AO}X_A}{1 + \varepsilon_XA}\left[C_{BO} - C_{AO}X_A\right]
- \frac{C_{BO} + C_{AO}X_A}{1 + \varepsilon_XA}\left[C_{SO} + C_{AO}X_A\right] \tag{19}
\]

Where;

\[
C_A = C_{AO}\left(1 - X_A\right)\frac{1}{1 + \varepsilon_XA} \tag{20}
\]

Equation (19) is factorized to give;

\[
-R_\varepsilon = \frac{C_{AO}^2}{1 + \varepsilon_XA}\left[K_1(1 - X_A)\left(C_{BO} - X_A\right)\right]
- K_2\left[\frac{C_{BO}}{C_{AO}} + X_A\right]C_{AO}\left(C_{SO} + X_A\right) \tag{21}
\]

If we assumed \(M =\frac{C_{BO}}{C_{AO}}, N =\frac{C_{SO}}{C_{AO}}, P = \frac{C_{BO}}{C_{AO}}\) and these are inserted into equation (21) it will result in equation (22) below which is the modified Abowei et al. (2020).

\[
-R_\varepsilon = \frac{C_{AO}^2}{1 + \varepsilon_XA}\left[K_1(1 - X_A)(M - X_A)\right]
- K_2(N + X_A)(P + X_A) \tag{22}
\]

Time of reaction for transesterification process is determined as follows;

\[
-R_\varepsilon = C_{AO}\frac{dX_A}{dt} = \frac{C_{AO}^2}{1 + \varepsilon_XA}\left[K_1(1 - X_A)(M - X_A)\right]
- K_2(N + X_A)(P + X_A) \tag{23}
\]

Equations 22-24 is the modified Abowei et al., (2020) for rate expression for the bimolecular reversible vegetable oils and C1 and C3 alcohol catalyzed transesterification reaction in biodiesel production and glycerol.

Determination of time of reaction for transesterification reaction:

**Case 1:** For isothermal condition from equation (25) at equilibrium assuming that \(C_{AO} = C_{BO}\) and \(C_{SO} = C_{PO} = 0;\)

\[
\frac{dC_{AE}}{dt} = 0, \text{ the fractional conversion of A (vegetable oil) at equilibrium conditions was established from equation (23) to be equation (25)}
\]

\[
K_e = \frac{K_1}{K_2} = \frac{(N + X_A)(P + X_A)}{(1 - X_A)(M + X_A)} \tag{25}
\]

Re-writing equations (23) and (25)

\[
\frac{1}{K_2}\frac{dX_A}{dt} = C_{AO}[K_e(1 - X_A)(M - X_A)]
- (N + X_A)(P + X_A) \tag{26}
\]

Substituting equation (25) into equation (26) we have

\[
\frac{1}{K_2}\frac{dX_A}{dt} = C_{AO}\left[\frac{(N + X_A)(P + X_A)}{(1 - X_A)(M + X_A)}\left[(1 - X_A)(M - X_A)\right] - (N + X_A)(P + X_A)\right] \tag{27}
\]

However, simplifying equation (27), applying induction and solving algebraically with subsequent integrations we get equation below;

\[
\ln\left(\frac{X_A(2X_A - 1)X_A}{X_A - X_A}\right) = \frac{1}{2K_1}\left[\frac{1}{X_A} - 1\right]C_{AO}t \tag{28}
\]

\[
t = \frac{1}{2K_1}\left[\frac{1}{X_A} - 1\right]C_{AO}\ln\left(\frac{X_A(2X_A - 1)X_A}{X_A - X_A}\right) \tag{29}
\]

where \(X_A\) and \(C_{AO}\) are the fractional conversion and concentration of vegetable oil (PKO).

**Case 2:** For non-isothermal equilibrium exothermic conditions, solving equation (29) above using integration by partial fraction technique as follows;

\[
\int\frac{(1 + \varepsilon_XA)dx_A}{K_e(1 - X_A)(M - X_A)} - \int\frac{(1 + \varepsilon_XA)dx_A}{K_e(N + X_A)(P + X_A)} = C_{AO}\int dt \tag{30}
\]

If we assumed

\[
B = \int\frac{(1 + \varepsilon_XA)dx_A}{K_e(1 - X_A)(M - X_A)} \tag{31}
\]

and

\[
C = \int\frac{(1 + \varepsilon_XA)dx_A}{K_e(N + X_A)(P + X_A)} \tag{32}
\]

Applying partial fractions method resulting to; for B keeping \(1/K_e\) as a multiplying factor, thus;

\[
\frac{A}{1 - X_A} + \frac{L}{(M - X_A)} = \frac{1 + \varepsilon_XA}{(1 - X_A)(M - X_A)} \tag{33}
\]

Let \(X_A = 1;\) then \(A(M - 1) + L(0) = 1 + \varepsilon_X; \text{ hence}\)
\[ A = \frac{1 + e_A \lambda}{M - 1} \]

For C; keeping 1/K as a multiplying factor; thus

\[ \frac{R}{N + X_A} = \frac{1 + e_A X_A}{(N + X_A)(P + X_A)} \]

Let \( X_A = -P \); then \( Q(0) + R(N-P) + R(0), 1 - e_A P; \) hence

\[ R = \frac{(1 - e_A) P}{(N - P)} \]

Let \( X_A = -P \); then \( Q(0) + R(P-N) + R(0), 1 - e_A N \)

\[ Q = \frac{(1 - e_A) N}{(P - N)} \]

\[ C_{act, t} = \frac{1}{K_1} \left[ \frac{1 + e_A}{M - 1} \ln(1 - X_A) + \frac{1 + e_A M}{1 - M} \ln(1 - X_A) \right] \]

\[ + \frac{1 + e_A N}{P - N} \ln(N + X_A) \]

\[ + X_A \frac{1 - e_A P}{N + P} \ln(P + X_A) \]

\[ C_{act, t} = \frac{1}{K_1} \left[ \frac{1 + e_A}{M - 1} \ln(1 - X_A) + \frac{1 + e_A M}{1 - M} \ln(1 - X_A) \right] \]

\[ - \frac{1 - e_A N}{P - N} \ln(N + X_A) \]

\[ + X_A \frac{1 - e_A P}{N + P} \ln(P + X_A) \]

\[ t = \frac{1}{C_{act}} \left[ \frac{1 + e_A}{M} \ln(1 - X_A) + \frac{1 + e_A M}{1 - M} \ln(1 - X_A) \right] \]

\[ - \frac{1 - e_A N}{P - N} \ln(N + X_A) \]

\[ + X_A \frac{1 - e_A P}{N + P} \ln(P + X_A) \]

Expanding the right hand of equation (41)

\[ \frac{dX_A}{dt} = \frac{C_{act}}{1 + e_A X_A} [K_1(1 - X_A)(M - X_A) - K_2(N + X_A)P(X + X_A)] \]

Equation (50) is the Abweie et al. (2020) generalized proposed mathematical models for the prediction of reaction time for vegetable oils and C; to C3 alkali catalyzed transesterification reaction process in the bio-diesel production.

**CONCLUSION**

It is highly undisputed that biodiesel production has come to stay, especially in times of the massive drift from the dependence on petroleum-based diesel to diesel from biogenic materials. Recently, researchers are very much interested in designing and characterizing sustainable catalysts to the safe production of biodiesel. The environmental sustainability of biodiesel is in the removal of sulphur content present. For the feedstock selection in biodiesel production, it is advisable to consider environmental sustainability. Recent research towards biodiesel production, utilize feedstocks from wastes and other non-edible lipids that don’t compete with food. It was found out that higher reaction temperature increased reaction rate and shortened reaction time due to reduction in viscosity of oils. Another major component of the transesterification process worth mentioning is the alcohol to oil ratio. While for chemical catalysis the process requires excess methanol above the stoichiometric 3:1 alcohol to oil ratio to drive the reaction well enough to the right, it varies from oil to oil but researchers generally prefer a ratio of about 6:1. Shahid et al. (2011) claimed that separation of biodiesel and glycerol was easier using KOH instead of NaOH. From several reviews, KOH was reportedly used as catalyst to obtain the maximum yield of biodiesel production specifically when using hydrodynamic cavitation technology. KOH was constantly used as a catalyst and this is due to the fact that KOH results in a higher conversion of biodiesel compared to other catalysts, as well as its high activity and low cost (Chua et al., 2015; Gupta et al., 2015; Kolhe et al., 2017). For enzyme catalyzed transesterification, the optimal ratio of alcohol to oil is the stoichiometric value of 3:1 as excess methanol will deactivate the enzyme. In summary, non-edible oils remain the best option as the most sustainable feedstock for biodiesel production although other promising sources could also complement. Also, as environmental concerns rise coupled with the need to valorize biodiesel production by utilizing or purifying the by product from the transesterification process, enzymatic catalysis using immobilized lipases could be the way to go because of the ease of separating the products and catalyst, reusability of the catalyst and ability to work with a wide range of oils. Biodiesel production from microalgae is considered to be the best efficient feedstock for biodiesel.
production to displace conventional feedstock’s and meet global demand of fuel (Hanifa et al., 2011). According to research, the geometric parameters for the shockwave reactor (A hydrodynamic cavitation reactor operating on the rotor-stator mechanism) showed efficient results with a ratio of rotor to stator diameter (Dr/Ds) as 0.73. In addition, it is essential to note that when the constriction used in the reactor is the orifice plate it should have geometry of 25 holes with the diameter measuring 2mm. However, when the constriction used in the reactor is a venturi meter, a slit venturi is most appropriate as power dissipated is 1.1 times higher than the circular venturi & 1.5 times higher than the orifice plate (Maddikeri et al., 2014).

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