State of the art in the optimization of physico-chemical operating parameters of hydrodynamic cavitation reactors for the production of FAME

Estado del arte en la optimización de los parámetros de operación físico-químico de los reactores de cavitación hidrodinámica para la producción de FAME

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#### Abstract

This article focuses on the results of several research studies that used mechanical devices in hydrodynamic cavitation reactors to intensify the transesterification reaction in biodiesel. Results show a pattern in the operating parameters of the hydrodynamic cavitation reactors where high rates of oil to biodiesel conversions were obtained. As a conclusion some parameters and their corresponding operating ranges are shown; these ranges are recommended to be considered in order to obtain de maximum efficiency in the production of biodiesel using hydrodynamic cavitation.

Biodiesel, hydrodynamic cavitation, mechanical devices, operating parameters, transesterification

#### Resumen

El presente artículo se enfoca en los resultados de los diferentes estudios de investigación que utilizaron dispositivos mecánicos en los reactores de cavitación hidrodinámica para la intensificación de la reacción de transesterificación del biodiésel. Los resultados de los diferentes estudios muestran un patrón en los parámetros de operación de los reactores de cavitación hidrodinámica donde se obtuvieron altas tasas de conversiones de aceites a biodiésel. Como conclusión al final se presentan los diferentes parámetros y rangos que se recomienda considerar para obtener la máxima eficiencia en la producción de biodiésel utilizando cavitación hidrodinámica.

Biodiésel, cavitación hidrodinámica, dispositivos mecánicos, parámetros de operación, transesterificación

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# Introduction

Global warming caused largely by fossil fuel exhaust gases has led to the establishment of multinational agreements (Kyoto Protocol and Paris Agreement) for the reduction of greenhouse gases (GHG). According to Yeletsky et., 2019 [1], forty-five percent of oil demand belongs to the transport sector, the latter being one of the main generators of ["CO"] \_"2". This has led to interest in the development of alternative fuels for diesel engines. Biodiesel is one of the most promising alternatives to the oil crisis, as it would replace petroleum-based diesel fuel.

Biodiesel can be blended in different concentrations, including B100 (pure biodiesel), B20 to B6. Blends at or below B20 are most commonly used in the United States, Europe and Brazil because they do not require special handling or engine modifications. A number of technical standards have been established for biodiesel fuel to maintain quality, including the European standard EN 14214, the international standard ASTM 6751 and ASTM D7467 (Sheinbaum-Pardo et al., 2013) [2].

Studies have been conducted to analyse the emissions generated by replacing diesel with biodiesel [3, 4, 5, 6]. A reduction in CO, HC and smoke opacity emissions was observed in the research results; however, CO2 and NOx emissions had a slight increase.

To produce biodiesel on an industrial scale, transesterification is considered the most suitable chemical reaction. Transesterification involves the production of long-chain fatty acid methyl esters (FAME) from vegetable or animal oils that are composed of triacylglycerols, i.e. glycerol esters of long-chain fatty acids with a low molecular weight alcohol. This reaction is catalysed using bases, acids or enzymes. The main disadvantage of transesterification is the slow conversion rate due to the immiscibility of the oil-alcohol phase, and longer reaction time [7]. Intensification of the transesterification reaction is necessary to reduce biodiesel production costs.

#### Hydrodynamic cavitation reactors

The chemical reaction of transesterification takes place in a reactor. There are different types of reactors that help to speed up the biodiesel production process by increasing the rate of the reaction. One of these reactors is the hydrodynamic cavitation reactor, which is mainly composed of a pump and a channel construction/mechanical device (orifice plates or Venturi tube) located downstream of the pump discharge shown in Figure 1. Then, vapour cavities are generated, the pressure increases again and the bubbles collapse, so that the surface contact experiences a considerable increase. In addition, a large amount of energy in the form of pressure and temperature is released due to the collapse of the vapour cavities [8].

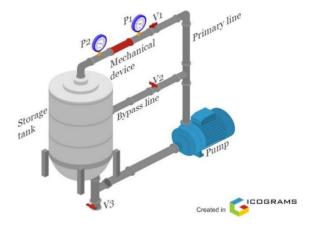


Figure 1 Hydrodynamic cavitation reactor

# Parametric operating effects on the transesterification reaction

The efficiency of a hydrodynamic cavitation reactor is determined by the physico-chemical operating parameters. The higher the reactor efficiency, the higher the biodiesel conversion and the lower the input energy. For this reason, several experimental studies [7, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23] report the optimal operating parameters with which conversions greater than ninety percent of biodiesel were obtained. Among the most important parameters are:

Oil to alcohol molar ratio,

Catalyst concentration,

Operating temperature,

Inlet pressure,

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Geometric parameter  $\alpha$ ,

Geometric parameter  $\beta$ ,

Geometric parameter " $\beta$ " \_"0".

Cavitation number C\_v.

#### Results

#### Oil to alcohol molar ratio

Experimental studies of biodiesel production using hydrodynamic cavitation reactors by [7, 10, 12, 13, 14, 16, 17, 19, 21, 22] obtained triglyceride to FAME conversion greater than 90%, when working with an oil to alcohol molar ratio of 1:6. This could be attributed to the generation of more cavities in methanol compared to oil, resulting in low mass transfer resistance [9].

The high viscosity of oil as a feedstock requires higher amounts of alcohol to increase the solubility of oil in alcohol [12]. Chitsaz et al., 2018 [17], states that for a molar ratio less than 1:4, no FAME is detected in the final product. This is because there was no liquid phase separation between the ester phase and the glycerol phase in the final product. However, beyond this, only a marginal increase in the degree of conversion is observed when the ratio increases to more than 1:8 [10]. This may be due to the nature of the reaction being reversible, an excess of alcohol limits the backward reaction giving an increase in the equilibrium conversion of the biodiesel sample [18].

However, the studies of [18, 20, 23, 11] were the only ones with an oil to alcohol molar ratio of 1:12 to obtain a triglyceride to FAME conversion of 99, 97, 96.5 and 90 % respectively..

### Catalyst concentration

The catalyst is used to create initial active ions in the esterification reaction and the amount of ions generally increases with catalyst concentration [10], but an overloading of the catalyst concentration results in an increase in wastewater during the washing stage [12]. According to the results of [10] he reports a twostage biodiesel conversion, obtaining a FAME conversion of 95%.

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In the study by [11] they obtained an increase in biodiesel yield from 79% to 89%, by increasing the catalyst concentration from 0.75% to 1% for a molar ratio of 1:12 and an inlet pressure of 3 bar. However, an increase from 1% to 1.25% of the catalyst concentration does not show any significant increase in biodiesel yield. One year later, [12] reports a considerable increase in the conversion of triglycerides to FAME from 63% to 98.1% by increasing the catalyst concentration from 0.5% to 1% for a molar ratio of 1:6 and a working temperature of 60 °C. In contrast, an increase in catalyst concentration from 1% to 1.25% has a decrease from 98.1% to 91% in the conversion of triglycerides to FAME. However, [16, 23] were the only studies that reported a biodiesel conversion of 94% and 96.5% with a catalyst concentration of only 0.5% respectively; for the study [16] they worked with a molar ratio of 1:4. 5 and an operating temperature of 45 °C, while [23] worked with a molar ratio of 1:12 and an operating temperature of 60 °C. Finally, [21] reported that for a FAME conversion of 75%, an optimum catalyst concentration of 1.2% was needed for a molar ratio of 1:6, inlet pressure of 5 bar and a temperature of 65 °C.

### Operating or working temperature

The operating temperature increases the solubility of triglycerides in methanol and gradually improves the contact between oil and methanol molecules, increasing thus the conversion of FAME. The high reaction temperature not only increases the reaction rate, but also the mass transfer [12]. The results of [12], report a significant increase from 79.0% to 98.1% in methyl ester conversion, when increasing the temperature from 50 °C to 60 °C, under conditions of 1:6 molar ratio and a catalyst concentration of 1% by weight of oil. In research by [13] they report a significant increase in biodiesel conversion from 44.8% to 97.1% by increasing the operating temperature from 40 °C to 55 °C, for the fixed operating conditions of 1:6 molar ratio and 1% catalyst concentration at a reaction time of 20 minutes. However, [21] reports a 75% conversion at a temperature of 65 °C.

### Inlet pressure

The effect of inlet pressure leads to an increase in the transesterification reaction rate, which in turn reduces the reaction time [21]. An increase higher than the optimum pressure causes that regions downstream of the mechanical device (orifice plate or Venturi tube), the local area is filled with a large number of cavities that merge and form a larger cavity, which causes a block in cavitation [14]. The results obtained by [11] when using a slit Venturi state that the optimum inlet pressure is 3 bar since, with an increase from 2 to 3 bar the ester conversion increased from 79 to 89% at the fixed conditions of 1:12 molar ratio and 1% catalyst loading in oil, but an increase in inlet pressure from 3 to 4 bar, there is no significant FAME conversion increase. A further increase in inlet pressure from 4 to 5 bar resulted in a marginal decrease of 89 to 85%. In research by [13] reports that for the optimum 21hole 1 mm diameter plate, an increase in ester conversion from 79% to 96.5% in 20 minutes was obtained by increasing the inlet pressure from 1 to 3 bar at a temperature of 55 °C; however, there was no difference in reaction rate from 3 to 3.5 bar. On the other hand, [18] reports that for the same 21-hole plate, there was an increase in ester conversion from 83% to 96.5% in 15 minutes, when the inlet pressure is increased from 1 to 2 bar at a constant reaction temperature of 60 °C. For pressure increase of 2-3 bar, there was no significant difference in reaction rate. In the investigation of [22], the highest conversion percentage of 98.3% was obtained at an inlet pressure of 4.4 bar, followed by a yield of 95.6% at a pressure of 2.9 bar and the lowest of 88.5% at an inlet pressure of 1.4 bar. The results of [7] was the only study to report a biodiesel yield of 99% at an optimum pressure of 7 bar using a 100-hole plate with a diameter of 0.3 mm.

#### Geometric parameter $\alpha$

The geometrical parameter  $\alpha$  is designated as the ratio of the total perimeter of the throat/orifices to the cross-sectional area of the mixture flow through the mechanical device. In an orifice plate, the smaller the orifice size and the higher the  $\alpha$  value, the better the conversion of the methyl ester. In the study by [9], he reports that for a 25-hole plate of 2 mm diameter with  $\alpha = [$ 2 mm ^(-1) he obtained a conversion of 97%. In the research of [14] they used orifice plates of four different values of  $\propto$  ( [4 mm] ^(-1), [2 mm ] ^(-1), [1.33 mm ] ^(-1), [0.87 mm ] ^(-1)). For hole plate  $\alpha = [4 \text{ mm}]^{-(-1)}$ , they reported that they obtained 98% conversion, followed by plate (  $[] \propto = 2 \text{ mm} ]]^{-(-1)}$  ) with 90% conversion, while for plate (  $[\alpha=1.33 \text{ mm}]$ ]  $^{(-1)}$  ) was 67% and for the plate of (  $\alpha=0.87 \text{ mm}$  ^(-1)) was 54%, with a reaction time of 15 min and inlet pressure of 2 bar. Similarly, [13] used the same mechanical devices, but with a constant reaction time of 20 minutes and inlet pressure of 3 bar. The results coincided in the order of the conversion plates, obtaining with the plate of (  $[\alpha=4 \text{ mm}]^{-(-1)}$ ) a 97.1 %, while the plate of (  $[\alpha=2 \text{ mm}]^{-(-1)}$ ) with 85.4%, plate of (  $[\alpha=1.33 \text{ mm}]^{-(-1)}$ ) 84.4% and plate of (  $\left[\alpha=0.87 \text{ mm}\right]^{-1}$ ) 64.3%.

### *Geometric parameter* $\beta$

The dimensionless geometric parameter  $\beta$  is defined as the ratio of the orifice diameter to the pipe diameter [9]. The smaller the value of  $\beta$ , the better conversion of the methyl ester tends to be. In the study by [9], he reported that for a 25-hole plate of 2 mm diameter with a value of  $\beta$ =0.10, a 97% conversion was obtained in a time of 20 minutes and with an inlet pressure of 1.5 bar. Three years later, [14] carried out an investigation using a plate with  $\beta$ =0.07. In the results, they obtained an ester conversion of 98%, using an inlet pressure of 2 bar and a reaction time of 15 minutes. On the other hand, [13] obtained a biodiesel yield of 97.10% with the same mechanical device.

### *Geometric parameter* $\beta_0$

The dimensionless parameter  $\beta$  0 is defined as the ratio of the total flow area to the crosssectional area of the pipe [9]. Conversion increases as the value of  $\beta_0$  decreases, i.e. the reaction rate increases with lower  $\beta_0$ . The lower the value of  $\beta_0$ , the better the conversion of the methyl ester tends to be. In the study carried out by [14] using two plates with the same value of  $\beta$  0=0.09, but with plate 2 he obtained 99% conversion of the ester while plate 1 obtained 55% conversion of the ester. Plate 2 has 21 holes with a diameter of (D=1 mm) and plate 1 has one hole with a diameter of (D=4.58 mm). On the other hand, [13] who worked with the same mechanical devices obtained a conversion of 99% for plate 2, and 65% for plate 1, using an inlet pressure of 3 bar and a constant reaction time of 20 minutes.

## *Cavitation number C\_v*

The cavitational events occurring inside the mechanical device and their subsequent effects on biodiesel conversion are characterised using the dimensionless number, known as the cavitation number [7]. This dimensionless parameter is defined by equation (1).

$$C_v = (P_2 - P_v) / (1/2 \rho V^2)$$
 (1)

Where P\_2 is the fully recovered water pressure, P\_ves the liquid vapour pressure of the reaction mixture depending on the individual moles of alcohol and oil, V is the velocity of the reaction mixture and  $\rho$  is the liquid density of the reaction mixture of the individual moles of alcohol and oil [19]. Cavitation is not favourable for conditions C\_v>1 while the cavitation intensity increases as the C\_v value decreases below 1 [7]. Cavitation number values in the range of 0.05 to 0.5 generally indicate enhanced cavitation activities in the reactor [9]. On the other hand, very low values of C\_v can lead to supercavitation resulting in vapour locking and absence of cavity collapse.

In the research by [11] they report the relationship between the inlet pressure in the mechanical device and the cavitation number. For the mechanical slit Venturi device with a working pressure of 3 bar and a C\_v=0.3 value, a maximum yield of more than 99% was obtained for a molar ratio of 1:12 and a catalyst loading of 1%.

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Also [14] confirmed that when the C v number was kept close to 0.3, maximum benefits were obtained in the conversion of methyl esters using the four different orifice plate geometries. For the investigation by [15] the methodology was applied to sixteen configurations of Venturi and cylinder arrangements and it was observed that the cylinder arrangement named 4510 (4 mm throat width arrangement, 5 mm cylinder diameter and 10 mm cylinder pitch) has the highest mean which, over the range of pressures investigated, is able to operate at the lowest C\_v (0.062), with the highest percentage of biodiesel conversion (95.2%). On the other hand, in the research carried out by [7], they obtained that for an optimum plate with 100 holes of 0.3 mm in diameter, a C\_v value of 0.34 at inlet pressure conditions of 7 bar and 5 minutes of reaction time, a biodiesel yield of 99% was obtained. Likewise, [21] reports that, with an orifice plate of 3 mm diameter, with a C\_v=0.63, a maximum biodiesel yield of 75 % was obtained at a working pressure of 5 bar.

### Conclusions

The molar ratio is one of the most influential parameters in the transesterification reaction, due to the higher cavity formation in methanol versus oil. An oil to alcohol molar ratio of 1:6 has given in most of the reported studies the maximum biodiesel conversion.

### Catalyst concentration

A catalyst concentration of 1% by weight of oil has in most studies given the maximum conversion of FAME. However, it is recommended to experiment in the range of 0.5% to 1.2% by weight of oil.

### Working temperature

It has been reported in the studies that the maximum conversion of esters was obtained in the temperature ranges of 55 °C to 63.5 °C. It should be noted that the boiling temperature of methanol is 64.7 °C.

### Inlet pressure

It has been observed that in the investigations presented in this article that the maximum FAME conversion was obtained in the pressure ranges of 2 to 3 bar.

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# Geometric parameter a

The maximum FAME conversion in the different studies was obtained in the condition that the higher the value of  $\alpha$ , the more orifices with the smallest possible diameter. The maximum conversions occur when  $\alpha > 2 mm$ -1.

# *Geometric parameter* $\beta$

The results reported from the different studies show that the lower the  $\beta$  value, under the condition of having the smallest possible diameter in the orifices, a FAME conversion of greater than 95% was obtained. The maximum conversions occur when  $\beta < 0.10$ .

# *Geometric parameter* $\beta_0$

It is observed in the different studies that the conversion increased with decreasing  $\beta 0$ , i.e. the reaction rate was higher at lower  $\beta_0$  values. The maximum conversions occur when  $\beta_0 < 0.25$ .

# Cavitation number Cv

Different studies reported in this research obtained a conversion higher than 90% when the value of cavitation number is equal to 0.3.

# Mechanical devices

Similarly, the mechanical devices that obtained better results in the different studies reported in this research are as follows:

The plate with 21 holes of 1 mm diameter obtained an ester conversion of 96.5 % in the studies [13, 14]. The operating parameters of [14] are inlet pressure of 2 bar, operating temperature of 60 °C while the constant parameters of the study of [13] are inlet pressure of 3 bar, operating temperature of 55 °C.

The plate with 25 holes of 2 mm diameter reported in the study of [9] obtained more than 95% ester conversion in 10 minutes. The operating parameters are inlet pressure of 3 bar.

The plate with 16 holes of 3 mm diameter reported in the study [16] obtained a 94 % ester conversion in only 20 minutes. The operating parameters are, an oil to alcohol molar ratio of 1:4.5, catalyst concentration of 0.55 wt.% oil.

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