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Review

Advancing Process Intensification with High-Frequency Ultrasound: A Mini-Review of Applications in Biofuel Production and Beyond

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Abstract: High-frequency ultrasound (HFU) is an ultrasound technology with a frequency higher than 1000 kHz. It has become increasingly recognized as an emerging process intensification technology in various fields, such as biofuel production, carbon dioxide absorption, and wastewater treatment. HFU is seen as a potential intensifier technology for biofuel production, as its mechanisms, such as cavitational phenomena, microstreaming, and fountain formation, can benefit biofuel production. Previous research has shown that HFU can decrease the reaction time required for biofuel production, aid in lipid extraction, increase carbon dioxide absorption rates, and be effective in destroying pathogens in wastewater treatment. However, despite the potential benefits, there are limited reports on the use of HFU technology for biofuel production, which has led to uncertainties and constraints in its industrial deployment. These constraints include equipment design, economic analysis, and safety concerns, which require further in-depth analysis. Despite these limitations, previous studies have shown promising results for the incorporation of HFU into various fields due to its unique characteristics and mechanisms. This paper presents a review of the theory and application of HFU for process intensification, with a focus on its potential for biofuel production. It also provides recommendations for the further exploration of the technology to overcome industrial deployment obstacles.

Keywords: biodiesel; biofuel; high-frequency ultrasound; microalgae cell disruption

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1. Introduction

Ultrasound is a type of mechanically oscillating sound wave that can be sustained by an elastic medium, such as air or water. Its frequency ranges from 20 kHz to 10 MHz, which is higher than the limit of human auditory perception (i.e., between 16 Hz and 20 kHz) [1]. According to Chuah et al. [2], ultrasound can be classified into two categories based on its frequency: high-frequency ultrasound (HFU), which has a frequency range of 1000–10,000 kHz, and low-frequency ultrasound (LFU), which has a frequency range of 20–100 kHz. While HFU is more commonly used in medical applications, its use in non-medical fields has been increasing due to its unique characteristics and mechanisms. The application of ultrasound technology has extended to different fields, such as carbon dioxide (CO_2) absorption [3–5], chemical synthesis [6], wastewater treatment [6,7], medical imaging [8], algae biomass disruption [9], biodiesel production [10], and the food industry [11,12].

Ultrasonic applicators can produce different forms of energy by converting electrical energy into heat and vibrational energy using an ultrasonic probe [13] or discs [14]. Figure 1 illustrates the transformation of electrical energy into various forms of energy using an ultrasonic applicator.

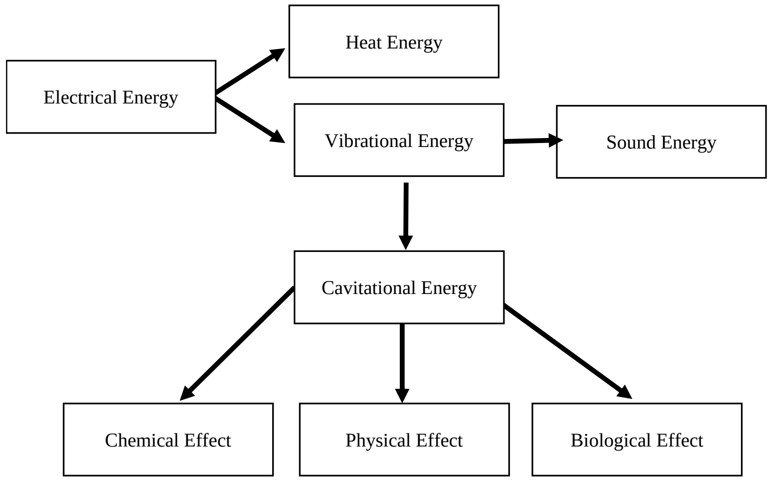

Figure 1. Energy transformation in ultrasonic applications.

Ultrasound technology can produce different types of effects, including biological effects, such as the break down of algal cell membranes for extraction [15]. Its physical effects, such as ultrasonic cleaning, emulsification, and atomization, have been extensively utilized in commercial products [16]. Atomization emits micro-droplets when the acoustic intensity exceeds a liquid-dependent threshold [17]. On the other hand, the implosion of bubbles during ultrasound irradiation can lead to the formation of highly reactive species, such as $OH^{\bullet}$, $HO_2{}^{\bullet}$, and $H^{\bullet}$, which contributes to the chemical effects of ultrasound technology [18]. The benefits of ultrasound technology extend to various fields, including wastewater treatment, where the vibrational energy from ultrasonic cavitation can help to break down pollutants. The cavitation phenomenon is the formation of bubbles that grow and collapse in a liquid that has been irradiated with ultrasound [19]. Figure 2 illustrates how bubble nuclei in water grow to reach resonance size through a rectified diffusion pathway and a coalescence pathway under the influence of an ultrasonic field. Resonance bubble size is important in ultrasound applications because it affects the effectiveness of the cavitation process. The size of the cavitation bubbles is influenced by the ultrasonic waves, with low-frequency ultrasound producing larger-diameter bubbles, resulting in stronger shear forces, while higher-frequency ultrasound generates smaller and more stable bubbles (stable cavitation) [20,21]. At lower frequencies, the implosion of cavitation bubbles is more intense compared to that at higher frequencies. At higher ultrasonic frequencies, a larger number of cavitational bubbles are formed, but the collapse intensity is lesser, and higher powers are needed for active bubble formation [22].

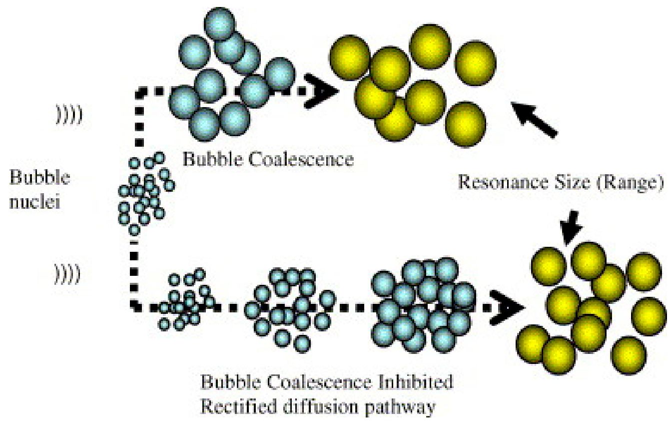

Figure 2. Schematic illustration of bubble nuclei growth in water through a pathway of rectified diffusion and coalescence under the influence of an ultrasonic field (adapted from Ashokkumar et al. [23] with permission from the publisher).

The frequency and cavitation intensity of ultrasound are inversely proportional to each other, as the formation of cavitation bubbles becomes more difficult with shorter rarefaction phases at higher frequencies [1]. At higher ultrasonic frequencies (>1 MHz), jet-like fountain formation and ultrasonic streaming force are enhanced [24]. Shokrollahi et al. [5] found that the fountain formation created under high-frequency ultrasound contributes to the enhancement of mass transfer process. Table 1 summarizes the differences between low-frequency ultrasound and high-frequency ultrasound, while Figure 3 shows the schematic drawing of the behavior of liquid under high-frequency ultrasound conditions.

Various parameters, such as power, frequency, reaction condition, operation mode, and mechanical vibrations, can affect the occurrence and intensity of ultrasonic effects [25]. Therefore, understanding the interaction between these parameters is crucial in enhancing the use of high-frequency ultrasound for different purposes.

Table 1. Comparison between high- and low-frequency ultrasound.

Low-Frequency Ultrasound	Characteristics	High-Frequency Ultrasound
20–100 kHz [2]	Frequency Range	1000–10,000 kHz [2]
Production of larger bubbles [20]	Cavitation Bubbles Size	Production of smaller bubbles [21]
Lesser [22]	Quantity of Bubbles	Higher [22]
Higher intensity [20]	Bubble Collapse Intensity	Lower intensity [20]
Lesser free radical formed [26]	Amount of Free Radical Formation	Higher free radical formed [26]
None or weak fountain formation [27]	Formation of Fountain (Figure 3)	Yes [28]

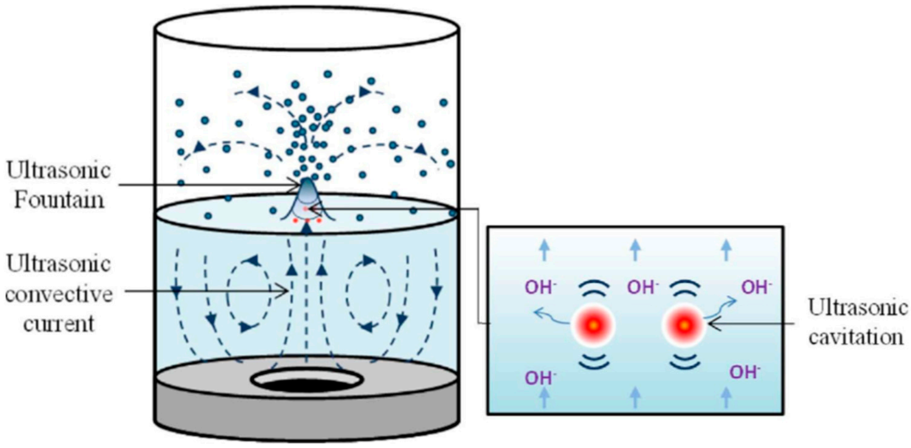

Figure 3. Schematic drawing of the formation of an acoustic fountain (adapted from Tay et al. [3] with permission from the publisher).

Schueller and Yang [29] observed that cavitation phenomena formed under lower frequencies aid in the desorption process more than absorption and adsorption processes. At higher frequencies, the cavitation phenomena are less intense, and fountain formation increases mass transfer coefficients for CO_2 absorption The role of high-frequency ultrasound is slowly gaining attention across different fields, particularly in biofuel production. The mechanism of HFU (e.g., cavitation phenomena, jet-like fountain formation, and ultrasonic streaming force) plays a major role in the formation of eddies, turbulence, and shear forces that enhance the transesterification process [30,31], directly influencing the production of biodiesel. Meanwhile, cavitation phenomena aid in the breakdown of algal cell walls, improving the extraction process of products such as biofuel.

According to Hasan et al. [32], research conducted on biofuel production increased from the year 2005 onwards, with an increase in the number of published articles on the subject being evident over the last 16 years, and 2019 marked the highest rate of publications. Climate change and increasing energy demands are major contributors to the expansion of biofuel economics, and biofuel plays a vital role in addressing the increasing energy demand as a potential energy source [33]. However, the production of biofuel is still not economically viable, and more research and development for technological enhancement is required [34].

Various intensifying technologies have been applied to the production of green biofuels. Still, each technology possesses different advantages and disadvantages. LFU is well-established for biodiesel production [35], but there is still a lack of reports on HFU as an intensification technology for this application [31]. Therefore, a great deal of uncertainty regarding the relationship between HFU technology and biofuel production exists. To our best knowledge, there is currently a lack of comprehensive literature on the application of high-frequency ultrasound in biofuel production. Hence, this review paper aims to assess its current progress and the issues that need to be studied for the future prospects of biofuel production.

2. High-Frequency Ultrasound Technology for Biofuel Production

Reliance on fossil fuels for energy and transportation is having detrimental effects on the environment. The combustion of fossil fuels releases greenhouse gases, such as CO_2, sulfur dioxide (SO_2), and nitrogen oxide (NO_x), which contribute to global warming. This situation is alarming as society and industries continue to rely heavily on the use of fossil fuels in their power plants and as a primary source of energy for transportation fuel. Therefore, clean, sustainable, and renewable energy sources are critically needed to reduce the negative environmental impact and ensure a continuous supply of energy for the future. Biofuel, a new potential energy source derived from different renewable feedstocks, may reduce the reliance on the usage of non-renewable fossil fuels. Demirbas [36] denoted biofuel as liquid, gas, and solid fuels produced from biomass. Meanwhile, according to Raboni et al. [37], biofuel comprises any product obtained from biomass and this includes biodiesel, biofuels, biogas, bioethanol, and bio-methanol. Biofuel production is highly supported by various countries, including Brazil, Germany, Switzerland, and Sweden, as efforts have been intensified in order to reduce the reliance on fossil fuels as an energy source.

Biodiesel, or fatty acid methyl esters (FAME), is regarded as a potential source to replace the use of fossil fuels and has been studied globally [38]. Biodiesel can be potentially utilized as an energy resource in the coming future, as it can be produced from numerous potential feedstocks, such as microalgae [38–41], waste cooking oil [42,43], animal fat [44,45], by products such as rice bran [46], and various vegetable seeds oil [47–50]. According to Mahbub et al. [51], the usage of biodiesel has several benefits, including a reduction in CO_2 and carbon monoxide (CO) emissions by 8–41%, based on life cycle assessment studies. Previous research has proposed the use of biodiesel as a complete or partially mixed alternative for diesel engines, since biodiesel usage reduces exhaust emissions as the composition of biodiesel contains less carbon, water, and sulfur with a higher amount of oxygen than conventional petroleum [52]. Biodiesel can be introduced into diesel engines without modifications because the current diesel engines are compatible with biodiesel [53].

2.1. High-Frequency Ultrasound-Assisted Transesterification Process for Biodiesel Production

Transesterification is a more popular method for producing biodiesel compared to other methods, such as the direct use of blended oils, micro-emulsion of oils, and thermal cracking (pyrolysis) of oils [30,54]. This process converts feedstock (i.e., oil) into methyl or ethyl esters using an alcohol source, such as methanol or ethanol, and a catalyst. Glycerol is produced as a side product during the reaction [55]. Transesterification is carried out at a mild temperature [56] and requires simpler conditions in comparison to other methods. For example, micro-emulsification requires different alcohol solvents with colloidal microstructures [57], while pyrolysis requires high temperatures ranging from 300 °C to 1300 °C, which may lead to changes in the chemical structure of the compounds [58]. Therefore, transesterification is more economically feasible and simpler, making it the preferred method for producing biodiesel compared to other methods [59].

However, the production of biodiesel via the transesterification process still faces some challenges. Firstly, the heterogenous nature of the reactants (alcohol and vegetable oil) does not form a homogenous mixture [43,60], which requires intensive mixing processes to increase the mass transfer rate, resulting in a higher power consumption. Additionally, the viscosity of the different feedstocks used may differ, leading to higher or lower agitation requirements. Moreover, the two-way reaction of the transesterification process requires a higher alcohol-to-oil molar ratio. An excess of alcohol aids in product formation instead of reactant formation, resulting in higher conversion rates that directly lead to increasing biodiesel expenditure [61].

Ultrasound-assisted transesterification is considered an approach to enhance biodiesel production. The characteristics of ultrasound technology, such as cavitation bubbles, microstreaming, and fountain formation, enhance mixing between heterogenous mixtures, reducing the reliance on additional mixing processes. The application of ultrasound technology allows for higher reaction rates and lower alcohol-to-oil molar ratios [62,63]. LFU has been widely researched for transesterification processes [15,30,64,65] due to the high-intensity collapse of cavitation bubbles, which aids in overcoming the mass transfer limitation. However, there is still a lack of reports on the use of HFU in the transesterification process [31], which requires further exploration.

Recent studies have shown that high-frequency ultrasound technology is beneficial for biofuel production. The phenomena of cavitation, jet-like fountain formation, and ultrasonic streaming force are prominent characteristics that facilitate the mixing of immiscible liquids, such as alcohol and oil. However, it remains unclear which of these characteristics is more advantageous for mixing immiscible liquids. Oliveira et al. [31] studied the influence of low power (3–9 W) and high frequency (1 MHz and 3 MHz) without an external heating source on the transesterification of soybean oil. The results showed that ultrasound-assisted transesterification increased soybean oil conversion from 48.7% to 79.5% when the alcohol-to-oil ratio was 6:1 at 1 MHz and 3 MHz, respectively, while a molar ratio of 8:1 at 1 MHz and 3 MHz achieved a conversion of 59.5% to 84.6%, kept at the same reaction time of 40 min. Within 10 min, the HFU of 3 MHz achieved a conversion of 79.8% compared to the HFU of 1 MHz, at a conversion rate of 53.6% after 20 min at a 8:1 M ratio. This shows that HFU along with excess methanol can result in a higher reaction rate within a shorter time period, as the excess methanol shifts. This is because the diameter of the cavitation bubbles produced by ultrasonic frequency is dependent on the frequency, causing variations in bubble size [31]. Since the size of the cavitation bubbles is inversely proportional to ultrasonic frequency [66,67], this may, to a certain extent, facilitate higher collision rates between reactants, leading to a greater conversion rate (Figure 4).

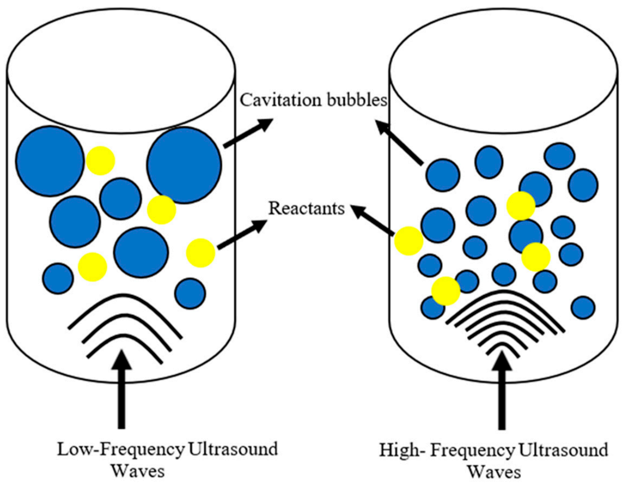

Figure 4. Differences between the amount of cavitation bubbles between LFU and HFU.

Another study conducted by Mahamuni and Adewuyi [18] focused on the synthesis of biodiesel via the ultrasound-enhanced base-catalyzed transesterification of soybean oil using a multifrequency ultrasonic reactor. The study revealed that high-frequency ultrasound at 1.3 MHz resulted in more than 90% conversion within 30 min. Aghbashlo et al. [35] conducted a separate study on the development and evaluation of a high-frequency piezoelectric-based ultrasonic reactor to intensify the transesterification reaction. The study noted that the transesterification of waste cooking oil (WCO) achieved the highest conversion efficiency of 99.3% at 10 min of ultrasonication with an elevated alcohol/oil molar ratio of 8:1. According to the authors, increasing the ultrasonification time and liquid media temperature improved the conversion efficiency. The longer exposure time allowed for an enhanced emulsification between the alcohol and oil phases due to the increased number of bubbles produced using HFU. The study suggested that HFU may outperform LFU in terms of conversion efficiency, energy requirement, and processing time. Table 2 provides a summary of the reaction conditions for the HFU-assisted transesterification studies conducted to date.

Table 2. Reaction conditions for high-frequency ultrasound-assisted transesterification.

	Authors		
	Oliveira et al. [31]	Mahamuni and Adewuyi [18]	Aghbashlo et al. [35]
Feed	Soybean oil		Waste cooking oil (WCO)
Ultrasonic Reactor Type	Transducer		Piezoelectric-based ultrasonic reactor
Catalyst	Potassium hydroxide (KOH)		
Alcohol	Methanol		
Conditions	Study was conducted at the frequencies of 1 MHz and 3 MHz with alcohol-to-oil molar ratios of 6:1 and 8:1, respectively, at a reaction time of 40 min (without external heating or mechanical stirring).	Study was conducted at the frequencies of 323 kHz, 581 kHz, 611 kHz, and 1.3 MHz with an alcohol-to-oil molar ratio of 6:1 and a power between 12 and 223 W, with a reaction time ranging from 60 min to 180 min.	Study was conducted at the frequency of 1.7 MHz with alcohol-to-oil molar ratios of 4:1, 6:1, and 8:1 and a power of 31 W, with the reaction times of 6, 8, and 10 min.

HFU has the potential to aid in biodiesel synthesis, particularly in enzyme-catalyzed biodiesel production. However, most studies have focused on acid- and base-catalyzed transesterification processes [22,68]. The lack of research on enzyme-catalyzed biodiesel production was highlighted by Veljković et al. [69], where it was noted that enzyme-catalyzed ultrasound-assisted transesterification could be beneficial due to its advantages of higher selectivity, better energy efficiency, and lesser generation of by-products. Nonetheless, the impact of HFU on enzyme-catalyzed biodiesel production needs to be investigated further.

Additionally, different feedstocks, such as soybean oil and waste cooking oil, may affect the HFU mechanism due to their viscosity, which can hinder the cavitation process and require higher power supplies, leading to additional operating costs. It is expected that, as viscosity increases, the interactions between molecules become stronger, further hindering cavitation [70], which may reduce the efficiency of HFU during transesterification, necessitating the use of alternative solutions.

2.2. High-Frequency Ultrasound on Microalgal Cell Disruption for Biofuel Production

Microalgae, which are photosynthetic microorganisms that require minimal growth requirements, are considered a promising source of lipids, proteins, and carbohydrates. They can produce large quantities of bioproducts in a short time and, thus, are an ideal feedstock for biodiesel production. Microalgae are environmentally and economically advantageous due to their high growth rate and biomass productivity, and their ability to accumulate bioproducts, such as carbohydrates and lipids, under nutrient-limited conditions [71]. Unlike crops, which require growing cycles ranging from three months to three years [72], microalgae have a short growth cycle, and bioproducts such as lipids can be harvested in just 3–5 days. Once the microalgae biomass is cultivated, harvesting and dewatering are carried out, followed by lipid extraction from microalgae [73]. The use of ultrasound pre-treatment is known to increase the lipid yield from the microalgal biomass, as it aids in the disruption of biomass for lipid extraction [74]. Ultrasonic irradiation breaks down the microalgal cell wall and reduces microalgal particle size, leading to a better release of chemical content, thus enhancing extraction efficiency [75]. Figure 5 illustrates the use of HFU technology to aid microalgal cell lysis.

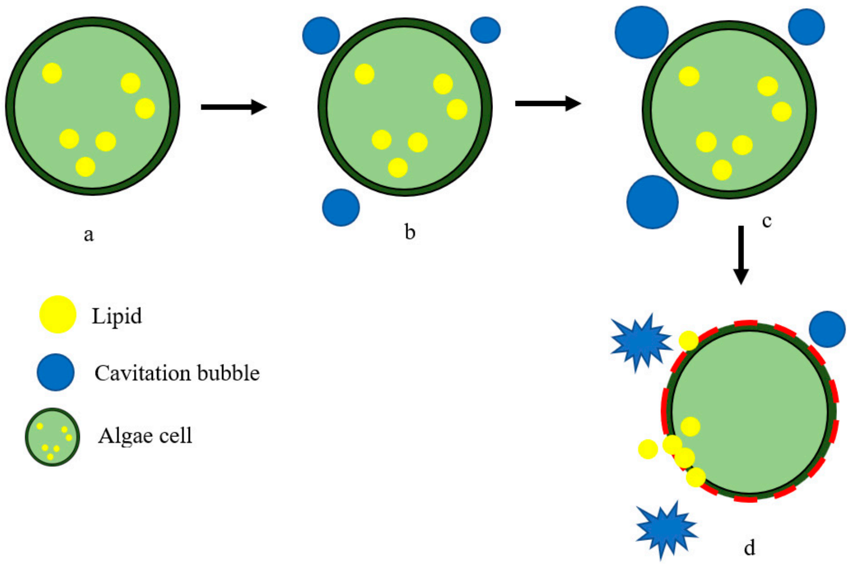

Figure 5. Disruption of algal cell using ultrasound: (**a**) condition of algal cell prior to disruption; (**b**) formation of cavitation bubbles; (**c**) growth of cavitation bubbles; and (**d**) breakdown of algal cell wall and release of lipids.

The incorporation of high-power, low-frequency (i.e., 100 W and 33 kHz) ultrasound as pre-treatment in the transesterification of microalgal oil showed an increase in biodiesel yield by almost 40% compared to the mechanical stirring method [76]. While previous studies have focused on low-frequency ultrasound for algal cell disruption [77–82], high-frequency ultrasound for cell lysis has only recently been explored [9]. Several studies have been conducted to investigate the use of high-frequency ultrasound for the disruption of microalgal cells to aid in lipid extraction [9,78,83]. Wang et al. [9] evaluated the effectiveness of high-frequency focused ultrasound (HFFU) on the cell lysis of the microalgae *Scenedesmus dimorphus* and *Nannochloropsis oculata*. The effects of HFU at 3.2 MHz and 40 W input power on microalgal cell were evaluated in the study. The cell size for both *S. dimorphus* and *N. oculata* was found to decrease after HFFU treatment, and the lipid extraction for *S. dimorphus* increased, ranging between 49% and 113%, as the processing time increased from 1-min HFFU to 5-min HFFU. For *N. oculata*, the relative lipid extraction increased from 25% to 49%, which was lower compared to the increase observed for *S. dimorphus*. Hence, this indicated that the effectiveness depends on the cell (biomass) being treated. The results from the study revealed that using HFFU for cell disruption to assist lipid extraction in microalgae is a feasible method. On the other hand, Yamamoto et al. [78] investigated the effect of ultrasonic frequency ranging from 20 to 1146 kHz and different powers on the disruption of microalgal cells. HFU was found to have a more prominent effect on microalgal cell disruption compared to LFU, and the frequency required for disruption depends on the specific characteristics of the cell. The effect of HFU on *Chlamydomonas concordia* and *Dunaliella salina* was investigated at the different frequencies of 20, 580, 864, and 1146 kHz, as well as at the different power inputs of low (~3 W), medium (~20 W), and high (~60 W). The results showed that the highest frequency for *C. concordia* cell disruption was 1146 kHz, while there was an insignificant difference for *D. salina* at 580 kHz, 864 kHz, and 1146 khZ. However, the higher ultrasound frequency of 580 kHz showed better results than the ultrasound frequency of 20 kHz. The authors explained that the bubble radius (μm) was significantly reduced from 160 μm, at a frequency of 20 kHz, to 3.3 μm at a high-frequency 1146 kHz. The resonance radius of 3.3 μm was almost similar to the size of *C. concordia*, ranging between 3 and 6 μm. Hence, HFU aids in the breakdown of vacuoles inside the cell as the size of the vacuole is similar to the resonance radius of the cavitation bubbles.

Varied frequencies for cell disruption are required for different species of algae, which can be explained by the distinct cell characteristics, as some species have thicker cell walls. Kurokawa et al. [83] conducted research on the effectiveness of ultrasonic waves with frequencies ranging from 0.02 MHz to 4.3 MHz on the disruption of algae cells using *Chaetoceros gracilis*, *Chaetoceros calcitrans*, and *Nannochloropsis* sp. The results showed that higher frequencies led to a higher cell disruption for all three microalga species. The most prominent cell disruption was reported at 2.2, 3.3, and 4.3 MHz for *C. gracilis*, *C. calcitrans*, and *Nannochloropsis* sp., respectively. *C. gracilis* showed approximately 100% of cell disruption in 2 min at 2.2 MHz, while at the lower frequency of 0.4 MHz, more than 60% of the cell remain undisrupted after 10 min. *C. calcitrans* showed, at the higher frequency of 3.3 MHz, almost 100% of cell disruption was achieved after 2 min, while at the lower frequency of 0.4 MHz, 60% of the cell remained undisrupted after 10 min. The same pattern was observed for *Nannochloropsis* sp., for which, at 4.3 MHz, nearly 90% of the cell was ruptured, while at lower frequency of 0.4 MHz, only 10% of the cells were ruptured. The research results suggest that HFU is more efficient than LFU at cell disruption, rupturing nearly 90% of cells in all three species studied. The ideal frequency for cell disruption differs among species and is dependent on frequency. This is due to the fact that the ideal frequency is determined by the mechanical properties of the cell, such as the thickness of the algal cell wall.

HFU technology can aid in the extraction of bioproducts from microalgal cells by disrupting their outer cell wall layer. The use of ultrasound is considered more environmentally friendly than using toxic solvents, such as chloroform, which can be harmful to the environment and human health. Since the bioproducts are synthesized inside the cell, the disruption of the cell wall (outer layer of the cell) is essential. However, the efficacy of this technology depends on the unique biological characteristics of the microalgal species, as some species have thicker cell walls that require higher frequency HFU for effective disruption, while others have thinner cell walls that may be susceptible to excessive disruption. An excessive disruption of the cell membrane may cause the bioproducts to disintegrate. Therefore, optimizing the conditions for HFU will require further research and investigation, particularly in the context of microalgae-based biofuel production.

3. Other Applications of High-Frequency Ultrasound

3.1. Chemical Absorption of CO_2

The technology for CO_2 removal and capture is crucial in various applications, such as power plants, natural gas purification, and carbon capture and storage (CCS) [3,84]. Post-combustion CO_2 removal is required for stack gas treatment produced from combustion plants [3,85]. CO_2 is captured from the pre-combustion treatment of power generation stations and reused in the industry [86]. In natural gas reservoirs, CO_2 capture could be used for enhanced oil recovery as natural gas reserves can contain up to 87% of CO_2 [3]. To facilitate the transportation of natural gas, it is necessary to reduce the CO_2 levels in the gas to specified limits [87]. Controlling the release of CO_2 into the atmosphere is also important to prevent climate change, as CO_2 is one of the major greenhouse gases. Climate change can lead to shifts in global temperature patterns, resulting in catastrophic events worldwide.

Various technologies can be used for carbon capture, including absorption [88], adsorption [89], packed bed columns, mechanical agitators, and membranes [84]. Nonetheless, the limitations of these technologies have resulted in the search for alternative methods that can improve carbon capture [84]. The use of conventional packed bed reactors for carbon capture is space-consuming [84,90]. Membrane technology with larger surface contact areas for mass transfer is a promising alternative. However, the costing, pressure, and selectivity are some of the issues to be addressed when using membrane technology [91]. Several studies have examined the potential of ultrasound for improving the absorption and desorption of CO_2 for capture purposes. However, most of these studies have utilized low-frequency ultrasound in the range of 20–500 kHz [4]. Tay et al. [92] reported that the generation of small gas bubbles by LFU nullified the physical absorption process. As a result, HFU was studied as an alternative technology to improve CO_2 absorption in a water batch system under increased pressure conditions. HFU was found to provide high mass transfer enhancement, which is beneficial for ultrasound absorption processes.

Tay et al. [84] conducted a study on the high-frequency ultrasonic-assisted chemical absorption of CO_2 using monoethanolamine (MEA) at the frequency of 1.7 MHz. The study showed that high-frequency ultrasonic irradiation was beneficial for CO_2 absorption, increasing the absorption rates up to 60 times compared to absorption without the HFU irradiation. The increase in absorption was attributed to the physical enhancement effect of convective dynamics and fountain formation. The authors found that temperature did not affect the CO_2 absorption rate, as the absorption rate is impacted by the transportation of the reactant to the fountain layer, which is affected by the ultrasonic force. Therefore, elevated temperatures did not contribute to increased CO_2 absorption in the solvent. HFU-assisted absorption helps with CO_2 capture by improving the mass transfer coefficient and the compact design, which reduces the need for large spaces, allowing the technology to be used even in limited space conditions [92].

Meanwhile, Yusof et al. [4] conducted a study on the mass transfer intensification of CO_2 absorption in MEA using high-frequency ultrasonic technology in a continuous system. They investigated the removal of CO_2 from natural gas using high-frequency ultrasonic (1.7 MHz) absorption technology, which showed effective mass transfer performance and high operating flexibility. The results of the study showed that the use of ultrasonic irradiation contributed to a seven-times-higher mass transfer compared to CO_2 absorption without the use of ultrasonic irradiation. This finding is consistent with those of Tay et al. [84], where CO_2 absorption was enhanced with high-frequency ultrasound usage. The study found that the fluid motion (acoustic streaming) is directly proportional to the ultrasonic power [92]. As the ultrasonic power increases, the fluid motion also increases, resulting in an increase in the fountain flow rate, which increases the contact area. Additionally, the fluid motion contributes to the agitation of the liquid.

According to Tay et al. [92], high-frequency ultrasonic power is more favorable for CO_2 absorption in high-pressure water batch systems. Low-frequency ultrasonic (LFU) power generates sound waves with insufficient physical force to break through the surface of the liquid layer, making it unsuitable for CO_2 absorption. The mass transfer coefficient is directly proportional to the ultrasonic power, and an increase in ultrasonic power leads to an increase in mass transfer coefficient due to the presence of fluid motion (streaming effect) and liquid fountain formation under high-frequency ultrasonic irradiation. The streaming turbulence generated by the ultrasonic power generates a convective dynamic current that helps in the thorough mixing of the liquid, contributing to an enhanced CO_2 absorption [24]. Therefore, HFU has greater potential for CO_2 absorption due to the presence of streaming turbulence and the ultrasonic fountain, making it a more efficient technology for CO_2 capture.

3.2. Inactivation of Harmful Microorganisms in Water

Water is a crucial resource in daily human activities, including drinking, washing, cooking, and industrial processes. In the food industry, it is vital to have clean water that is free from pathogens and microorganisms, which can pose health risks to consumers. Disinfection is a crucial step in maintaining water quality, and chlorine disinfection is commonly used to eliminate microorganisms through chemical oxidation [93]. Despite the effectiveness of chlorine disinfection in deactivating microorganisms present in water, there are some issues with its continuous usage. One of the problems is the presence of chlorophenols, which results in unpleasant odors in the water [94]. Additionally, some pathogens, such as *Cryptosporidium parvum*, are known to be tolerant to chlorine sanitation [95,96]. The continuous usage of disinfection methods such as chlorine and antibiotics may also lead to higher resistance in pathogens and other harmful microorganisms [97]. To address the issue of pathogen resistance to disinfection methods such as chlorine and antibiotics, higher concentrations of these chemicals may be necessary. As a result, finding alternative methods to inactivate pathogens is an ongoing concern in water treatment. Ultrasound has recently been investigated as a potential solution, as it has been shown to break down the cell membranes of pathogens, resulting in cell lysis, damage to DNA molecules, and organelle damage that ultimately leads to their inactivation [98]. Moreover, the implosion of bubbles during ultrasound irradiation has the potential to generate free radicals, which can assist in the water treatment [99]. Olvera et al. [98] reported that microbial pathogens are susceptible to inactivation due to the penetration of free radicals from cavitation collapse. Additionally, other studies [97] suggest that the increase in temperature, pressure, and free radical action also contribute to the deactivation of pathogens.

Olvera et al. [98] conducted a study on the use of ultrasonic treatment at 1 MHz ultrasound to inactivate *C. parvum* oocysts in water. The study found that, within 2 min of ultrasound irradiation, the cell viability of the samples was reduced by up to 87.82%, and, by 4 min, the cell viability was reduced to 94.02%. In a study by Gao et al. [94] high-frequency ultrasound (850 kHz) was used to inactivate bacteria and yeast. The results showed that the inactivation rate was more than 99%, mostly due to the formation of free radical action attacking the bacterial cells. The inactivation of bacteria is mostly contributed by the formation of free radical action attacking the bacterial cells. HFU was found to be effective in deactivating more than 99% of bacteria. However, the yeast *Aureobasidium pullulans* was found to be highly tolerant to ultrasound treatment, while most bacterial cells were ruptured within 10 min of treatment. The difference in tolerance could be attributed to the thickness of the cell wall of each species, with bacterial cells having a thinner cell wall compared to yeast cells.

Brayman et al. [100] investigated the effectiveness of 1 MHz ultrasound on the inactivation of planktonic *Escherichia coli*. After 10 min of a high-frequency ultrasound treatment, approximately 80% of the bacteria were inactivated, and after 20 min of the treatment, the inactivation percentage increased up to 95%. The authors suggested that the inactivated cells were lysed into small fragments and that the tiny holes observed on the surface of the bacterial cell membrane could be caused by cavitation jets. Despite its potential for water disinfection, ultrasound treatment may not be effective against all pathogens, as demonstrated by the resistance of the yeast *A. pullulans*. Therefore, combining HFU with other disinfection methods may be necessary to improve water quality.

4. Challenges to Be Addressed in Scaling up HFU for Biofuel Production

The use of HFU has shown promise in various industries, such as biofuel production, carbon dioxide absorption, and water treatment. Optimizing the conditions for these applications could have a positive impact on these industries. For instance, this technology could improve the commercial production of biofuels by increasing production yields. However, there is still a need to fill research gaps and fully understand the potential applications of HFU technology, especially for the enhancement of large-scale biofuel production.

Scaling up HFU for biofuel production poses a significant challenge due to a lack of fundamental or mechanistic data for up-scaling processes [6]. Equipment design, such as vessel size and configuration for HFU vessels, must also be taken into consideration. The height of the vessel should be sufficient to allow for the occurrence of fountain formation, which is affected by different factors, such as input pressure and the characteristics of the liquid [101]. A previous study on the relationship between water depth and acoustic fountain formation found that the optimal depth was directly proportional to the radiation area of the ultrasonic transducer [102]. This is due to the impact of varying ultrasonic intensity. Asakura and Yasuda [16] reported an observation of the quenching phenomenon, wherein the reaction rate decreased after reaching a maximum rate, even with an increase in ultrasonic power. The higher the frequency and sample volume, the greater the ultrasonic power required for the reaction quenching. The reason behind this is that, as the ultrasonic power increases, the traveling wave field increases while the standing wave field decreases, resulting in a reduction in the chemical reaction field. Hence, the authors suggested that a standing wave field with a stable and large volume should be established in developing a highly efficient ultrasound system. Thus, additional research is required to examine the impact of ultrasound frequency and power with regards to vessel dimensions for an optimal reactor configuration that achieves the desired HFU characteristics.

The thickness of the vessel wall, types of transducer used, and diameter of the ultrasonic reactor may also play a major role in equipment design. Fang et al. [103] found that different sonotrode tips have varying effects on the pattern of acoustic flow and the efficiency of cavitation patterns. Nanzai et al. [104] investigated the influence of the reaction vessel diameter on sonochemical effectiveness and cavitation dynamics. According to their findings, an increase in vessel diameter from 20 mm to 90 mm led to a rise in the reaction yield, but when the diameter was further increased to 120 mm, the yield decreased. The authors suggested that active cavitation bubbles were formed at specific zones, and in a larger diameter reaction vessel, bubble nuclei that had not grown to the resonance size were able to escape from the sonication zone to the non-sonication zone, resulting in their dissolution. Consequently, the number of active cavitation bubbles decreased in the larger diameter reaction vessel, which had a negative impact on the reaction process. In addition, when it comes to large-scale operations, it is necessary to use multiple transducers. It is essential to investigate the selection of the appropriate type of transducers and their optimal arrangement for the targeted biofuel production purpose. It may also be beneficial to incorporate mechanical stirring with HFU to promote synergistic effects, particularly in heterogeneous systems, which could result in a more uniform bulk mixing [105]. Hence, an in-depth analysis of various details is necessary for designing equipment for HFU-assisted processes to ensure their viability. Additionally, the mixing pattern between the immiscible liquids (e.g., oil and alcohol) should also be investigated based on the liquid's characteristics. It would also vary with liquid volume and reactor size. Employing particle image velocity (PIV) to observe the mixing or flow pattern may help in the mass and heat transfer prediction for the design of the vessel.

In the transesterification process, the type of catalyst used and the amount of catalyst required may vary depending on the feedstock utilized. The feedstock's characteristics, such as viscosity, must also be taken into account as higher intramolecular forces can impact cavitation phenomena [70]. Higher ultrasound frequency results in a shorter acoustic wavelength, which, in turn, would affect the bubble formation. Under a water medium, the range of ambient radius for an active bubble becomes narrower as the frequency is increased from 0.2 MHz to 1 MHz. At 1.5 atm, the optimal bubble radius reduced from 5.0 μm to 1.85 μm with the mentioned increase in frequency. At higher pressure of 2.5 atm, the reduction in size for the optimal bubble radius was even more pronounced, as it reduced from 8.0 μm to 1.5 μm [106]. Different characteristics and the hydrophicity of molecules, for systems other than water as the medium, could result in the retardation of bubble–bubble coalescence as a pathway for the growth of cavitation bubbles [23]. Therefore, a comprehensive understanding of the complex cavitation process in transesterification system is crucial to enable a more precise control of chemical reactions and improving the reaction efficiency. As different factors are interrelated, with the variation acoustic wavelength at different frequencies, it is important to understand the relationship between these factors, be it through modeling or experiment-based research.

Additionally, it is worth noting that most studies were conducted in small-scale laboratory batches. Therefore, the application of high-frequency ultrasound in large-scale production remains uncertain in terms of duration and electricity consumption. Moreover, the impact of different mechanism characteristics on the transesterification process should be investigated to determine which mechanism (e.g., bubble implosion, fountain formation, and acoustic streaming) may promote the reaction. At the moment, it is still unclear which mechanism is the main contributor towards the enhancement of the process. The primary mechanisms for microalgal cell disruption using ultrasound are acoustic cavitation, bubble implosion, heat, pressure, and free radicals induced by ultrasound [107]. The direct effects of ultrasound, such as radiation force and acoustic streaming, have insignificant impact on microalgal cell disruption. This is partly due to the small size of algae compared to the wavelength of ultrasound. For example, the estimated wavelength of the highest frequency ultrasound at 4.3 MHz in water is 350 μm, which is 270 times greater than the 1.3 μm radius of a *Nannochloropsis* sp. cell. Consequently, the ultrasound is not scattered by the algae [83]. On the other hand, Simon et al. [17] found that cavitation bubbles play a significant role in fountain formation and atomization. The atomization threshold increases with increasing shear viscosity as it damps surface instabilities. An increase in viscosity leads to the higher absorption of the standing spherical wave, resulting in a reduction in the acoustic pressure level at the drop center, which in turn increases the cavitation threshold and decreases the heating of the drop center. Meanwhile, as the temperature of liquid is increased, the cavitation threshold is affected, resulting in time reduction for the commencement of atomization. For the three frequencies tested (2.165 MHz, 1.04 MHz, and 155 kHz), the authors also observed that the diameter of the drops emitted from the fountain approximately equalled the ultrasonic wavelengths. With various raw material mixtures applicable for biodiesel production, it is important to investigate the appropriate ultrasonic frequency (which would result in the corresponding wavelength size), intensity, and duration based on the specific characteristics of the biofuel production system. This is due to the fact that it involves a mixture of liquids with varying viscosities, such as alcohol–oil blends utilized in transesterification.

To assess the viability of HFU in the industry, an economic analysis is necessary to be conducted. Since most studies are conducted on a laboratory scale, the unit cost estimate of the actual process is not thoroughly explored. To date, the economic analysis of such a process is still not available, even for a pilot scale. The availability of such data would eliminate the need for unavoidable assumptions in the unit cost estimation of the process [108]. Other important factors for scaling up, such as operational costs and detailed analysis of energy requirements, have not been fully investigated either. Consequently, the economic analysis may be considered incomplete or insufficient in comparison to the actual costs required. This creates a gap in determining the practicality of utilizing HFU in the industry since there is no clear differentiation between the feasibility of employing HFU for biofuel production versus other intensification technologies.

Furthermore, there is a lack of comprehensive reports on the safety analysis for utilizing HFU in specific fields, such as biofuel production. Since acoustic energy can disperse into the surrounding environment, there is a risk of energy loss to the external environment. This poses a threat to the safety of workers since long-term exposure to high-frequency wavelengths can cause hearing impairments [109]. Additionally, as cavitation collapse can cause extreme conditions, such as high temperature (up to 5000 K) and high local pressure (up to 1000 atm) [110], a safety analysis must be performed if the project is carried out in a pilot plant or on a bigger scale. This ensures the safety of workers, and proactive measures for preventing accidents can be implemented effectively. Safety analysis is also critical in determining preventive measures or safety precautions to be implemented. For instance, the prolonged use of ultrasound for the transesterification process may cause the overheating of equipment, requiring additional cooling procedures to prevent its breakdown and overheating. Similarly, the feasibility of employing HFU in other processes, such as CO_2 absorption and the inactivation of microorganisms in water treatment, is expected to be clarified by revealing these underlying fundamental mechanisms and conducting detailed economic, safety, and effectiveness analyses of the processes.

5. Conclusions

The primary objective of this mini-review paper was to highlight the potential benefits of HFU technology as a process intensifier in biofuel production as well as the further studies that can be conducted to bridge its potential towards scaling-up. The cavitation phenomena (bubble implosion), formation of jet-like fountain, microstreaming, and free radical formation make this technology advantageous for various fields, and there is growing interest in its application. The effect of HFU mechanisms enhances the mixing of immiscible liquids during the transesterification process and cell lysis, resulting in a higher biofuel production. In addition to improving biofuel production, this technology is also suitable for other areas, such as carbon dioxide absorption and the inactivation of harmful microorganisms in water. A more concrete fundamental understanding on the effect of the technology is necessary for a suitable equipment design at a larger scale. Conducting thorough safety and economic analyses can also reduce uncertainties and enable the further scaling up of the technology for deployment in the industry.

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