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Continuous and pulse sonication effects on transesterification of used vegetable oil

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ABSTRACT

This study reports on the effects of direct application of continuous and pulse sonication on transesterification reaction of used vegetable oil. Specific to this research, thermal effects of ultrasonics in transesterification reaction without external conventional heating along with the effects of different ultrasonic intensities and power densities were reported. Two process parametric evaluation studies were conducted to compare the effects of continuous and pulse sonication. These included methanol to oil ratio, catalyst concentration and reaction time effects on the transesterification reaction. For continuous sonication, a catalyst amount of 0.5% (wt/wt), methanol to oil ratio of 9:1 was sufficient to complete the transesterification reaction in 1–2 min at a power output of 150 W with a biodiesel yield of 93.5%. For pulse sonication, a maximum biodiesel yield of 98% was achieved at 2.5 min of reaction time, 9:1 methanol to oil ratio, and 1.25% catalyst. Generally, higher biodiesel yields were observed for pulse sonication compared to continuous sonication under any given process condition. Power density and ultrasonic intensity tests revealed that biodiesel yields were more sensitive to continuous sonication due to intense mixing. A plug-flow or contact-type reactor design may improve overall ultrasonic utilization in the transesterification reaction under continuous sonication.

1. Introduction

Biodiesel production from renewable feedstock is a promising alternative to combat the present dual crises of fossil fuel energy source depletion and the associated greenhouse gas emissions due to their rapid consumption. For sustainable biodiesel production, feedstock should be available in sufficient quantities at affordable prices in all seasons [1]. Waste cooking oils, though not available in substantial quantities, meet these criteria and provide for the local needs at reasonable supplies. Apart from that, they are rich in triglycerides suitable for biodiesel production, which eliminate the need for extraction step and environmental pollution associated with the process [2].

With feedstock like oil originating from fresh or used resources, the most commonly used method to produce biodiesel is "Transesterification". Transesterification reaction involves replacement of an organic group of an ester with the organic group of an alcohol [2]. Transesterification of oil feedstock can be performed in the laboratory by conventional heating and mixing methods such as jacketed reactors, oil and sand baths mixing provided by mechanical stirring [2]. Recently, non-conventional methods like microwaves [3–7] and ultrasound [2,8] were investigated to evaluate their process intensification effects. Ultrasound, a sophisticated non-conventional technique, was reported to enhance the transesterification reaction at reduced energy consumption and reaction times. Ultrasonic mixing can induce effective emulsification and increase mass transfer rates among the reaction compounds. Thus, the reaction rates under ultrasonic conditions can be much higher than the reaction rates under conventional stirring conditions [9–12]. Fig. 1a shows the different forms of energy that can be generated through ultrasonic applicators. When electrical energy is passed through an ultrasonic probe, it is mainly converted into two forms as heat and vibrational energy (i.e. mechanical energy). This vibrational energy is converted to cavitational energy in the reaction medium which is identified as a bubble effect and some of the vibrational energy is lost through sound reflection via harmonics and subharmonics [13]. The cavitational energy is further converted into chemical, physical, and biological effects depending on the application and reaction environment. In biodiesel process applications, chemical, and physical (thermal) effects are evidenced by many researchers through a significant rise in the reaction temperature, reaction yields, intense mixing [8,14–18] and in some applications through destruction of cell membranes for oil extraction such as in algae biodiesel production which can be considered a biological effect [19–24]. Lin et al. [25] attributed three important phenomena

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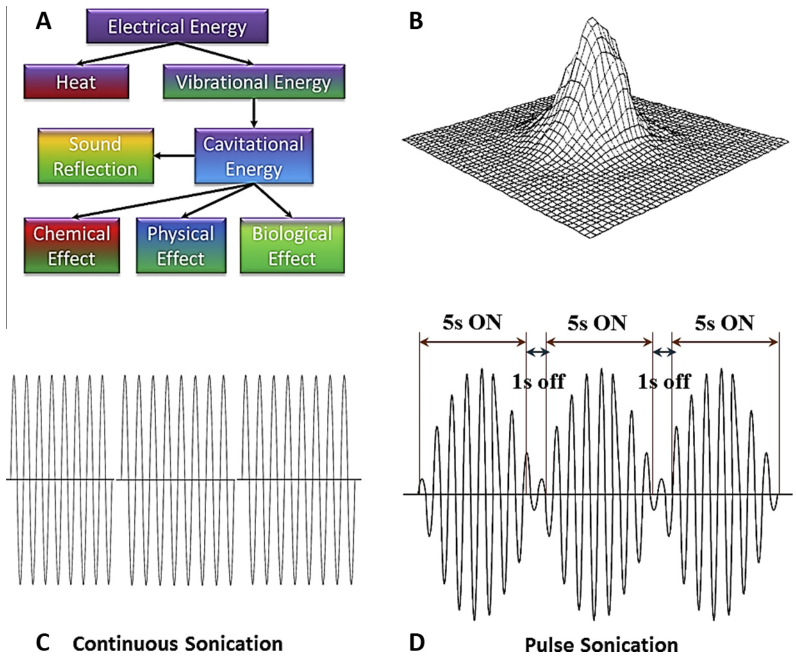

Fig. 1. (a) Energy forms, losses and effects in ultrasonic application; (b) acoustic wave propagation; (c) continuous sonication wave profile; and (d) pulse sonication wave profile.

to the effect of ultrasonic irradiation: (1) rapid movement of fluids caused by a variation of sonic pressure causes solvent compression and rarefaction; (2) cavitation; and (3) microstreaming where large amount of vibrational energy is confined in small volumes of reaction medium with less heating [25,26]. Cavitation phenomenon has been found as effective tool for intensification of the esterification of fatty acids [27]. When using ultrasound irradiation, high reaction medium temperatures are not required to carry out the transesterification reaction. In addition, lower methanol to oil molar ratios and smaller amounts of catalyst are required. Ultrasonication enhances the mass transfer between methanol and oil which results in a higher biodiesel yield. It enhances mass transfer and reaction rates in both multiphase reactors and homogeneous systems [28,29].

Many studies reported on the effects of pulse sonication on the transesterification of fresh and used oils in both direct and indirect (via water bath) applications [26,30–32]. Indirect application was shown to have much lower efficiency reflected by prolonged reaction times when compared to direct ultrasound application [14]. A few studies reported on the direct and continuous application of ultrasonic processing of biodiesel [14,31,33,34]. Nevertheless, none of the studies compared the continuous and pulse effects of direct ultrasound application for biodiesel production. This study compares the effects of continuous and pulse application of direct ultrasonication on used vegetable oil feedstock, and using sodium hydroxide catalyst and methanol as the reactant for transesterification reaction because the aforementioned chemical and physical (thermal) effects may vary largely for different types of ultrasound application. The following sections describe the experimental procedures and compare the principal differences between the continuous and pulse sonication effects on transesterification reaction with discussions on the power density and ultrasonic intensity effects.

2. Materials and methods

2.1. Materials

Used vegetable oil (UVO) was obtained from the Perry Market Cafeteria located in Starkville and methanol and catalyst (sodium hydroxide, NaOH) were purchased from Fisher Scientific®. Methanol and all other reagents used in this study were of analytical grade purchased from Fisher Scientific®. The acid value of UVO was found to be 3.5 mg KOH/g, corresponding to a free fatty acid (FFA) level of 1.7%; therefore a base catalyzed transesterification reaction scheme is suitable for feedstock with FFA content less than 4% [3].

2.2. Ultrasonic irradiation units

The pulse sonication effect on transesterification reaction was performed using a NO-MS100 ultrasonicator manufactured by Columbia International Technologies with a maximum of 1000 W power output capacity. The ultrasonic frequency was 20 kHz. The horn is made of titanium alloy with variable power output rates to vary the effect of ultrasonic application. The ultrasound horn was made with titanium alloy and has the following dimensions: 2.54 cm diameter tapered to 0.254 cm tip diameter. The continuous ultrasonication was performed using a sonicator purchased from Fisher Scientific (Model 550). The sonicator (20 kHz) is rated at 500 W maximum power output and ten levels of power control application. The length of the titanium alloy horn was 12.7 cm with a 3.8 cm diameter tapered to 1.25 cm at the tip.

2.3. Ultrasonic irradiation mechanisms

Fig. 1b shows the ultrasonic wave propagation in a reaction medium regardless of continuous or pulse application. Fig. 1c and d show the wave patterns for continuous and pulse sonication (5 s ON and 1 s OFF). The continuous sonication (Fig. 1c) delivers the acoustic radiation continuously such that the time for relaxation among fluid layers in the reaction medium is not possible increasing thermal energy of the reaction medium. This phenomenon can be considered as loss of some applied energy which could otherwise have been converted to vibrational or cavitational energy. The continuous sonication may energize the transesterification reaction by its thermal effect and is suitable for those reactions requiring thermal excitation. Too much of an exposure to direct sonication may result in undesired reactions and byproduct