Ultrasonics Sonochemistry xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

journal homepage: www.elsevier.com/locate/ultson

Identification of active sonochemical zones in a triple frequency ultrasonic reactor via physical and chemical characterization techniques

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ARTICLE INFO

Article history:
Received 29 November 2015
Received in revised form 18 April 2016
Accepted 24 April 2016
Available online xxxx

Keywords:
Triple frequency
Reactor
Cavitation
Ultrasound
Degradation
Characterization

ABSTRACT

Coupling multiple frequencies in ultrasonic systems is one of the highly desired area of research for sonochemists, as it is known for producing synergistic effects on various ultrasonic reactions. In this study, the characteristics of a hexagonal-shaped triple frequency ultrasonic reactor with the combination frequencies of 28, 40 and 70 kHz were studied. The results showed that uniform temperature increment was achieved throughout the reactor at all frequency combinations. On the other hand, sonochemiluminescence emission and degradation rate of Rhodamine B varies throughout different areas of the reactor, indicating the presence of acoustic 'hot spots' at certain areas of the reactor. Also, coupling dual and triple frequencies showed a decrease in the hydroxyl radical (·OH) production, suggesting probable wave cancelling effect in the system. The results can therefore be served as a guide to optimize the usage of a triple frequency ultrasonic reactor for future applications.

1. Introduction

Power ultrasound is widely used in different applications, such as cleaning [1], enhanced chemical reactions [2], wastewater treatment [3] etc. Over the past few decades, the use of ultrasound has become more established on the laboratory scale. Many researchers have produced reliable results upon using power ultrasound in various applications. However, due to the unique nature of ultrasonic wave, specific optimized ultrasonic setting is required for each unique application. In the field of removal of dye effluent, different ultrasonic settings were required for different dye removal. For example, Siddique et al. [4] have reported that the optimum ultrasonic frequency to remove reactive blue 19 dye was 80 kHz while Kobayashi et al. [5] have shown that sonication at 127 kHz and 490 kHz were able to remove methylene blue dye more effectively. In the field of sonochemical synthesis, it was reported that the effectiveness in synthesizing iron oxide nanoparticles loaded with folate and cisplatin increases with an increase in the ultrasonic frequency, under the same acoustic power [6]. On the other hand, Naddeo et al. [7] have reported that using a lower frequency ultrasound of 35 kHz slowed down the fouling formation in an ultrafiltration system, but higher frequency of 130 kHz favoured the removal of organic matter.

The unique optimized operating conditions of ultrasonic systems have gathered the attention of various researchers to combine different ultrasonic frequencies to enhance the efficiencies of their applications. In recent years, there has been an emerging trend of combining two frequencies for a particular sonication. Research has shown that coupling two different frequencies produced synergistic effects in an ultrasonic process, hence improving the overall efficiency of the process. For example, Koufaki et al. [8] coupled 20 kHz and 40 kHz ultrasound in synthesizing 3,5-disubstituted isoxazoles which resulted in a significant reduction in the reaction time. Sivakumar et al. [9] have reported the observation of synergistic effects upon combining 25 kHz and 40 kHz of sonication for the degradation of *p*-nitrophenol whilst Ninomiya et al. [10] have reported enhanced hydroxyl (·OH) radical generation upon coupling 0.5 and 1.0 MHz ultrasound. On the other hand, Yasuda et al. [11] performed an extensive study on coupling different dual frequency modes ranging from 176 to 635 kHz and found that coupling similar frequencies resulted in better synergistic effects.

Further to the improvements of using dual frequency ultrasound, there have also been several reports on the use of triple frequency ultrasonic systems. Some researchers have reported an enhancement upon coupling three frequencies in their system. For example, Feng et al. [12] showed that combining 28, 1000 and 1870 kHz ultrasound produced 1.3–2.0 times more yield than the summation of individual frequency settings, whilst Zhao et al.

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http://dx.doi.org/10.1016/j.ultsonch.2016.04.029
1350-4177/

[13] reported an enhancement in the degradation of nitrobenzene upon sonication with the frequency combinations of 20, 28 and 40 kHz. However, not all frequency combinations resulted in the enhancement. For example, Gogate et al. [14] combined 20, 30 and 50 kHz ultrasound and did not observe any synergistic effects on the destruction of Rhodamine B, even though the oxidation of potassium iodide showed significant synergistic effects.

Previous work performed by Manickam et al. [15] on the removal of chemical oxygen demand (COD) of palm oil mill effluent (POME) showed successful enhancement upon combining 28, 40 and 70 kHz ultrasonic irradiation, with the aid of H_2O_2 addition, in a triple frequency ultrasonic reactor (TFUR). In the work, it was also reported that the COD pattern fluctuated across the duration of the experiment for different frequency combinations, in the absence of H_2O_2, suggesting that there may be some external factors causing the inhibition of COD removal. The inconsistencies in the experimental outcome for triple frequency ultrasonic systems bring the need for further investigation. Hence, in this investigation, a series of physical and chemical characterization is carried out at specific areas across the TFUR to have a clear understanding on the performance of ultrasonic reactor.

2. Materials and methodologies

2.1. Materials

The hydrogen peroxide (H_2O_2, 30 w/v%) and sodium hydroxide (NaOH) pellets were obtained from R&M Chemicals, Malaysia. Luminol (3-aminophthalhydrazide) was obtained from Friendemann Schmidt Chemicals whilst the ethylene diamine tetraacetic acid (EDTA) and Rhodamine B powder were obtained from Sigma–Aldrich, Malaysia. All chemicals were used as-received without further purification.

The 16.5 L pilot-scale triple frequency ultrasonic reactor (TFUR, Sonictron, Malaysia) is a hexagonal shaped reactor (Fig. 1) that operates at 28 kHz, 40 kHz and 70 kHz. Each frequency mode has a maximum input power of 300 W. The TFUR can operate at seven different frequency modes of 28 kHz, 40 kHz, 70 kHz, 28 + 40 kHz, 28 + 70 kHz, 40 + 70 kHz and 28 + 40 + 70 kHz respectively. The vibrating plate on each transducer is a very thin plate of ca. 2 mm thickness. Six single open-ended glass cylinders (internal diameter: 46.4 mm, outer diameter: 50 mm, length: 450 mm) purchased from Donewell Engineering & Services, Selangor, Malaysia, were used in the experiments and were placed at specific positions of the TFUR as shown in the schematic diagram in Fig. 1. All experiments were performed under a constant temperature of 25 ± 1 °C using the existing cooling coil system from the TFUR, unless stated otherwise.

2.2. Methodologies

2.2.1. Calorimetry

14 L of air saturated deionized water was placed into the TFUR, followed by 350 mL of deionized water into each of the six glass cylinders. The TFUR was sonicated at all seven different frequency modes for an hour each, without temperature regulation. Temperatures at all the six positions in the glass cylinders were recorded at 30 s intervals. The power output of the system, Q was calculated based on Eq. (1).

$$Q = \frac{mCp\Delta T}{\Delta t} \tag{1}$$

where m is the total mass of water, C_p is the specific heat capacity of water (4.18 kJ $kg^{-1}{}^{o}C^{-1}$), ΔT is the temperature change (°C) and Δt is the time difference (s). The heat loss of the system was obtained by finding the temperature profile from heating the TFUR with the same volume of deionized water as the sonicated system with a 1000 W (Q_H) electrical heater for 15 min. The heat loss, Q_{HL}, can be calculated based on Eq. (2).

$$Q_{HL} = Q_H - \frac{mCp\Delta T}{\Delta t} \tag{2}$$

The intensity of the ultrasonic system, I, can then be computed using Eq. (3).

$$I = \frac{Q + Q_{HL}}{A} \tag{3}$$

where $Q + Q_{HL}$ is the output power, P_{out}, and A is the cross-sectional area of the transducer, which are 0.083 m^2, 0.1652 m^2 and 0.3304 m^2 for single, dual and triple frequency modes respectively.

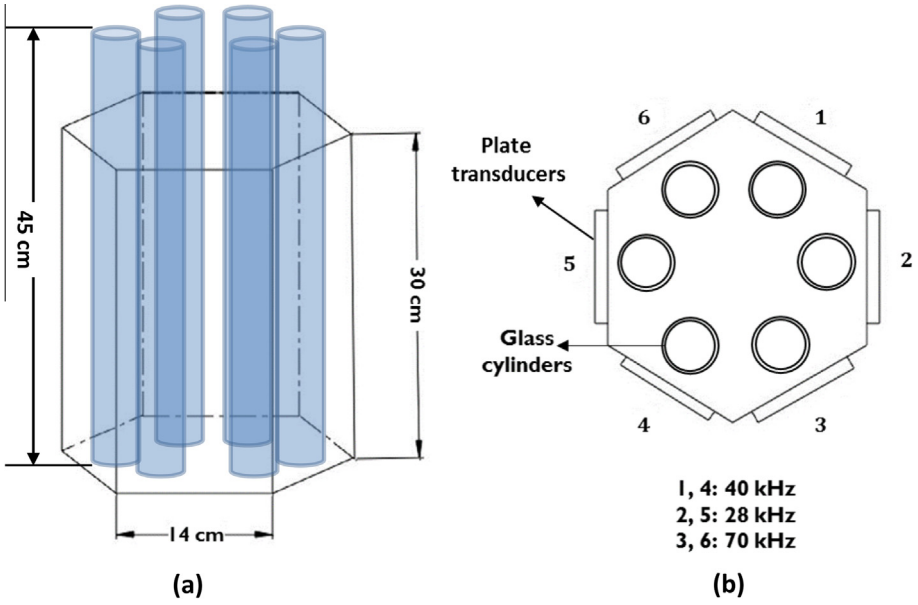

Fig. 1. Schematic diagram of the triple frequency ultrasonic reactor. (a) Side view of the hexagonal shaped reactor with 6 glass cylinders placed in the reactor; (b) top view of the reactor showing the sides of the reactor with different frequency emissions and the positions of glass cylinders and the piezoelectric ultrasonic transducers.

Please cite this article in press as: T.J. Tiong et al., Identification of active sonochemical zones in a triple frequency ultrasonic reactor via physical and chemical characterization techniques, Ultrason. Sonochem. (2016), http://dx.doi.org/10.1016/j.ultsonch.2016.04.029