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Experimental investigation on proper use of dual high-low frequency ultrasound waves—Advantage and disadvantage

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ABSTRACT

An experimental study was conducted to investigate the proper use of dual frequency ultrasound irradiation in sonoreactors. For this purpose, two waves with frequencies of 1.7 MHz and 24 kHz were combined. A dual frequency rig was equipped with a high frequency 1.7 MHz ultrasound transducer and a low frequency one generating 24 kHz waves. Dushman competitive reactions were used to study micromixing, Weissler reaction for evaluating cavitation activity and ammonia degradation experiment to investigate the superposition effect of two coupling waves on decomposition strength. The results revealed that combining these waves has a positive effect on number of generating cavitation bubbles and micromixing performance. However, this combination reduced the strength of the low frequency wave and diminished the ammonia degradation process efficiency. The micromixing enhancement in terms of the segregation index in dual frequency irradiation mode is approximately 46.9% and 40.1% more than single low and high frequency irradiation, respectively. On the other hand, the ammonia removal efficiency is reduced by 10.3% in dual frequency irradiation compared to single low frequency layout.

1. Introduction

Ultrasound is an acoustic wave with frequencies higher than human hearing range (20 kHz) [1]. The chemical effects of ultrasonic irradiation were first reported in 1927 when Richards and Loomis announced the acceleration of conventional reactions and the reduction–oxidation process by ultrasound [2]. The transmission of ultrasound waves through liquids can induce or accelerate a wide variety of chemical reactions. The chemical effects of ultrasound are due to a phenomenon called cavitation, which is the nucleation, growth and implosive collapse of the bubbles in a liquid [3]. Ultrasound waves propagate through the liquid by compressions and rarefactions and during the rarefaction, the negative pressure period of the sound wave, micro bubbles are formed. Bubbles growth takes place in areas with pressure lower than the liquid vapor pressure [4,5]. The cavitation bubble size is related to the resonance frequency as shown in Eq. (1) [6]:

$$Rr = \frac{1}{2\pi f\mathrm{r}}\left(\frac{3kp}{\rho \mathrm{L}}\right) \tag{1}$$

Where, K, is the polytropic coefficient and R_r is the resonant radius of the bubble. According to this equation, low frequency wave propagation leads to the formation of larger bubbles, which have more energy per bubble during a collapse in comparison with smaller bubbles [7]. Zhang [8] reported that acoustic energy releasing during cavitation depends on bubble size according to following relation:

$$\frac{Eac}{(4/3)\pi PhR^3\ \mathrm{max}} = \frac{1}{27C}\left(\frac{2Ph}{\rho}\right)^{1/2}\left(\frac{Ph}{Q}\right)^{3/2} \tag{2}$$

This leads to the fact, that low frequency waves make more violent bubble implosions. D'Agostino and Brennen [9] discovered that the number of bubbles is proportional to the square of the frequency, $N \propto \omega^2$, where N is the number of bubbles and ω is the radial frequency, which its value is calculated by $2\pi f$, and f expresses the ultrasound frequency. Therefore at higher frequencies more bubbles are formed which is due to the more cycles of compression and expansion [7,10]. The heat from the bubble implosion decomposes water into highly reactive hydrogen atoms ($H^\bullet$) and hydroxyl radicals ($OH^\bullet$). Hydroxyl radicals and hydrogen atoms then recombine to form hydrogen peroxide and molecular hydrogen, respectively. Therefore, because of this molecular environment, organic and inorganic compounds may be oxidized or reduced depending on their reactivity [11]. This makes the ultrasound one of the advanced oxidation processes (AOPs) widely used

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Nomenclature

A	light absorption
C	sound velocity, m s^{-1}
C_i	concentration of species i in the fluid after the reaction, mol l^{-1}
C_{i0}	concentration of species i in the fluid before the reaction, mol l^{-1}
C_p	specific heat capacity, J kg^{-1} K^{-1}
E	input power, W
g	incorporation function
I	ionic strength, mol m^{-3}
IUS	intensity of ultrasound, W m^{-2}
K_{eq}	equilibrium constant
k_i	kinetic constant
l	cell length, m
n_i	mole number of i component
P	acoustic pressure, Pa
q	Engulfment flux, m^3 s^{-1}
$\Re_{ij}$	net rate of production of species i for the reaction j, mol m^{-3} s^{-1}
r_i	reaction rate i, mol m^{-3} s^{-1}
T	temperature, K
t	time, s
t_c	critical feed time, s
t_{inj}	injection time, s
t_m	characteristic micromixing time, s
X_S	segregation index
Y	selectivity of iodide
Y_{TS}	selectivity of iodide in the case of total segregation
z_i	charge number of ion i

Greek letters

ε	molar extinction coefficient
υ	volume of acid cloud at t, m^3
υ_0	initial of volume of acid cloud, m^3
γ	relative segregation index
ω	angular frequency, s^{-1}

in wastewater treatment [11–14]. There is another mechanism for removing pollutants, pyrolysis, which is expected to be the main reaction path for the degradation of more volatile compounds [11,15].

The implosive collapse of micro bubbles, also results in mechanical effects caused by shock waves or by micro-jets and micro-streaming that is considered to cause turbulence-jets in a fluid [4]. These phenomena have significant effects on the rate of various processes in chemical industries such as cleaning [16], emulsification [17], extraction [18], wastewater treatment [19] and chemical reactions [20]. Micro-jets and micro-streams are quite important in liquid mixing at the molecular scale (micromixing) [21]. In crystallization, precipitation, polymerization involving processes [22–25] and competitive reactions [26–28] micromixing is more prominent [29].

Conventionally, only waves of single frequency are used in a given system. However, there have been investigations on subjecting a given system with two different frequencies simultaneously [11,30–33].

The aim of this study is to discuss the proper use of dual frequency ultrasound irradiation in sonoreactors. The presented dual frequency sonoreactor was equipped with ultrasound wave transducers with 1.7 MHz and 24 kHz frequencies. These frequencies were selected to be sure that they are exactly in the low and high frequency ranges of ultrasound waves. Ammonia removal efficiency, quantification of cavitation activity and micromixing performance were investigated. The effects of initial concentration, position of piezoelectric transducers and injection zone position in this sonoreactor were examined. The main new issue in this investigation compared with the previous ones [11,30,33] is to illustrate for which purposes combining a high frequency ultrasound wave with a low frequency one can be useful.

2. Theory

Ultrasound travels in the fluid as a cyclic sound pressure, which is defined as follows [34]:

$$PS = PA\cos\left[2\pi f\left(t + \frac{y}{c}\right)\right] \quad (3)$$

Where c is the speed of the sound in the medium and f, y and t are the frequency of ultrasound wave, the space coordinate and time, respectively.

PA is the acoustic pressure amplitude which can be related to the intensity of the ultrasound source as:

$$PA = \sqrt{2\rho LIUSC} \quad (4)$$

In which, IUS and ρL are the ultrasound intensity and the liquid density, respectively [34]. As mentioned earlier, cyclic sound pressure consists of alternating compressions and rarefactions. During the rarefaction period, the local pressure decreases adequately below the liquid vapor pressure causing the formation of cavities in the liquid [1,34]. When cavitation bubbles are formed, they continue to oscillate until they degenerate or eventually become larger to a size that collapse and release a shock wave that beams in a jet from the implosion point. This effect is called transient cavitation [34]. When the bubbles only oscillate and do not implode there's another type of cavitation termed stable cavitation. The effect of this type of cavitation is generation of fluid motion that is called acoustic and micro streaming [35] it can provide micro and macro agitations inside the liquid, thus it can improve the mixing processes. There will be no strong shock waves or jets associated with the violent implosion in stable cavitation [29,36,37]. The value of acoustic streaming velocity depends on the frequency and power of the ultrasound waves and is in a range between 1 and 100 cm s^{-1}.

As shown previously in Eq. (1), the size of cavitation bubbles is proportional to the inverse of its frequency. Moreover, according to Eq. (2) the shock wave energy and intensity, resulted from the cavitation bubble collapse, is proportional to the size of the bubble and is stronger when a larger cavitation bubble implodes. In the case of constant power input, a low frequency wave will produce fewer number of cavitation bubbles with stronger implosions while a higher frequency produces more numbers of cavitation bubbles with a smaller size [38].

3. Materials and methods

In order to generate the low frequency ultrasound wave in the sonoreactor, a wave transducer manufactured by UP400S with a frequency of 24 kHz and the horn tip diameter of 20 mm was placed at top of the reactor. Its electric power supply is adjustable in the range between 0 and 400 W. An ultrasonic power intensity input of 280 W (89.17 W/cm^2) was used for all experiments. A high frequency (1.7 MHz) piezoelectric transducer (Model ANN-25 17GRL, ANNON PIEZO TECHNOLOGY CO. LTD., China) with a diameter of 1.5 cm and an input power of 9.5 W was installed at the bottom of the reactor.

The chemical materials in this work, ammonia solution of 25% extra pure, Nessler reagent, (KI), (NaCl), (H_3Bo_3), (KIO_3) and (NaOH) all were obtained from Merck Inc and the sulfuric acid with a purity of 98% was provided from Fluka.