Ultrasonics Sonochemistry 28 (2016) 276–282

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

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

Dual frequency cavitation event sensor with iodide dosimeter

Ali Ebrahiminia [a], Manijhe Mokhtari-Dizaji [a,*], Tayebeh Toliyat [b]

[a] Department of Medical Physics, Faculty of Medical Sciences, Tarbiat Modares University, Iran
[b] Department of Pharmaceutics, Faculty of Pharmacy, Tehran University of Medical Sciences, Tehran, Iran

ARTICLE INFO

Article history:
Received 1 June 2015
Received in revised form 3 July 2015
Accepted 3 July 2015
Available online 4 July 2015

Keywords:
Dual frequency
Sonication
Acoustic cavitation
Iodide dosimeter

ABSTRACT

The inertial cavitation activity depends on the sonication parameters. The purpose of this work is development of dual frequency inertial cavitation meter for therapeutic applications of ultrasound waves. In this study, the chemical effects of sonication parameters in dual frequency sonication (40 kHz and 1 MHz) were investigated in the progressive wave mode using iodide dosimetry. For this purpose, efficacy of different exposure parameters such as intensity, sonication duration, sonication mode, duty factor and net ultrasound energy on the inertial cavitation activity have been studied. To quantify cavitational effects, the KI dosimeter solution was sonicated and its absorbance at a wavelength of 350 nm was measured. The absorbance values in continuous sonication mode was significantly higher than the absorbance corresponding to the pulsed mode having duty factors of 20–80% ($p < 0.05$). Among different combination modes ($1\ MHz_{100\%} + 40\ kHz_{100\%}$, $1\ MHz_{100\%} + 40\ kHz_{80\%}$, $1\ MHz_{80\%} + 40\ kHz_{100\%}$, $1\ MHz_{80\%} + 40\ kHz_{80\%}$), the continuous mode for dual frequency sonication is more effective than other combinations ($p < 0.05$). The absorbance for this combined dual frequency mode was about 1.8 times higher than that obtained from the algebraic summation of single frequency sonications. It is believed that the optimization of dual frequency sonication parameters at low-level intensity (<3 W/cm^2) by optically assisted cavitation event sensor can be useful for ultrasonic treatments.

1. Introduction

Acoustic cavitation, which is defined as the oscillation of bubbles in an acoustic field, consists of three stages: nucleation, growth and collapse of bubbles. It is well known that the acoustic cavitation can be classified into stable and inertial cavitations [1,2]. The collapse event can be violent enough for generating temperatures of about 4000 K and pressures in excess of 1000 atmospheres. The extremely high pressure and temperature are capable of causing sonochemical reactions [3,4]. When water is sonicated with threshold intensity, collapse of cavitation bubbles leads to the formation of reactive radical species, such as hydrogen peroxide (H_2O_2), hydroxyl radicals ($^{\cdot}OH$) and hydroperoxyl radicals ($HOO^{\cdot}$) [5]. Several methods for determining and quantifying inertial cavitation are available, including sonoluminescence, subharmonic analysis, acoustic imaging, laser holography and electron spin resonance. Some of these methods such as electron spin resonance are an extremely sensitive method for detecting the radical produced but its application needs specialist and expensive equipments [6]. Chemical products also may be used to measure cavitation activity. Various sonochemical dosimeters have been developed to monitor the hydroxyl radical generation, which are known as indirect cavitation monitors. Chemical methods namely terephthalate dosimeter, Fricke dosimeter and iodide dosimeter are suitable for monitoring the chemical effects of inertial cavitation [3,4,7]. Terephthalic acid dosimetry is a time consuming procedure and needs to have a spectrofluorometer. The dosimeters based on the photometry, are not high sensitive. Fricke dosimeter is used in sonochemistry too, but iodide dosimeter is very simple and widely acceptable [3,7]. The oxidation of potassium iodide (KI) is widely regarded as a standard to calibrate sonochemical efficiency. In addition, preparation and handling of KI solution is simple and easy [7]. Therefore, in order to quantify the bubbles generated in dual frequency sonication, the cavitation activity induced by the ultrasound waves on different exposure parameters has been measured by potassium iodide dosimetry. Therefore, in this study, chemical effects of sonication parameters were investigated as optically assisted dual frequency cavitation event sensor using iodide dosimeter.

Several researches have been carried out using multiple frequencies to enhance cavitation events [8,9]. Multiple frequency ultrasound is increasingly being employed to achieve enhancement of sonochemical reaction performance by the

* Corresponding author at: Department of Medical Physics, Tarbiat Modares University, Tehran, Iran.
E-mail address: mokhtarm@modares.ac.ir (M. Mokhtari-Dizaji).

http://dx.doi.org/10.1016/j.ultsonch.2015.07.005
1350-4177/

superimposition of ultrasound fields [6,10,11]. It has been suggested that ultrasonic fields of a few tens of kHz cause main cavitation activities in the medium while the MHz fields lead to nucleation of bubbles which in combination with kHz frequency results in cavitation activity increase [6]. Therefore, we believe that by combining 40 kHz and 1 MHz frequencies, it is possible to enhance cavitation and concentrate energy in a restricted area in a media such as human tissue. Numerous studies have been carried out of dual frequency enhanced cavitation activity, however, the experimental settings and conditions differ from one another and a universal setting protocol is not available. In our previous studies, it has been shown that a combination of two ultrasonic fields including a kHz plus a MHz continuous field has the highest subharmonic amplitude and the most localized effective irradiation area [6].

In this study, the effect of different sonication parameters for the mentioned combined field was optically assessed in several combinations; mode of sonication, duty cycle, intensity and duration at low level intensities ($I \leqslant 3$ W/cm^2) by dual frequency cavitation event sensor. To quantify cavitational effects, the KI dosimeter solution was sonicated and its absorbance at a wavelength of 350 nm was measured for I_3^- spectrophotometrically. The purpose of this work is development of dual frequency inertial cavitation meter for therapeutic applications of ultrasound waves. This method can be defined as optically assisted dual frequency cavitation event sensor.

2. Materials and methods

2.1. Dosimetry solution

Cavitational activity were estimated under various sonication conditions using a potassium iodide (KI) dosimeter as a standard method [7,12–14]. In this method, when potassium iodide (KI) solution is sonicated, oxidation occurs and I^- ions are oxidized by the generated reactive radicals to give I_2. The excess of I^- ions present in the solution reacts with I_2 to form triiodide ion (I_3^-) [11]. The concentration of I_3^- ions can be quantified via UV spectrophotometer at 350.00 nm [13]. When a potassium iodide solution is sonicated, the following reactions occur [15] (reactions 1–5):

$$^{\cdot}OH + I^- \rightarrow OH^- + I \quad \text{(Reaction1)}$$

$$H_2O_2 + 2I^- + 2H+ \rightarrow I_2 + 2H_2O \quad \text{(Reaction2)}$$

$$I + I^- \rightarrow I2^- \quad \text{(Reaction3)}$$

$$2I_2^- \rightarrow I_2 + 2I^- \quad \text{(Reaction4)}$$

$$I_2 + I^- \rightarrow I_3^- \quad \text{(Reaction5)}$$

Indeed, this is an indirect method for inertial cavitation activity estimation in ultrasound fields. In these experiments, a modified iodide dosimeter was prepared from a 0.1 M potassium iodide solution (KI, 16.6 g, Merck, Darmstadt, Germany) and then 0.2 M chloral hydrate ($CCl_3CH(OH)_2$, 33 g, Merck, Darmstadt, Germany) was added to the dosimeter solution. Chloral hydrate was added to enhance the oxidation of iodide [8]. Double distilled water was used as a solvent for experiments and resultant solution was made up 1 dm^3. Sonication was carried out at 22 °C before which, dosimeter solution was aerated for 15 min. In all experiments, 50 ml of KI solution were added to the reaction cell [12]. The sonicated dosimeter was analyzed for I_3^- by spectrophotometer (Optizen 2120 UV Plus spectrometer, Mecasys Co Ltd., Daejeon, Korea). The absorbance was measured in the individual samples immediately after sonication at 350 nm spectrophotometry.

2.2. Equipment setup

The applied MHz ultrasound system was a 1 MHz therapeutic probe (Sonopuls 492, Enrof Nonius Co., Rotterdam, Netherland) with a 30 mm diameter and 5 cm^2 effective radiation area (ERA). By this 1 MHz apparatus, it was possible to change Duty factor (20–80%), sonication mode and intensity (0–2 W/cm^2). For 40 kHz sonication, a system was designed and constructed with a probe diameter of 30 mm in our lab (Tarbiat Modares University, Ultrasound Lab, Tehran, Iran) in collaboration with Pars Nahand Engineering Company (Pardis Technology Park, Tehran, Iran). This system was capable of working either in continues or pulsating mode within a range of different duty factors (20–80%), different intensity (0–1.34 W/cm^2) and also, at a wide range of pulse repetition frequencies (PRF) from 0.5 Hz to several kHz. All reported experimental intensity values include the spatial average temporal average intensity (I_{SATA}). Acoustic frequency and intensity calibration of the applied systems was performed in degassed water in a cubic $25 \times 20 \times 20$ cm^3 Perspex tank. The inner surfaces of the tank in front of the transducers were covered by ultrasonic absorbent material to reduce acoustical reflections. A calibrated PVDF-type hydrophone (PA124, Precision Acoustics Ltd., Dorchester, Dorset, UK, calibration range: 20 kHz–1 MHz, with a sensor diameter of 25 mm) was positioned at a distance of 5 mm from the probe surface on the transducer axis to record the ultrasonic signals at the place of chemical dosimetry chamber. The hydrophone was connected to a digital oscilloscope and spectrum analyzer (TNM 20080, TNM Electronics Ltd., Tehran, Iran) and then to the computer (Intel Pentium IV, 3.00 GHz, Taiwan) for the observation, measurement and processing of the spectrum [16]. Actual frequencies of the therapeutic and our designed probes were 980.95 kHz was 39.5 kHz respectively. In order to compare the effect of the sonication mode at different duty factors (DF%), having equal ultrasound energy, on the inertial cavitation activity, the ultrasound energy (E) was defined as [10]:

$$E\ (\mathrm{J}) = I\ (\mathrm{W/cm^2}) \times t\ (\mathrm{s}) \times \mathrm{D.F}\ (\%) \times \mathrm{ERA}\ (\mathrm{cm^2}) \quad (1)$$

where I, t and ERA are spatial average temporal average intensity, exposure duration and effective radiation area respectively. The sonication field was compensated for duration and intensity in 1 MHz frequency pulsed wave mode to maintain the ultrasound energy constant. The compensation processes were as following. According to the Eq. (1), the ultrasound energy transferred to the sonicated solution is proportional to the ultrasound intensity (SATA), exposure duration, ERA and duty factor. For 1 MHz probe, the ultrasound intensity was set on 2 W/cm^2. Then the duty factor (DF) was changed from 20% to 100% (20, 50, 80 and 100) and sonication duration was changed accordingly to compensate the effect of changing DF and maintain the transferred energy constant (1200 J).

For the next experiment, the sonication duration was set on 20 min and again the DF values were changed from 50% to 100% (50, 80,100) and the intensity (SATA) was modified accordingly (2, 1.3 and 1 W/cm^2 respectively). This way, the net ultrasonic energy transferred to the sonicated solution remained constant (6000 J).

For sonication, a cubic reaction cell with a volume of 50 cm^3 was constructed from Perspex and attached to the inner surface of the cubic $15 \times 15 \times 10$ cm^3 Perspex tank. In order to maximize the acoustic intensity entering the reaction cell, the ultrasonic