The 7th International Conference on Applied Energy – ICAE2015

Energy Conversion Efficiency of Pulsed Ultrasound

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Abstract

Energy characterization of a pulsed ultrasonic system was carried out using a modified calorimetric method. Sonochemical efficiency (SE) for the oxidation of Fe^{+2} and the formation of H_2O_2 was determined for selected *on:off* ratios (R) and different power levels. The measured efficiency of the pulsed ultrasonic system of 60-70% in converting electrical energy into calorimetric energy was found to be constant for all R ratios and equivalent to that for continuous operation. SE of Fe^{+2} and H_2O_2 for pulsed ultrasound was higher than that of continuous ultrasound. The ratio R=0.2:0.1 had the highest SE values overall, while for long off-time ratios, R=0.1:0.6 recorded the highest value of SE. These results were supported by the production rates results for Fe^{+2} and H_2O_2.

1. Introduction

Ultrasound technology refers to the use of sound energy in the frequency spectrum that exceeds the human hearing limit (> 20 kHz) [1]. Ultrasound is a chemical-free energy that can be harnessed in various applications such as water treatment [2], fuel preparation [3], food processing [4, 5] and chemical synthesis [6]. Ultrasound has the disadvantage of being high energy demand technology [4]. Therefore, the current research in ultrasound field focusses on finding ways to reduce energy requirement for this technology.

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There are several ways suggested in the literature [4, 7] for reducing energy demand in ultrasound. One of these ways is operating ultrasound on pulsed mode (pulsed ultrasound).

Pulsed ultrasound is defined as the operation of ultrasound on a chosen *on:off* ratio *R*. The period during which ultrasound is turned on is known as pulse length whereas the off-time is known as interval length. Although pulsed ultrasound is mostly proposed as energy saving operational mode, it can be just as or even less effective than continuous ultrasound in some instances [8]. Such low efficiency of pulsed ultrasound is mostly related to the choice of operating parameters (e.g. *R* ratio, pulse length and interval length) and the way through which the energy characterisation is carried out. Hence, it is of utmost importance to scrutinize energy conversion in pulsed ultrasonic system and optimize it with regards to the aforementioned parameters. The objectives of this study are to improve on the current techniques for determining energy conversion in pulsed ultrasound and to investigate the effect of the operating conditions on the chemical yield of pulsed ultrasound. The second objective will help in identifying the optimum process parameters for pulsed ultrasound. The electrical energy conversion into vibrational energy for a low frequency high power ultrasonic horn reactor was characterized based on a modified calorimetric method reported in our previous study [9]. Sonochemical effects of ultrasound are normally determined via measuring the production of $HO^{\cdot}$ and H_2O_2 [7, 10, 11]. SE of $HO^{\cdot}$ and H_2O_2 for pulsed and continuous ultrasound were measured and the operating conditions with the highest SE were identified. The production rates of $HO^{\cdot}$ (measured from ferrous oxidation) and H_2O_2 were also determined to further understand the effect of the operating parameters on the chemical throughput of ultrasound in both modes pulsed and continuous.

Nomenclature

R	*on:off* ratio
P_{cal}	calorimetric power (W)
P_{out}	output power from the system (W)
P_{accum}	accumulated power in the system (W)
m	mass of the irradiated water (kg)
C_p	heat capacity per unit mass (J/kg.K)
dT/dt	slope of the temperature rise versus time (K/s)
$\bar{h_c}$	average convection heat transfer coefficient (W/m^2.K)
A	surface area of the cavitation chamber (m^2)
T_w	average temperature of the cavitation chamber surface (K)
T_∞	ambient temperature (K)
η	efficiency
P_{in}	input electrical power (W)
SE	sonochemical efficiency
C	concentration (mole/L)
V	volume of the treated water (L)
t	treatment time
t_p	treatment time for pulsed ultrasound
$t_\circ$	treatment time for continuous ultrasound

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2. Materials and methods

2.1. Experimental setup

The system used in the experimental work is consisted of electrical power measurement equipment, ultrasonic horn reactor, temperature sensors and data acquisition system. The electrical power withdrawn

by the system components (i.e. generator, transducer and horn) was measured using a high precision wattmeter (EDMI MK7C Single Phase Smart Meter) and oscilloscope (Tektronix, TDS5034B Digital Phosphor Oscilloscope). Ultrasonic horn reactor (maximum power of 400 W, 20 kHz) equipped with 3/4″ titanium horn was used to perform the experiments. In-house fabricated stainless steel cavitation chamber was used to hold 400 mL of reaction solution. Eight calibrated platinum thin film detectors (supplied by RS Australia) were fixed in and outside the cavitation chamber to measure temperature rise of the irradiated solution and the walls of the chamber during ultrasound operation. Detailed description of the experimental setup is available in [9].

2.2. Power measurements

Electrical and calorimetric power measurements were performed on the system components to determine the energy conversion efficiency of the system as whole and its components for continuous and pulsed modes. Power measurements of the continuous mode were discussed in our previous work [9], and the results showed overall efficiency of the system between 60-70% and for system components ranged between 80-98%. To check whether efficiency figures of the continuous mode can be applied on the pulsed mode, the precision of the generator in controlling the output power needs to be tested. This was achieved by monitoring the output current wave of the generator using current probe (Tektronix, TCP 202) for two R ratios at the same power.

The temperature rise of the sonicated liquid (deionized water) and the walls of the cavitation chamber was collected to measure the calorimetric energy of ultrasound by applying energy balance on the irradiated water as follows:

$$P_{cal} = P_{out} + P_{accum} \tag{1}$$
$$P_{accum} = m\bar{C}_p \ dT/dt \tag{2}$$
$$P_{out} = \bar{h}_c \ A \ (T_w - T_\infty) \tag{3}$$

The energy exiting the irradiated water is consumed in heading up the cavitation chamber. Hence, energy balance equations (1-3) can be applied on the chamber walls to calculate the convective energy lost to the atmosphere. More details on the calorimetric calculations can be found in [9]. The overall efficiency of the system is calculated from the equation bellow:

$$\eta = P_{cal}/P_{in} \tag{4}$$

The input electrical power into the pulsed ultrasonic system was measured by averaging instantaneous power over treatment time. It is noteworthy that calorimetric measurements of long off-time ratios at low power requires long operating time to achieve tangible temperature rise in the liquid and the chamber

2.3. Experimental procedure

Ultrasound device was run on continuous and pulsed modes. The energy conversion efficiency of pulsed ultrasound was investigated for three groups of R ratios within 1 second time window. Long interval group include 0.1:0.8, 0.1:0.6, 0.1:0.4 and 0.1:0.2. Long pulse group include 0.8:0.1, 0.6:0.1, 0.4:0.1 and 0.2:0.1. The third group only contained one ratio of 0.1:0.1 (pulse=interval). Ultrasound was operated for 15 minutes for each R ratio and for the continuous mode at different power levels. Samples were withdrawn after each 5 minutes and the concentration of the targeted chemical species was quantified as explained in

the following section. The production rates of the chemical species were determined from the concentration vs. time curves.

2.4. Chemical dosimetry

Fricke and H_2O_2 dosimeters were applied in this study to evaluate ultrasonic energy conversion into chemical yield. Frick dosimeter is based on the oxidation of ferrous to ferric in an acidic environment under the effect of oxidants. Fricke dosimeter is used to measure the generation of $HO^{.}$ and other radical species as illustrated in equations 5-7 [11]. H_2O_2 dosimeter is based on the conversion of iodine ion to triiodide ion under the oxidative effects of H_2O_2 (equations 8 and 9). Peroxide dosimeter is used to measure the recombination activity of $^{.}OH$.

$$HO_2^{.} + Fe^{+2} + H^+ \rightarrow Fe^{+3} + H_2O_2 \tag{5}$$
$$HO^{.} + Fe^{+2} + H^+ \rightarrow Fe^{+3} + H_2O \tag{6}$$
$$H_2O_2 + Fe^{+2} + H^+ \rightarrow Fe^{+3} + H_2O + HO^{.} \tag{7}$$
$$H_2O_2 + 2I^- + 2H^+ \rightarrow I_2 + 2H_2O \tag{8}$$
$$I_2 + I^- \leftrightarrow I_3^- \tag{9}$$

Fricke solution was prepared by dissolving 10^{-3} mole $Fe(NH_4)_2(SO_4)_2.6H_2O$, 0.4 mole H_2SO_4 and 10^{-3} mole NaCl in 1 L of deionised water. The amount of the produced ferric ions was measured spectrophotometrically at wavelength 304 nm ($\varepsilon = 2197$ L/mol.cm) [12]. Fricke solution was air saturated prior to ultrasound exposure by bubbling filtered air through it for 30 mins as suggested in [13].

H_2O_2 was produced by sonicating deionised water and its concentration was measured using iodometric method [14]. Aliquot sample of irradiated water was withdrawn from the reaction solution and added to the spectrophotometer cuvette that contained equal amount of solutions A and B. Solution A was prepared by dissolving 33 g of KI, 1 g of NaOH and 0.1 g of $(NH_4)6Mo_7O_{24}.4H_2O$ in 500 mL of deionised water. Solution B was prepared by dissolving 10 g of KHP in 500 mL deionized water. Solution A was always kept in the dark to avoid any inadvertent Photo-oxidation. The spectrophotometric measurements were performed using JENWAY UV/Vis spectrophotometer, model 6705 and 1 cm path length Quartz cuvette. The absorbance of Fricke solution and the mixture of A and B solutions was subtracted from the absorbance of the samples. Deionised water was used as baseline. The measurements were conducted in triplicate. All the chemicals used for the dosimetry measurements were AR grade supplied by Sigma-Aldrich and Chem-Supply, Australia.

SE of Fe^{+3} and H_2O_2 were calculated based on the yielded concentrations after 10 minutes of ultrasonic operation by applying the formula below:

$$SE = CV / P_{cal} t \tag{10}$$

3. Results and discussions

3.1. System efficiency based on calorimetric measurements

Figure 1 shows the sinusoidal current wave of pulsed ultrasound at 50% amplitude for two *R* ratios. It can be noticed from figure 1 that the generator produced precise *on:off* ratios for the supplied power. This confirms that the efficiency of the system components with continuous mode is the same as that of the pulsed mode which is supported by the overall efficiency results (Figure 2). The overall efficiency of pulsed ultrasound is slightly higher than that of continuous ultrasound due to the more sever collapse of the bubbles form at the start of each pulse as compared to collapse of the steady state bubbles (continuous ultrasound)

[15]. The results shown in figures 1 and 2 affirm that operating ultrasonic system on pulsed or continuous modes has no effect on the electrical power conversion into vibrational energy (calorimetric power). Hence, the efficiency figures of the continuous system components measured in our previous study [9] can perfectly represent the pulsed mode. The time required for pulsed ultrasound to produce the same level of energy as that of continuous ultrasound can be appropriately calculated using equation 11.

$$t_p = t_\circ (1+1/R) \tag{11}$$

Monitoring the energy conversion in ultrasound system is an important practice for ultrasound technology especially when such technology is criticized as being high energy demand technology. This practice is mostly overlooked which leads to overestimation to the electrical power required for a particular ultrasonic application. It is also important to measure the efficiency of individual component in the ultrasonic system as this would enable manufacturers to identify the most power consuming components and improve on their performance.
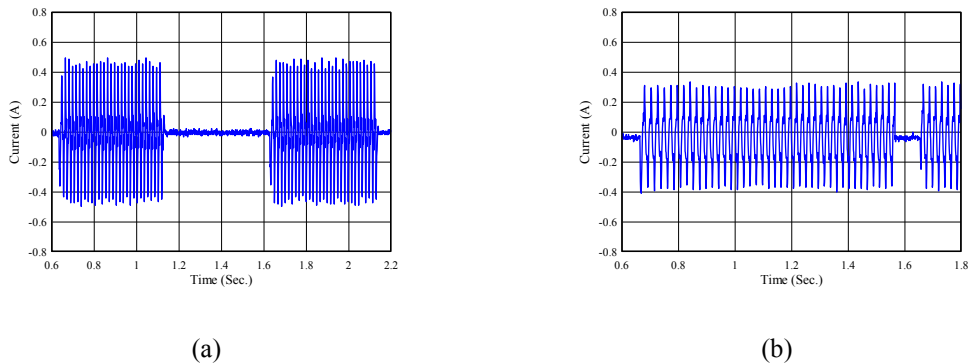

(a)   (b)

Figure 1: Sinusoidal current wave Vs. time of pulsed ultrasound for a) *R*=0.5:0.5 and b) *R*=0.9:0.1
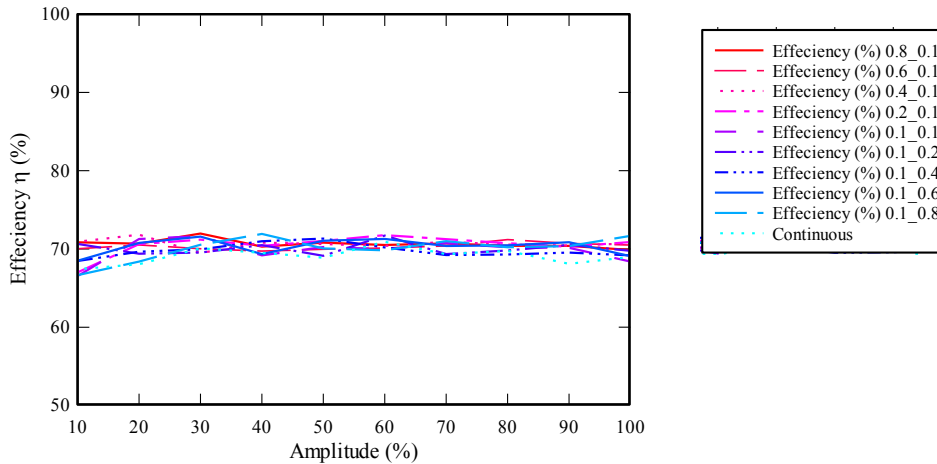

Figure 2: Overall efficiency of ultrasound with continuous and pulsed mode

3.2. Sonochemical efficiency

Sonochemical efficiency of ferric denoted as SE_{Fe+3} for continuous and pulsed ultrasound at various amplitude percentages are shown in Figure 3a. Generally, SE_{Fe+3} of pulsed ultrasound for all the examined R ratios showed a multiple fold increase as compared to continuous ultrasound over the amplitude range of 10-100%. The increase of Fe^{+3} yield in pulsed ultrasound reflects an increase in the production of free radicals and their recombination products (equations 5-9). This trend is supported by the results of the SE for H_2O_2 (SE_{H2O2}), Figure 3b.

The increment of SE_{H2O2} production of pulsed ultrasound as opposed to continuous ultrasound peaked at approximately 1.5 fold, while the maximum increment of SE_{Fe+3} was more than 8 fold. These results are in close agreement with the observation of Casadonte Jr, Flores and Petrier [8]. The large difference observed in sonochemical enhancement of SE_{Fe+3} compared to SE_{H2O2} suggests that the oxidation of ferrous happened mostly via free radicals and H_2O_2 might have evaporated into the collapsing bubbles providing a substrate for the formation of further hydroxyl radicals [16]. Interestingly, the maxima of SE for both Fe^{+3} and H_2O_2 shifted from 10% amplitude in continuous ultrasound to 30-40% amplitude for pulsed ultrasound. This shift in the maxima highlights the importance of these power levels (30-40% amplitude) in a pulsed mode ultrasonic system.
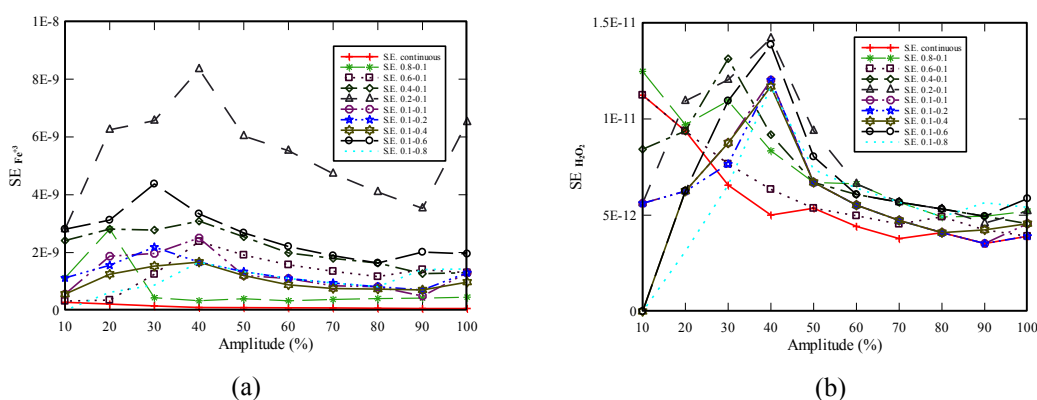

Figure 3: Sonochemical efficiency vs. amplitude percentage of pulsed and continuous ultrasound for (a) Fe^{+3} and (b) H_2O_2

The improvement in sonochemical activities of pulsed mode is attributed to different mechanisms. Yang, Rathman and Weavers [17] and Casadonte Jr, Flores and Petrier [8] suggest that the oxidation of chemical species with an ionic or polar nature such as Fe^{+3} under the effect of cavitation occur via a diffusion mechanism. The intervals between the subsequent pulses allow enough time for the chemicals of interest to diffuse into the seeding bubbles of the following pulse leading to better reactions. Dekerckheer, Bartik, Lecomte and Reisse [15] found that the residual acoustic pressure and active bubbles in the *off* period of pulsed ultrasound could be behind the difference in the levels of sonochemical activities for pulsed mode as compared to the continuous mode. Tuziuti, Yasui, Lee, Kozuka, Towata and Iida [18] have ascribed sonochemical enhancement of pulsed ultrasound to residual acoustic pressure and the spatial enlargement of the chemically active zone in the irradiated solution. However, they concluded that the residual pressure has a superior role in the chemical enhancement. Others [7] suggest that the enhancement in the chemical activities of pulsed ultrasound particularly at higher power levels (> 70% amplitude), is attributed to the lower shielding effects which allow better energy transmission through the irradiated liquid compared to continuous ultrasound. This trend was evident in this study, as it can be seen from figure 3 (a) and (b) that after the maxima, SE for most of R ratios decreased as the amplitude increased until 90 % amplitude after

which SE went up again. To further elaborate on this point, the production rates of Fe^{+3} and H_2O_2 were plotted against amplitude percentages as shown in figures 4 and 5 respectively. The data in these figures clearly shows an increase in the chemical yield of pulsed ultrasound at higher power level. It is worth mentioning that the production rates of Fe^{+3} and H_2O_2 exhibited zero-order kinetics with values close to those reported in the literature [19]. Using high power and low frequency ultrasonic energy results in a low number of powerful bubbles with each pulse [1]. Thus the residual number of bubbles would be very small and that would confine the enhancement mechanisms in this study to residual acoustic pressure, spatial enlargement and lowering of shielding effects. These analyses are reasonable as a high power wave can propagate through the liquid for longer distances ($P_A/P_{A\circ} = e^{-\alpha x}$ [15]) and its residual effect can last longer in liquid.

   Regardless of the chemical enhancement mechanisms, pulsed ultrasound had higher chemical effects than continuous ultrasound for the same energy level. This confirms that pulsed mode can reduce the energy demand of ultrasound technology considerably. To make this mode more energy efficient, the suitable pulse and interval lengths need to be optimized for specific ultrasonic systems as they vary from case to case [7]. In this study, it was observed that R=0.2:0.1 resulted in the highest SE for all the tested groups of R ratios. While R=0.1:0.6 had the highest SE among the long time-off ratios.
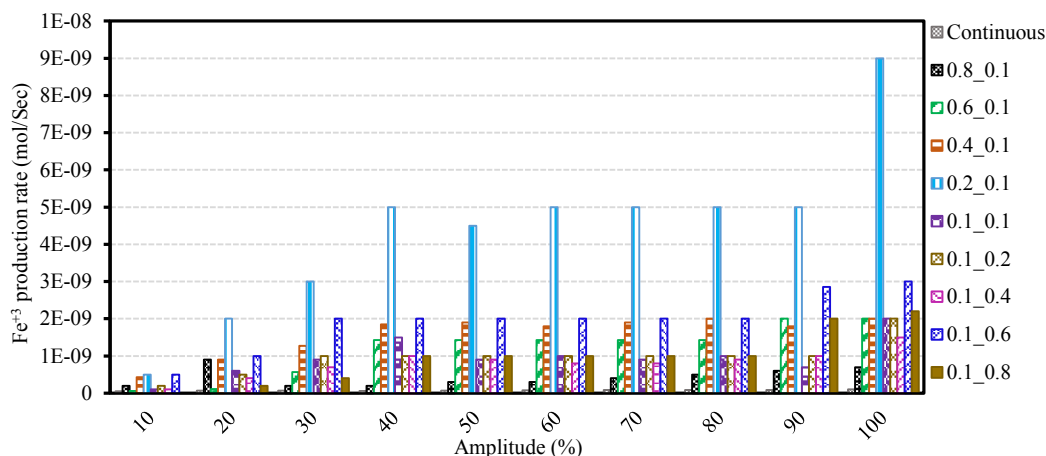

Figure 4: Fe^{+3} production rates vs. amplitude percentage of pulsed and continuous ultrasound
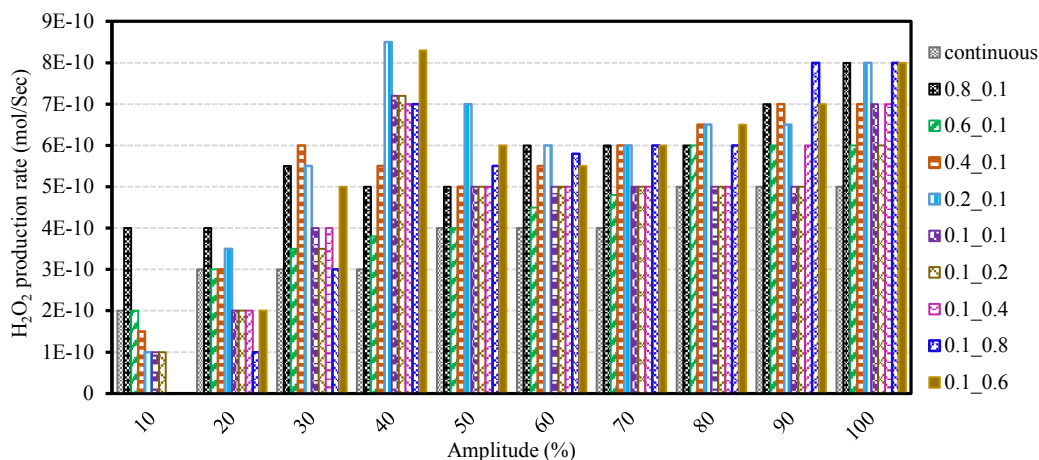
Figure 5: H_2O_2 production rates vs. amplitude percentage of pulsed and continuous ultrasound

4. Conclusion

Systematic investigation into the conversion of electrical energy into calorimetric energy and chemical yield was carried out for a low frequency high power ultrasonic horn reactor. It was found that operating the ultrasonic system in pulsed mode did not affect the performance of the system with regards to its efficiency in converting the electrical power to calorimetric power. However, the conversion of electrical power to chemical yield expressed as SE was more efficient for pulsed ultrasound compared to continuous ultrasound. SE_{Fe+3} and SE_{H2O2} of pulsed ultrasound was higher than that of continuous ultrasound by a small fraction to several fold depending on the applied R ratio. R=0.2:0.1 had the highest SE compared to other ratios, whilst R=0.1:0.6 resulted in the highest SE values among the long off-time ratios. The effectiveness of these two R ratios in ultrasonic applications needs to be examined in the future to confirm their potential in saving energy in ultrasonic systems.

References

[1] Al-Juboori, RA, T Yusaf. Identifying the Optimum Process Parameters for Ultrasonic Cellular Disruption of E. Coli. *International Journal of Chemical Reactor Engineering* 2012;**10**: 1-32.
[2] Al-Juboori, RA, T Yusaf, V Aravinthan. Investigating the efficiency of thermosonication for controlling biofouling in batch membrane systems. *Desalination* 2012;**286**: 349-57.
[3] Al-lwayzy, SH, T Yusaf, RA Al-Juboori. Biofuels from the Fresh Water Microalgae Chlorella vulgaris (FWM-CV) for Diesel Engines. *Energies* 2014;**7**: 1829-51.
[4] Yusaf, T, RA Al-Juboori. Alternative methods of microorganism disruption for agricultural applications. *Applied Energy* 2014;**114**: 909-23.
[5] Chemat, F, H Zill e, MK Khan. Applications of ultrasound in food technology: Processing, preservation and extraction. *Ultrasonics Sonochemistry* 2011;**18**: 813-35.
[6] Wongpisutpaisan, N, P Charoonsuk, N Vittayakorn, W Pecharapa. Sonochemical synthesis and characterization of copper oxide nanoparticles. *Energy Procedia* 2011;**9**: 404-9.
[7] Chen, D, SK Sharma, A Mudhoo, Handbook on Applications of Ultrasound: Sonochemistry for Sustainability, in, CRC Press, 2011.

[8] Casadonte Jr, DJ, M Flores, C Petrier. Enhancing sonochemical activity in aqueous media using power-modulated pulsed ultrasound: an initial study. *Ultrasonics Sonochemistry* 2005;**12**: 147-52.
[9] Al-Juboori, RA, T Yusaf, L Bowtell, V Aravinthan. Energy characterization of ultrasonic systems for industrial processes. *Ultrasonics* 2014, http://dx.doi.org/10.1016/j.ultras.2014.10.003.
[10] Nowak, FM. *Sonochemistry: Theory, Reactions, Syntheses, and Applications*. Nova Science Publishers, Incorporated; 2010.
[11] Virot, M, L Venault, P Moisy, SI Nikitenko. Sonochemical redox reactions of Pu(iii) and Pu(iv) in aqueous nitric solutions. *Dalton Transactions* 2015.
[12] Koda, S, T Kimura, T Kondo, H Mitome. A standard method to calibrate sonochemical efficiency of an individual reaction system. *Ultrasonics Sonochemistry* 2003;**10**: 149-56.
[13] Sugimoto, Ki, R Tsujino, S Musha. A Continuous Dosimeter by Use of the Fricke Dosimeter. *Bulletin of University of Osaka Prefecture. Series A, Engineering and natural sciences.* 1963;**11**: 75-80.
[14] Klassen, NV, D Marchington, HC McGowan. H2O2 determination by the I3-method and by KMnO4 titration. *Analytical Chemistry* 1994;**66**: 2921-5.
[15] Dekerckheer, C, K Bartik, J-P Lecomte, J Reisse. Pulsed Sonochemistry. *The Journal of Physical Chemistry A* 1998;**102**: 9177-82.
[16] Chakma, S, VS Moholkar. Investigations in Synergism of Hybrid Advanced Oxidation Processes with Combinations of Sonolysis + Fenton Process + UV for Degradation of Bisphenol A. *Industrial & Engineering Chemistry Research* 2014;**53**: 6855-65.
[17] Yang, L, JF Rathman, LK Weavers. Degradation of Alkylbenzene Sulfonate Surfactants by Pulsed Ultrasound. *The Journal of Physical Chemistry B* 2005;**109**: 16203-9.
[18] Tuziuti, T, K Yasui, J Lee, T Kozuka, A Towata, Y Iida. Mechanism of Enhancement of Sonochemical-Reaction Efficiency by Pulsed Ultrasound. *The Journal of Physical Chemistry A* 2008;**112**: 4875-8.
[19] Petrier, C, M-F Lamy, A Francony, A Benahcene, B David, V Renaudin, et al. Sonochemical Degradation of Phenol in Dilute Aqueous Solutions: Comparison of the Reaction Rates at 20 and 487 kHz. *The Journal of Physical Chemistry* 1994;**98**: 10514-20.

Biography

Raed A. Al-Juboori is a researcher with a background in Chemical and Environmental engineering. Read's research interests are advance oxidation techniques, membrane technology, disinfection techniques, water treatment and alternative energy solutions.