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# Combining COMSOL modeling with acoustic pressure maps to design sono-reactors

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ABSTRACT

Scaled-up and economically viable sonochemical systems are critical for increased use of ultrasound in environmental and chemical processing applications. In this study, computational simulations and acoustic pressure maps were used to design a larger-scale sono-reactor containing a multi-stepped ultrasonic horn. Simulations in COMSOL Multiphysics showed ultrasonic waves emitted from the horn neck and tip, generating multiple regions of high acoustic pressure. The volume of these regions surrounding the horn neck were larger compared with those below the horn tip. The simulated acoustic field was verified by acoustic pressure contour maps generated from hydrophone measurements in a plexiglass box filled with water. These acoustic pressure contour maps revealed an asymmetric and discrete distribution of acoustic pressure due to acoustic cavitation, wave interaction, and water movement by ultrasonic irradiation. The acoustic pressure contour maps were consistent with simulation results in terms of the effective scale of cavitation zones (~10 cm and <5 cm above and below horn tip, respectively). With the mapped acoustic field and identified cavitation location, a cylindrically-shaped sono-reactor with a conical bottom was designed to evaluate the treatment capacity (~5 L) for the multi-stepped horn using COMSOL simulations. In this study, verification of simulation results with experiments demonstrates that coupling of COMSOL simulations with hydrophone measurements is a simple, effective and reliable scientific method to evaluate reactor designs of ultrasonic systems.



## 1. Introduction

Many laboratory studies have reported the chemical processing of materials, water contaminants, and waste streams using ultrasound [1–3]. However, few studies report methods to scale up these bench-scale studies to larger systems. The most commonly used bench-scale device (e.g., horn type probe) for sonication has low energy efficiency, localized cavitation, and a non-uniform acoustic field in the reactor [4–6]. In our previous work, a scaled-up multi-stepped horn was designed and characterized showing higher energy efficiency, multiple cavitational zones, and more widely distributed acoustic pressure as compared to typical horns [7]. To date, there are still limited strategies that have been investigated to design new ultrasonic devices [7,8], improve reactor performance [9–12], and scale up sonolytic processes [13,14].

In the design process, computational simulations are used to investigate how different reactor geometries, horn configurations, and operational parameters (e.g., frequency) impact optimizing performance of ultrasonic systems [15–20]. Of the available computational tools, COMSOL Multiphysics applies a finite element method to solve different physics and engineering problems (e.g., acoustic propagation and heat transfer) governed by partial differential equations (PDEs). The numerous modules and corresponding analytical solutions in the software allow it to combine different phenomena into one model, which is required to simulate ultrasonic systems that feature electromechanical and elastic mechanical effects [21,22]. Therefore, COMSOL Multiphysics has been applied to simulate acoustic fields and sonochemistry in reactors and has provided results consistent with laboratory measurements [15,16,18].

A hydrophone is a piezoelectric device that detects sound pressure underwater and converts the pressure signals to electrical signals. Hydrophone measurements are used to determine an acoustic pressure distribution in solution and through frequency spectral analysis, locate cavitation regions [23–25]. Bubble oscillations in an acoustic field, together with shock waves/micro-jets that follow bubble collapse, introduce many subharmonic/harmonic frequencies and a broad range of frequencies (i.e., background noise) [26–29]. This emitted broadband signal is indicative of transient

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**Nomenclature**

| | |
|---|---|
| $a_n$ | normal acceleration of solid horn (m s$^{-2}$) |
| $A_i$ | amplitude in radius change for *i*th harmonic |
| $c$ | speed of ultrasound propagation in the water (m s$^{-1}$) |
| $c_E$ | elastic coefficients ($6 \times 6$ matrix; Pa) at constant electric field strength |
| $d$ | piezoelectric strain constant ($3 \times 6$ matrix; m V$^{-1}$) |
| $d^t$ | transposed piezoelectric strain constant matrix ($6 \times 3$; m V$^{-1}$) |
| $\mathbf{D}$ | electric flux density vector ($3 \times 1$ matrix; C m$^{-2}$) |
| $e$ | dielectric permittivity ($3 \times 6$ matrix; C m$^{-2}$) |
| $e^{i\varphi}$ | alternating current (AC) |
| $e^t$ | transposed dielectric permittivity matrix ($6 \times 3$; C m$^{-2}$) |
| $\mathbf{E}$ | electric field intensity vector ($3 \times 1$ matrix; V m$^{-1}$) |
| $f$ | frequency of ultrasound (Hz) |
| $f_h$ | bubble oscillation frequency (Hz) |
| $f_R$ | resonance frequency of bubble oscillation (Hz) |
| $\mathbf{F}_V$ | force per volume (N m$^{-3}$) |
| $m$ | integral number |
| $n$ | integral number |
| $\mathbf{n}$ | unit vector |
| $P$ | acoustic pressure (Pa) |
| $P_A$ | maximum acoustic pressure (Pa) |
| $P_{stat}$ | hydrostatic pressure (Pa) |
| $P_{vapor}$ | vapor pressure (Pa) |
| $\mathbf{q}$ | dipole source (m s$^{-2}$) |
| $R$ | bubble radius at time $t$ (m) |
| $R_0$ | bubble radius at equilibrium (m) |
| $s_E$ | elastic compliance ($6 \times 6$ matrix; m$^2$ N$^{-1}$) in a constant electric field |
| $\mathbf{S}$ | strain vector ($6 \times 1$ matrix; m m$^{-1}$) |
| $\mathbf{T}$ | stress vector ($6 \times 1$ matrix; Pa) |
| $t$ | time (s) |
| $\mathbf{u}$ | particle displacement (m) |
| $x$ | defined power series |

*Greek letters*

| | |
|---|---|
| $\alpha$ | characteristic exponent |
| $\beta$ | ratio of driving frequency to bubble oscillation frequency |
| $\gamma$ | ratio of specific heats |
| $\varepsilon_S$ | dielectric permittivity matrix ($3 \times 3$; F m$^{-1}$) at constant mechanical strain |
| $\varepsilon_T$ | dielectric permittivity matrix ($3 \times 3$; F m$^{-1}$) at constant mechanical stress |
| $\mu$ | fluid viscosity (Pa s) |
| $\rho$ | water density (kg m$^{-3}$) |
| $\rho_m$ | material density (kg m$^{-3}$) |
| $\rho_s$ | density of horn rod (kg m$^{-3}$) |
| $\sigma$ | surface tension (N m$^{-1}$) |
| $\varphi$ | phase difference (rad) |
| $\varphi_i$ | phase difference for *i*th harmonic (rad) |
| $\omega$ | angular frequency (rad s$^{-1}$) |

cavitation [30]. Hydrophone measurements of acoustic emissions have been used to characterize acoustic fields and sonochemical reactivity in many ultrasonic systems [23,30,31].

The coupling of computational simulation with mapping the acoustic field using hydrophone measurements provides a method for designing ultrasonic reactors. This work presents a protocol for a sono-reactor design using this coupled method. First, acoustic field surrounding the newly designed multi-stepped horn was simulated in COMSOL Multiphysics to evaluate ultrasound propagation and the resulting cavitation zone in water. The simulation results were then verified using acoustic pressure maps from hydrophone measurements in a plexiglass box, followed by spectral analysis of ultrasound signals to determine the cavitation region and scope. Finally, the configuration of an approximately sized sono-reactor was proposed and modeled. We propose this method for reactor design as a rational way to design and characterize sono-reactors.

## 2. Methodology

### 2.1. COMSOL simulation

An ultrasonic system, composed of a transducer and a horn, involves different physical phenomena [6,21,22]. The piezoelectric material in the transducer converts electricity into mechanical vibrations which pass through the ultrasonic horn rod and are amplified at the end of the horn [22]. These amplified mechanical waves (i.e., ultrasonic waves) are emitted and propagate through a medium, such as water. Thus, three different modules were selected to simulate these physical effects in the COMSOL Multiphysics software (version 4.2): (1) a piezoelectric material module for the transducer; (2) a linear elastic material module for the horn rod; and (3) a pressure acoustics module for water [32,33]. Each module is governed by its own equations that describe the specific physics as discussed in the following section.

#### 2.1.1. Applied physical modules

A piezoelectric effect is a phenomenon in which an applied stress on a piezoelectric material induces electric polarization or an applied electric field induces a dimensional change in the piezoelectric material [34]. In an ultrasonic transducer, the piezoelectric material, often a lead zirconate titanate (PZT) ceramic, generates a mechanical strain under an applied electrical field (i.e., alternating current or AC). Thus, these electromechanical behaviors of the isotropic PZT are expressed by linearized constitutive equations as follows [34,35]:

$$\begin{cases} \mathbf{T} = c_E\mathbf{S} - e^t\mathbf{E} \\ \mathbf{D} = e\mathbf{S} + \varepsilon_S\mathbf{E} \end{cases} \tag{1a}$$

$$\begin{cases} \mathbf{S} = s_E\mathbf{T} + d^t\mathbf{E} \\ \mathbf{D} = d\mathbf{T} + \varepsilon_T\mathbf{E} \end{cases} \tag{1b}$$

where $\mathbf{T}$ is the stress vector ($6 \times 1$ matrix; Pa), $\mathbf{S}$ is the strain vector ($6 \times 1$ matrix; m m$^{-1}$), $\mathbf{E}$ is the electric field intensity vector ($3 \times 1$ matrix; V m$^{-1}$), $\mathbf{D}$ is the electric flux density vector ($3 \times 1$ matrix; C m$^{-2}$), $c_E$ is the elastic coefficient ($6 \times 6$ matrix; Pa) at constant electric field strength, $e^t$ is the transposed dielectric permittivity matrix ($6 \times 3$; C m$^{-2}$), $e$ is the dielectric permittivity ($3 \times 6$ matrix; C m$^{-2}$), $\varepsilon_S$ is the dielectric permittivity matrix ($3 \times 3$; F m$^{-1}$) at constant mechanical strain, $s_E$ is the elastic compliance ($6 \times 6$ matrix; m$^2$ N$^{-1}$) in a constant electric field, $d^t$ is the transposed piezoelectric strain constant matrix ($6 \times 3$; m V$^{-1}$), $d$ is the piezoelectric strain constant ($3 \times 6$ matrix; m V$^{-1}$), and $\varepsilon_T$ is the dielectric permittivity matrix ($3 \times 3$; F m$^{-1}$) at constant mechanical stress.

The vibration generated in the piezoelectric transducer is then transmitted to the horn rod. Assuming both the stainless steel structure of the horn rod and PZT are isotropic and elastic, their linear elastic behavior is governed by Newton's Second Law [32,33]:

$$-\rho_m\omega^2\mathbf{u} - \nabla \cdot \mathbf{T} = \mathbf{F}_V e^{i\phi} \tag{2}$$