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Review

Simulation of the spatial distribution of the acoustic pressure in sonochemical reactors with numerical methods: A review

Ignacio Tudela [a,*], Verónica Sáez [b], María Deseada Esclapez [c], María Isabel Díez-García [c], Pedro Bonete [c], José González-García [b,1]

[a] The Functional Materials Applied Research Group, Faculty of Health and Life Sciences, Coventry University, Priory Street, Coventry CV1 5FB, United Kingdom
[b] Grupo de Nuevos Desarrollos Tecnológicos en Electroquímica: Sonoelectroquímica y Bioelectroquímica, Departamento de Química Física e Instituto Universitario de Electroquímica, Universidad de Alicante, Ap. Correos 99, 03080 Alicante, Spain
[c] Grupo de Fotoquímica y Electroquímica de Semiconductores, Departamento de Química Física e Instituto Universitario de Electroquímica, Universidad de Alicante, Ap. Correos 99, 03080 Alicante, Spain

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ABSTRACT

Numerical methods for the calculation of the acoustic field inside sonoreactors have rapidly emerged in the last 15 years. This paper summarizes some of the most important works on this topic presented in the past, along with the diverse numerical works that have been published since then, reviewing the state of the art from a qualitative point of view. In this sense, we illustrate and discuss some of the models recently developed by the scientific community to deal with some of the complex events that take place in a sonochemical reactor such as the vibration of the reactor walls and the nonlinear phenomena inherent to the presence of ultrasonic cavitation. In addition, we point out some of the upcoming challenges that must be addressed in order to develop a reliable tool for the proper designing of efficient sonoreactors and the scale-up of sonochemical processes.

1. Introduction

Sonochemistry [1] is the area of high-energy chemistry which studies chemical reactions and processes involving acoustic cavitation formed by the application of an ultrasonic field in a frequency range which commonly varies between 20 kHz and 10 MHz. It allows chemists to increase the conversion, improve the yield, initiate and change the reaction pathways in all sorts of biological, chemical or electrochemical processes [2], becoming a prominently used technique in a wide variety of research areas, including: (i) material science [3] (ii) synthetic chemistry [4,5], (iii) water remediation [6,7], (iv) biotechnological applications [8], (v) electrochemical processes [9], (vi) food technology [10], and (vii) spent nuclear fuel reprocessing [11], among others. The versatility of the use of ultrasound in chemistry permits its combination with other technologies such as photocatalysis [12] or microwaves [13], proving the enormous potential of Sonochemistry.

Despite this extensive research at laboratory scale, a limited number of applications have been industrially scaled-up due to two main reasons: (i) the lack of expertise in diverse areas such as ultrasonics or sonochemical engineering, and (ii) the lack of proper reactor designing strategies. Related to this, Sutkar and Gogate have stated that understanding the cavitational activity and its distribution would yield efficiently designed sonochemical reactors and systems [14], and for this purpose, theoretical analysis of the cavitational activity distribution with proper experimental validation could be used for the optimization of sonochemical processes taking into account building materials, geometry of the reactor and working frequency of the sonochemical system. A correct understanding of the acoustic field structure inside a sonochemical reactor is therefore needed to proceed with its optimization and scale-up in order to design efficient large scale reactors [15].

The numerical simulation of the spatial distribution of the acoustic pressure inside sonochemical reactors has widely emerged in the last 15 years to shed new light on this issue, and quite a few groups around the world have tried to model the acoustic field inside sonoreactors with the aim of predicting the cavitation events within the reactor. To our knowledge, the most recent review on this topic found in the literature was published more than 10 years ago [16], and no exhaustive literature revisions are usually found in most of the papers that deal with the simulation of the acoustic field in a sonoreactor. Therefore, the goal of the present paper is to introduce numerical methods for the development of sonochemical reactors to a wider audience of scientists by summarizing the continuous development of numerical methods employed by the scientific community from the late 1990s until now. In this paper, basic

* Corresponding author. Tel.: +44 7521160565.
E-mail address: i.tudela@ua.es (I. Tudela).
[1] Deceased.

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methodologies and results from many works are briefly commented, pointing out the strong and weak points in the most representative cases from a qualitative point of view. And new trends and future challenges on the problem are also discussed.

2. Numerical simulations of the acoustic field inside sonochemical reactors using linear-based models

2.1. Basic linear-based models

The vast majority of the works dealing with the simulation of the acoustic field inside a sonochemical reactor rely on the resolution of the equations that describe the linear propagation of sound in a liquid. Such equations, which are derived from the linearization of the Euler equations [17], yield the well-known Helmholtz equation for the linear propagation of sound waves:

$$\nabla^2 P + k^2 P = 0 \tag{1}$$

being P the acoustic pressure and $k = (\omega/c_l)$ the wave number, where ω is the angular frequency and c_l is the sound speed of the liquid. The Helmholtz equation can be easily solved by setting the next boundary conditions:

- $P = 0$ for 'infinitely soft' boundaries.
- $\nabla P \cdot \mathbf{n} = 0$ for 'infinitely hard' boundaries, being $\mathbf{n}$ the normal vector pointing outward the liquid.
- $P = P_0$ at the emitter surface of the ultrasonic transducer, being P_0 the amplitude of the wave.

The simplicity of the Helmholtz equation and the boundary conditions defined above facilitate the task of calculating the linear propagation of sound in a liquid by numerical methods. In the last years, the wider availability of commercial FEM (Finite Element Methods) software packages presenting acoustic modules based on the Helmholtz equation have allowed an increasing number of different research groups to employ numerical simulations as a powerful tool to design and characterize sonochemical systems.

Among the different commercial software codes available, COMSOL Multiphysics, formerly known as FEMLAB, has probably been the most employed in recent years, not only because the Helmoltz equation and the different boundary conditions are implemented, but also because it does not require deep knowledge on either advanced acoustics modeling or numerical methods. With this software, Sáez et al. [18] tried to characterize a 20 kHz sonochemical reactor by considering the vessel boundaries as infinitely rigid walls and putting special efforts on the discretization of the domain following the rules by Ihlenburg, Babûska and co-workers [19,20]. In their work, the simulations were compared with aluminum foil experiments, observing in both cases a main active zone just along the axial direction located at the emitter center. Their numerical results also indicated a gradual decrease in the ultrasonic field activity with the distance from the emitter, which was roughly confirmed by aluminum foil experiments. Klíma et al. [21] faced the optimization of a 20 kHz sonochemical reactor based on the three-dimensional simulation of the acoustic pressure with the same software. Despite the limitations of the linear Helmholtz equation, which does not take into account non-linear wave propagation and generation of transversal elastic waves, and the consideration of the reactor walls as infinitely soft (which may not be realistic enough), the authors demonstrated that a proper design of the reactor geometry could yield an increase in the acoustic intensity due to multiple reflections rather than the fast decrease in intensity commonly observed when increasing distance from the horn tip (Fig. 1). Bargoshadi and Najafiaghdam [22] tried to optimize an ultrasonics dispersion system using multiple transducers, observing that the optimum distance between two transducers would yield a more uniform distribution of pressure antinodes, and Shao et al. employed basic numerical simulations to estimate the agglomeration position of oxidation inclusions and the ultrasonic field propagation in a ultrasonic cell employed in the purification of magnesium alloy melt [23,24], and the combination of an ultrasonic field with an electromagnetic field for casting AZ80 Mg alloy billets [25]. Kim et al. [26] tried to simulate the temperature and pressure profiles of various solvents at different levels of ultrasonic power by combining COMSOL's acoustic and heat transfer modules. More recently, Thiemann et al. [27] simulated the sound field in a 230 kHz system, observing similar trends in both experimental and numerical works. These simulations, which accounted for partially reflecting boundaries, no only showed the standing wave planes away from the transducer, but also the more complex near field. They also observed that the acoustic pressure amplitude in pressure nodes did not reach zero in the observed standing wave (which indicates the presence of partially absorbing boundaries), being this issue considered by the authors as an hypothetical reason for the cavitation structure experimentally observed.

COMSOL Multiphysics has also been used by different authors in order to solve the Helmholtz equation accounting for damping of the ultrasonic effect by introducing the complex density ρ_c and the complex sound speed c_c:

$$\rho_c = \frac{Z_c k_c}{\omega} \tag{2}$$

$$c_c = \frac{\omega}{k_c} \tag{3}$$

where k_c is the complex wave number and Z_c is the complex impedance. Different expressions can be found in the literature for Z_c and k_c. For example, we can find that

$$k_c = \frac{\omega}{c_l} \frac{1}{\sqrt{1 + (i\omega\mu_l/\rho_l c_l^2)}} \tag{4}$$

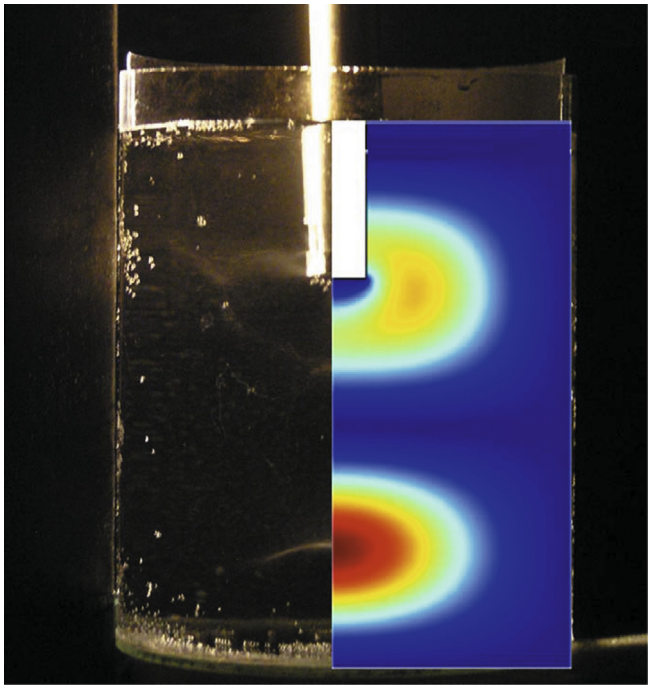

Fig. 1. Photograph of cavitating bubbles in a 20 kHz optimized cell (left) and simulated intensity distribution for the same geometry (right). Reprinted from [21], with permission from Elsevier.