DOI: 10.61782/fa.2025.0926

ULTRASONIC INTENSIFICATION REACTOR APPLIED TO THE AGRI-FOOD AND PHARMACEUTICAL INDUSTRY

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ABSTRACT

Ultrasound has a wide range of applications in fields such as medicine, architecture, or chemistry. In particular, ultrasound-intensified reactors (where chemical and/or physical reactions take place) have proven to be more efficient than traditional reactors in multiple situations, offering a benefit to the industry. This is due to the increased control over the conditions in the mixture, helping to accelerate reactions and replace solvents, among other things. Two examples of this are the agri-food and pharmaceutical sectors. In the first case, ultrasound reactors can improve continuous production, product stability, and allergen removal. In the second, issues such as capillary blockages and crystallization treatments can be mitigated through this technology. This paper presents a preliminary design of an ultrasonic reactor aimed at these applications. The device utilizes a long sonotrode to enhance energy transmission to a continuous flow. This sonotrode consists of a solid cylinder with a machined helical groove along its surface, into which a tube is inserted through which the sample to be treated is passed. Analytical and numerical methods have been used for its design, which have subsequently been reflected at the experimental level for the agri-food and pharmaceutical industry applications.

Keywords: *ultrasound, flow reactor, agri-food industry, pharmaceutical industry*

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1. INTRODUCTION

Ultrasound technology has emerged as a powerful tool across various scientific and industrial sectors, including medicine, chemistry, and engineering. Its application in reactors, particularly ultrasound-intensified reactors, has shown significant advantages over conventional methods [1, 2]. These reactors enhance reaction efficiency by providing precise control over the conditions within the mixture, leading to faster reaction rates and the potential for solvent-free processes. Such benefits have been demonstrated in industries such as agri-food and pharmaceuticals, where ultrasound-assisted techniques improve production stability and mitigate common challenges like crystallization and capillary blockages [1, 3].

The synthesis of particles with controlled properties at the nanoscale and microscale has garnered significant attention due to their diverse applications in fields such as adsorption, ceramics, catalysis, and drug delivery [4]. Among various fabrication methods, continuous flow approaches utilizing microreactors have emerged as powerful tools, offering advantages like enhanced mass and heat transfer, precise control over reaction conditions, and improved reproducibility [5]. However, a critical challenge in continuous flow systems, particularly when synthesizing particles or performing crystallization, is the potential for clogging of the microchannels by solid formation, including the aggregation or precipitation of products or by-products [1, 3, 6].

To address the issue of clogging in continuous flow reactors, several strategies have been explored. These include modifying the wettability of the channel surface, adjusting the viscosity of the solution [7], and employing multiphase flow [1, 5, 7]. Another promising technique involves the application of ultrasound irradiation within the microreactor [1,2]. Ultrasound has demonstrated its effectiveness in preventing and even removing clogging in capillary systems, attributed to the strong cavitation effects it induces [1, 3]. Furthermore, ultrasound can enhance mass transfer rates at gas-liquid or liquid-solid interfaces and promote mixing, which can be particularly beneficial in processes like continuous crystallization and nanoparticle synthesis [3, 5, 6].

In this context, a significant need exists for robust and scalable continuous flow reactors. Particularly, they can be applied to the agri-food and pharmaceutical industries as they can effectively handle solids and prevent clogging [3]. While continuous flow reactors offer numerous advantages, such as precise control and enhanced mass transfer [4, 6], their widespread adoption is hindered by challenges related to solid handling, including clogging from precipitate formation, solid catalysts, or insoluble byproducts [3].

To address these limitations, the design of a large-scale flow reactor should incorporate strategies for efficient solid management. Ultrasound irradiation emerges as a particularly promising technique for both preventing and clearing blockages in continuous flow capillary systems [3]. Ultrasound can induce cavitation, which provides a mechanical means to disrupt particle aggregation and resuspend settled solids [5, 8].

Notably, even low-power acoustics without cavitation has been shown to effectively unclog and prevent blockages [3].

The fabrication of such a reactor should consider several key aspects. The choice of materials must ensure chemical compatibility with the intended applications in the agri-food and pharmaceutical sectors. For efficient ultrasound transmission, materials such as glass can be advantageous [5]. The reactor design should incorporate optimized sonotrode geometries, such as blade, block, cylindrical, or even helicoidal shapes, to achieve wider and more uniform sonication areas suitable for larger reactor volumes and longer capillary lengths [3]. Acoustic design principles are crucial to ensure that the applied frequency resonates effectively with the reactor system, maximizing the conversion of electrical power into acoustic energy focused on the reaction channels. Finite element analysis (FEA) can be a valuable tool in the design process to simulate and optimize the acoustic field distribution within the reactor [6].

Validation of the large-scale flow reactor will be essential to confirm its efficacy in handling solids and preventing clogging while maintaining process performance. This should include characterizing the hydrodynamics of the reactor using residence time distribution (RTD) measurements to ensure predictable flow behavior, potentially approaching plug flow to minimize dispersion [8]. Techniques like high-speed microscopic imaging can visualize flow patterns and the behavior of solid particles under different operating conditions and with the application of ultrasound [3]. The reactor's ability to handle multiphase flow and maintain suspension of solid catalysts or reagents should be rigorously evaluated [5]. Furthermore, the performance of the reactor should be tested in relevant applications, such as continuous crystallization of pharmaceutical compounds with control over particle size and polymorph [9], and continuous catalytic reactions involving solid catalysts [5], demonstrating sustained, clog-free operation and desired product quality.

By focusing on integrated design, incorporating effective solid handling strategies like ultrasound, and thorough validation, the development of large-scale continuous flow reactors can be significantly advanced for the benefit of the agri-food and pharmaceutical industries. Nonetheless, their widespread adoption is hindered by challenges related to solid handling, including clogging from precipitate formation, solid catalysts, or insoluble byproducts [3].

Thus, ultrasound-intensified flux reactors serve as a tool to study the effects of ultrasound irradiation on substances of interest. Flux reactors, in contrast to ultrasonic batch reactors, allow continuous operation, which is beneficial at the industry level.

The reactors that will be studied consist of a system which contains a Langevin transducer (or two) coupled to a long sonotrode. This sonotrode consists of a solid aluminum cylinder with a helical machining along its surface, into which a tube is inserted through which the sample to be treated is passed by means of a peristaltic pump. The objective of this study is to design, build and experimentally test this kind of reactors.

The first section will be dedicated to the analytical and numerical design of the reactor. The second section will treat the experimental part, in which the experimental setup will be described, as well as the methodology and results.

2. DESIGN OF THE SONOTRODE

The system will consist of a Langevin transducer coupled to a cylindrical sonotrode, which will be an aluminum cylinder with a length of 630 mm, with a machined helical groove on its surface to locate the tube. The first design was developed considering a 60 W Langevin transducer with two piezoelectric ceramics PZT8. The scheme of the system with its dimensions is shown in Fig. 1.

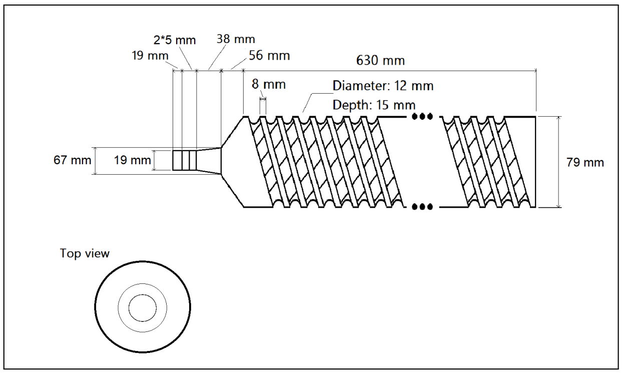

Figure 1. Initial scheme of the system.

Nonetheless, new high-power Langevin transducers were bought (with 4 piezoceramics PZT8), with dimensions different from those of the original Langevin transducer. Thus, the final scheme of the system has a step between the cone and the Langevin transducer, as shown in Fig. 2.

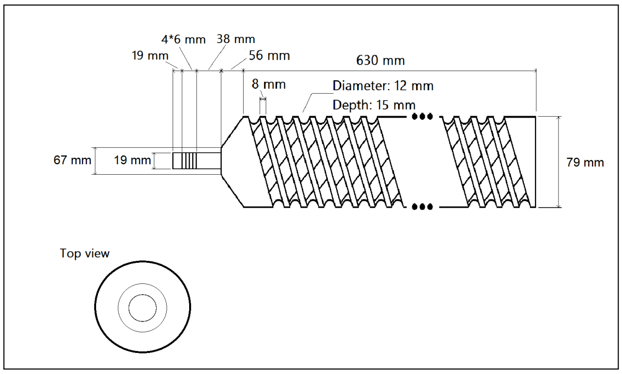

Figure 2. Initial scheme of the system.

2.1 Analytical design

The design of the system was made through the Mason model. The system was modeled to be resonant at 28 kHz. The equivalent circuit of the system can be seen in Fig. 3. An approximation has been used for the analytical solution. It has been considered that there is not a helical slit but a separated transversal ring slit sequence, in a way that there are maximum and minimum radius variations for the section. The part of the circuit corresponding to this ring sequence (the sonotrode) is shown in Fig. 4.

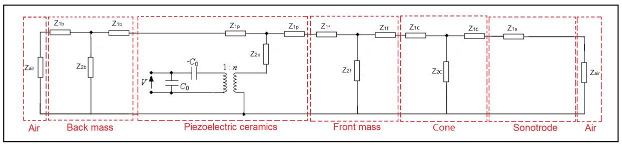

Figure 3. Equivalent circuit for the entire system. The sonotrode part is indicated as an equivalent impedance.

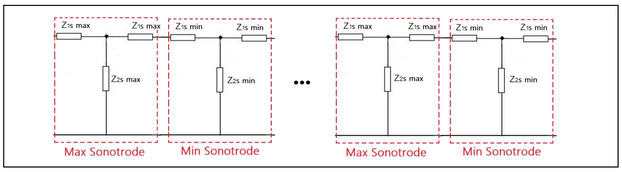

Figure 4. Equivalent circuit for the sonotrode part, where it is considered the sequence of maximum and minimum radius section of the sonotrode as if they were coupled blocks of different sections.

The result of the impedance of the system as a function of the frequency according to this method was solved using Python. The result is shown in Fig. 5, where it can be seen that the resonance of the system is approximately 28 kHz.

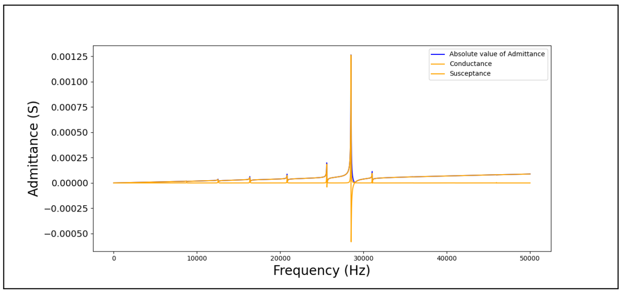

Figure 5. Absolute value of admittance, as well as conductance and susceptance of the system according to Mason model.

2.2 Numerical design

The system has also been numerically simulated in order to obtain the optimum design. The first attempt was to optimize the system at a frequency of 28 kHz. The software COMSOL Multyphysics has been used. The libraries used for this model have been "Solid mechanics", "Electrostatics" and "Piezoelectric effect". The result of this first attempt can be seen in Fig. 6, in which the transducer coupling to the sonotrode has been enlarged for better visualization.

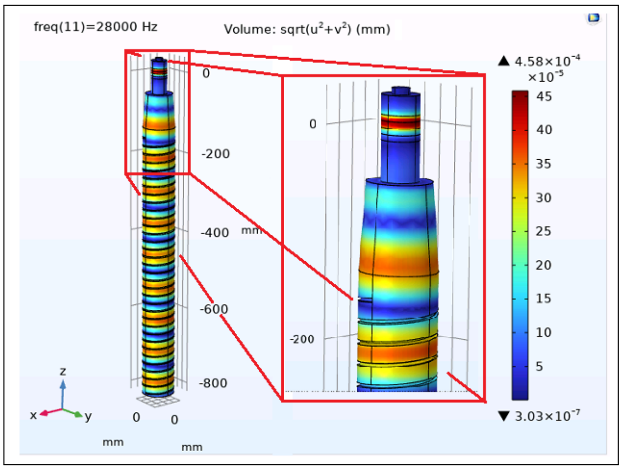

Figure 6. Numerical result of 28 kHz frequency response of the system. The parameters u and v correspond to the displacement on the x-axis and on the y-axis, respectively. Thus, $\sqrt{u^2+v^2}$ gives the total radial displacement of the system.

The absolute value of admittance, the conductance and the susceptance of the system has been computed from the numerical model by COMSOL Multphysics. The result is shown in Fig. 7.

Later, more designs were developed in order to increase the acoustic power. Thus, the new designs were mainly symmetric with two transducers of different resonant frequencies (20 kHz and 28 kHz), one in each side of the cylinder, as shown in Fig. 8, in which the transducer coupling to the sonotrode has also been enlarged for better visualization. Symmetric systems were modeled in 2D axisymmetric because they require less computational power and, due to the symmetry of the problem, the desired vibration modes can be studied in this way. In Fig. 9 the built sonotrodes are shown.

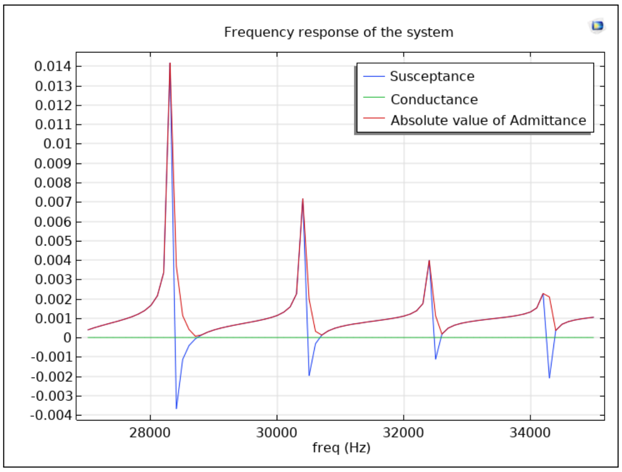

Figure 7. Numerical result of the absolute value of admittance, the conductance and the susceptance of the system. The result is not exactly the same as the one shown in Fig. 5, because that was an approximation.

3. EXPERIMENTS

The scheme of the experimental setup is shown in Fig. 10. The sample is forced to pass through the PFA tube by a peristaltic pump.

In order to control the experimental system, a LabVIEW program has been created. This program consists of a system of 3 tabs, each one in charge of a specific task. In the first tab, frequency sweep is performed in order to find the resonant frequencies of the system experimentally. In the second tab, the emission at the optimum frequencies can be controlled, making it possible to emit only at the desired frequency or at multi-frequency, with a continuous or totally customized pulsed signal. The third tab provides basic control of the pumps and monitoring of the potential pressure sensors and flow meters that can be added to the system.

Once the experimental setup has been completed (Fig. 11), tests can be started. The methodology is the same for all the experiments: the sample is passed through a tube coiled around the sonotrode, driven by a peristaltic pump. The pump flow rate and the type of ultrasound treatment (continuous or pulsed) can be changed depending on the sample and the purpose of the experiment.

In the laboratory, it has only been possible to perform preliminary studies which need to be systematically repeated in order to obtain more accurate and solid conclu-

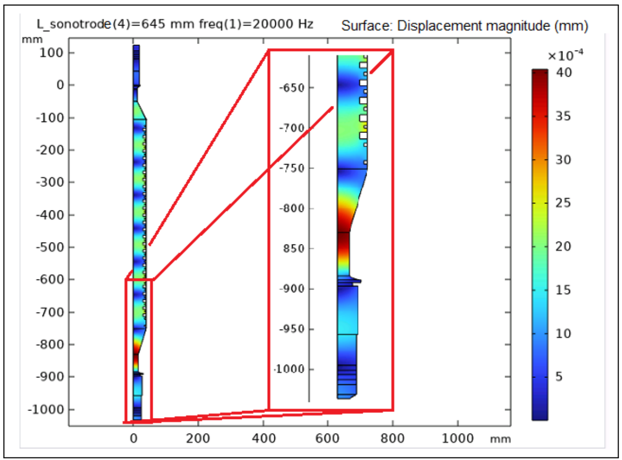

Figure 8. Numerical result of 20 kHz frequency response of the symmetric system.

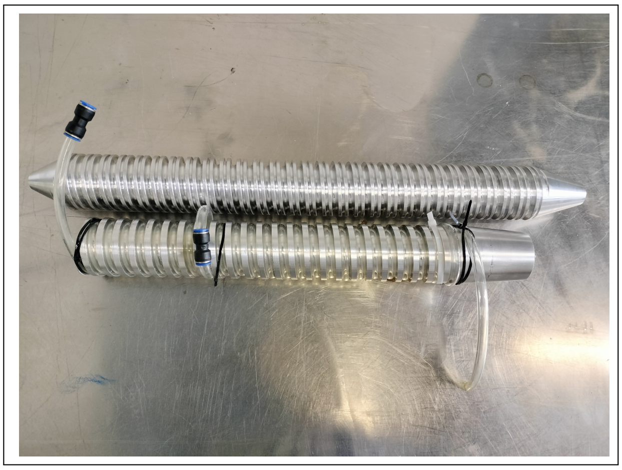

Figure 9. Built sonotrodes.

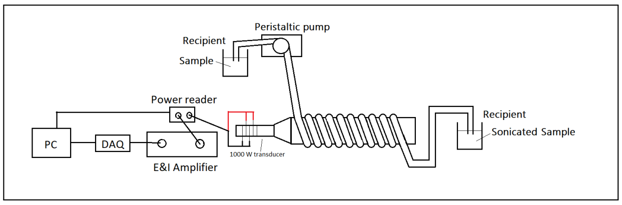

Figure 10. Experimental scheme (not to scale).

sions. Thus, only qualitative results can be summarized in this work.

The experiments performed coincide with the main problems found in continuous production in the industry, which are mentioned earlier. Thus, each experiment is a test to verify if the built device is able to solve the corresponding problem.

3.1 Antifoaming capacity

To check if the system is able to remove the foam, first it has to be generated. To create foam, the sample for the liquid-gas system was beer. When ultrasounds were applied some effects on the bubbles were observed, mainly the reduction of the large ones.

3.2 Unclogging capacity

The purpose of this experiment is to create a clog, and check if the ultrasound application can remove it. The sample for the liquid-solid system was water with sand.

The generation of the clog is made by means of a 3D printed tube reducer. The inner diameter of the tube goes from 2.5 mm to 1 mm in one single step.

When ultrasounds are applied, the effectiveness of removing and preventing clogs was qualitatively observed.

3.3 Homogenization of dispersed systems

The objective of this experiment is to increase the homogeneity of dispersed systems.

The sample for the dispersed system was pineapple juice. When ultrasounds are applied, a slight reduction of the juice pulp was observed.

More experiments are needed in order to prove the actual reduction of the juice dispersion.

3.4 Viscous systems

The objective of this experiment is to reduce the viscosity of the sample, in order to make it easier to transport. This is the case of sauces, for example. The sample in this case was mayonnaise.

When ultrasound is applied, a clear reduction in viscosity (slight liquefaction) was observed.

Another objective of this experiment is to test whether the device is capable of generating stable emulsions. This has not been made yet in the laboratory.

More quantitative, replicable and representative experiments are needed in order to properly test the proposed ultrasonic flux reactor in an industry context.

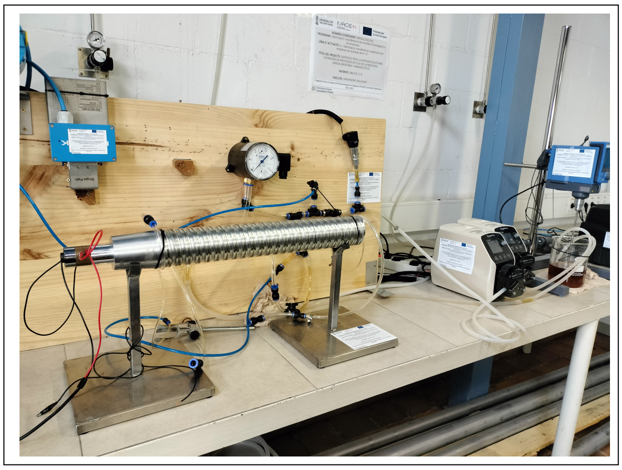

Figure 11. Experimental setup.

4. CONCLUSION

The application of ultrasonic reactors in the agri-food and pharmaceutical industries is explored, highlighting their advantages in enhancing reaction efficiency, preventing clogging, and improving product quality. The study presents the design and testing of a novel ultrasonic reactor, featuring a solid cylindrical sonotrode with helical slit for optimized energy transmission. The potential of the reactor has been experimentally suggested in terms of foam reduction, unclogging capacity, and liquefaction of sauces, although there are limitations in dispersion improvements. The results suggest that ultrasound-assisted reactors could be a valuable tool for industrial processes, offering potential scalable and effective solutions for continuous flow applications.

5. ACKNOWLEDGMENTS

This project (INNVA1/2022/36) was financed by the Agència Valenciana de la Innovació (IVACE)/Programa Fondo Europeo de Desarrollo Regional (FEDER) Comunitat Valenciana 2021-2027.

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