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Broadening the sonochemistry horizon: hurdles and challenges to address in cavitation

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This article provides an overview of the current challenges associated with cavitation, highlighting the technological and experimental limitations in elucidating complex bubble dynamics. It also examines how the limited availability of experimental data constrains the development of numerical models. Additionally, the paper reviews recent advances in cavitation and their influence on the development of physical and chemical technologies, with a particular focus on sonochemical applications.

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Introduction

The pursuit of improved performance and sustainability across chemical and physical processing has driven the exploration of cavitation as a mechanism to enhance technologies in the medical, biomedical, and chemical fields [26,5,28]. The formation and subsequent collapse of transient cavitation bubbles in a liquid presents a unique opportunity to induce physical and chemical transformations under conditions that are challenging to achieve using conventional techniques [32,60,12,14]. Cavitation bubbles collapse under strong pressure differences with the surrounding liquid (Figure 1), which may result from ultrasonic waves, laser pulses, shock waves, or liquid flow. The collapse triggers a plethora of phenomena, including microjetting, shock wave radiation, localized temperature and pressure increases, and light emission [48,49,33]. Events such as microjetting and shock waves induce the mechanical action of bubbles with benefits mainly for physical processing. Temperature rise and likely light emission aid the occurrence of chemical reactions within the bubble and in the liquid bulk.

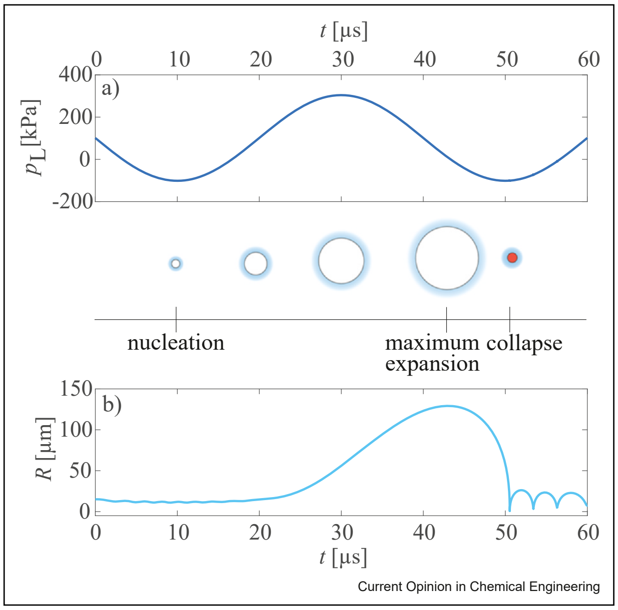
The mechanical effects of collapsing bubbles have been extensively studied and have received more attention over the past few decades. It is therefore unsurprising that many cavitation-based technologies for physical processing have been successfully implemented in real-world applications. Prominent examples include wastewater treatment [54,55], chemical extractions [4], and lithotripsy [13]. In contrast, industrial adoption of sonochemical technologies, especially concerning sonochemical synthesis, has progressed more slowly, although most often providing a greener solution with respect to conventional technologies. This is largely due to the relatively low efficiency of sonochemical reactors, as well as a limited control over the bubble collapse and comprehension of the underlying phenomena.

Control of cavitation collapse

Significant advances made in controlling bubble collapse for cavitation-based technologies are increasingly finding real-world applications. Yet, the understanding of the bubble dynamics and the resulting phenomena remains incomplete. The initial potential energy of the bubble, taken as the potential energy at maximum expansion, is distributed among all collapse-related phenomena, such as shock waves, microjetting, and chemical reactions. These damp the bubble oscillation, whereas the remnant energy contributes to the bubble rebound. The partition of the energy is dictated by the collapse conditions [51]. For example, a bubble collapsing in a nonhomogeneous pressure field deviates from a spherical collapse. A more pronounced bubble deformation lowers the fraction of the bubble's initial potential energy deployed for, among the others, the emission of shock waves in favor of microjetting.

Ongoing research aims to enhance the effectiveness and precision of cavitation-related technologies by achieving an accurate control over the various collapse phenomena, with an energy partition tailored to the specific

Figure 1



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(a) Liquid pressure p_L oscillation due to oscillating acoustic pressure and **(b)** single bubble radius R as a function of time t. A graphical representation of the bubble evolution is also included. Simulation was conducted using the [16] model with the following parameters: initial nucleus radius 15 μm, frequency of acoustic pressure 25 kHz, and pressure amplitude twice the atmospheric pressure. All other liquid physical constants were set to those of pure water.

application. In sonochemical processes, each bubble acts as a microreactor. Thus, maximizing energy density within the bubble through spherically symmetric bubble collapse, and therefore maximizing bubble temperature, is crucial to enhance process efficiency while minimizing energy loss to the surrounding liquid [33]. Inducing a more spherically symmetric collapse in a cavitating fluid requires control over several factors, such as liquid density, bubbles radii, cavitation driving pressure, nearby free or rigid surfaces, and fluid flow [49]. Moreover, precise control over higher-order deformation effects, such as gas–liquid instabilities, is sought, which negatively influence the compression efficiency of the bubble in the final moments of collapse [3,33]. Paradoxically, increasing the amount of reactants inside the bubble reduces the liquid contraction during implosion, which in turn diminishes both bubble deformation and collapse strength. An optimal sonochemical process requires a delicate balance between these factors, necessitating an ideal bubble composition that provides enough matter for chemical reactions while preventing excessive deformation and minimizing any negative impact on implosion strength. Finally, the properties of the cavitating liquid have been shown to significantly influence bubble collapse, thereby affecting various cavitation-related applications [29].

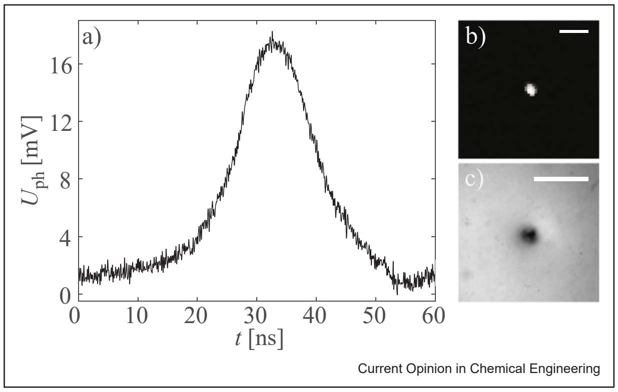
Limitations of cavitation experimental setups

The limited understanding of the final collapse stages is mainly due to the limited spatiotemporal resolution of experimental equipment. Figure 2a shows the light emission signal from a cavitation bubble that lasts for only ~20 ns, highlighting the rapid nature of phenomena at bubble collapse. Capturing and resolving the highly nonlinear behavior of cavitation bubbles, especially during the final collapse stages, present significant challenges. Visualizing bubbles at this stage requires cameras with extremely high acquisition rates (millions of frames per second) paired with complex optical systems due to the supersonic velocities of the bubble–liquid interface and the small size of the bubble [44]. A detailed visualization of subsurface interfaces such as microjets is also challenging. Additionally, optical access is hindered at the final collapse stage by light emission due to luminescence (Figure 2b) and optical refraction caused by pressure buildup at the bubble interface (Figure 2c).

Experimental challenges extend to describing the internal bubble contents, where direct measurement of the gaseous composition and behavior is prohibitive. The bubble composition, whether comprising non-condensable gas, condensable vapor, or a mixture of the two, remains largely unknown. Likewise the temporal evolution of the bubble contents stays concealed.

Consequently, an accurate control of the bubble contents is also complicated. However, a rough modification of the bubble contents is possible by directly modifying the surrounding liquid. For instance, changing the liquid temperature plays a fundamental role in the bubble

Figure 2



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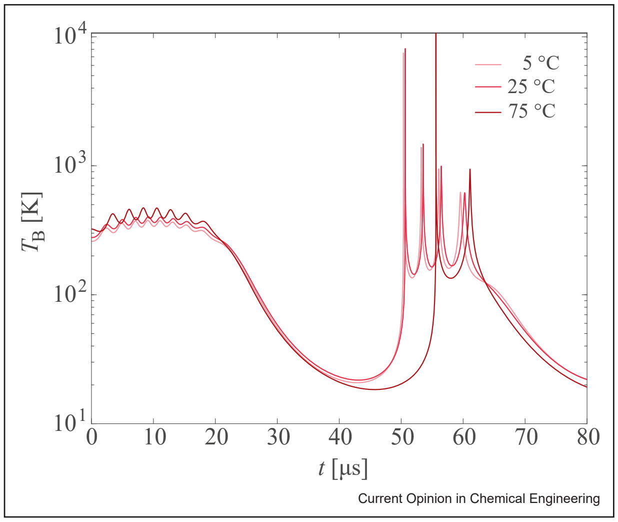
(a) Electric signal U_{ph} as a function of time t showing the light emitted at the collapse of a laser-induced bubble, collected using a photodiode with a 1-ns rise time. **(b)** Snapshot captured in dark conditions at the final instants of the collapse, showing light emission from a laser-induced bubble with a maximum radius of around 4 mm. **(c)** Shadowgram of a laser-induced bubble of maximum radius of around 1.5 mm at the instant preceding the final collapse. The white solid lines indicate the 1-mm scale.

Figure 3



Temporal evolution of bubble internal temperature T_B from the Keller-Miksis model for different initial liquid temperatures of 5, 25, and 75°C. The water vapor pressure was set to 873 Pa, 3169 Pa, and 38 563 Pa, respectively. All other inputs were consistent with those used in Figure 1.

contents [30]. It influences vapor pressure and gas dissolution, which directly impact the bubble's internal composition, hence the collapse strength (Figure 3).

Challenges in numerical simulation

The development of theoretical and numerical models has been pivotal in advancing cavitation collapse control and improving cavitation-based technologies. The temporal evolution of a spherical bubble radius can be described using Rayleigh–Plesset-based ordinary differential equations:

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{p_b - p_\infty}{\rho} - \frac{2S}{\rho R} + \phi \tag{1}$$

where R is the bubble radius function of time t, p_b is the bubble internal pressure, p_∞ is the liquid pressure in the far field, which equals the liquid static pressure p_0 plus any acoustic pressure p_{ac} if present, and S is the liquid surface tension. The dot notation indicates time derivatives. ϕ is an oscillation-damping term that includes effects from factors such as liquid compressibility, viscosity, phase change, and chemical reactions. More complex models can be extended to account for damping due to bubble deformation.

The effectiveness and reliability of numerical models for cavitation are often constrained by the technological limitations in experimental research. The unknown bubble composition is a major factor of uncertainty in cavitation simulations, often requiring strong assumptions behind their choice. This hinders the estimation of temperature and pressure inside the bubble. More accurate predictions will require detailed knowledge of the initial composition of the bubble nuclei, interface phenomena throughout the lifetime of the bubble [31], and complex chemical reaction schemes dependent on temperature and pressure [15].

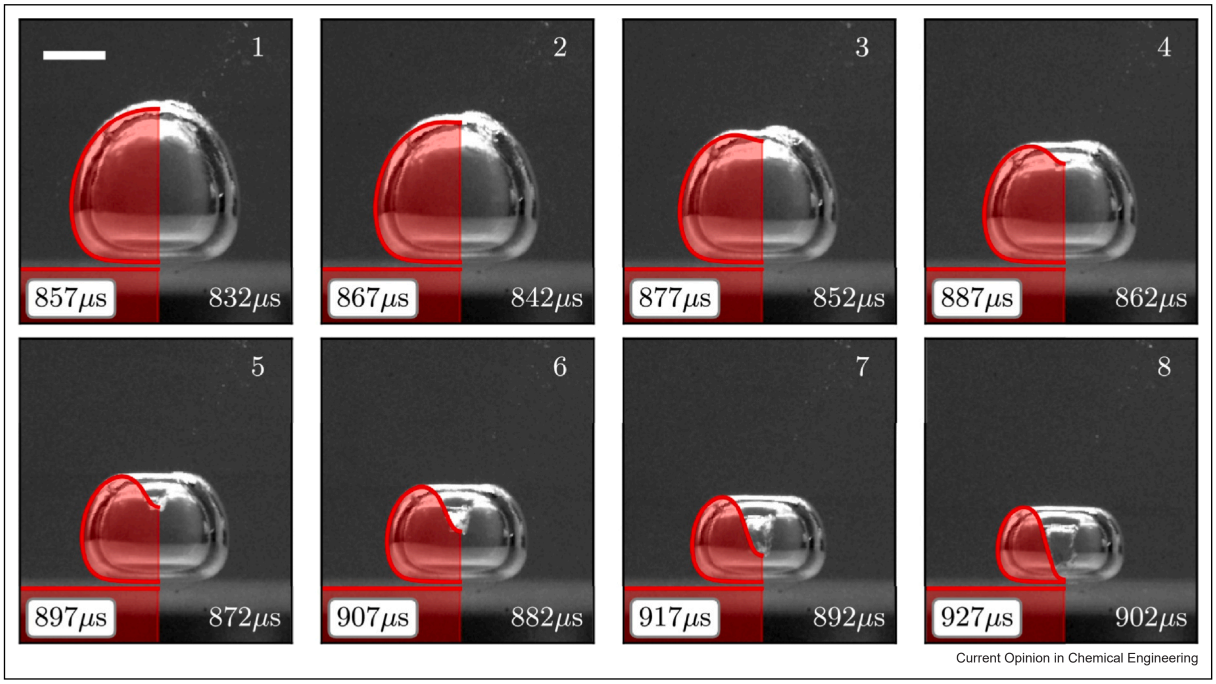
Although numerical simulations have achieved high accuracy in predicting bubble dynamics, these models still struggle to resolve the behavior of bubbles near their minimum volume where the radius can differ by more than an order of magnitude from its maximum expansion [39].

An accurate description of the thermodynamic properties inside the bubble is also essential. Selecting an appropriate equation of state is critical for reproducing bubble collapse accurately, ensuring proper coupling between the gas and liquid phases. Equations of state are also crucial for determining the peak temperature and pressure at collapse and estimating interface temperatures. For example, a comparative work between the Tait and the Noble–Abel Stiffened-Gas equation of state [7] has shown how the employment of the latter can provide a more consistent prediction of cavitation thermal effects. Furthermore, chemical reactions inside collapsing bubbles are often modeled under local thermal equilibrium. However, this assumption may not hold throughout the entire lifetime of the bubble, as the volume rate of change may become faster than the vapor-to-liquid phase change [1,33]. Such conditions may lead to nonequilibrium or supercritical states during collapse, whose effect on the sonochemical yield is unknown. Additionally, the reaction schemes themselves pose challenges. These reactions may involve multiple reactants depending on the liquid nature and dissolved gases, and produce a range of products that potentially become new reactant species in successive bubble cycles [15]. Furthermore, it is still unclear whether a particular reaction occurs inside the collapsing bubble, in the bulk of the liquid due to radical recombination, or both [27].

Multibubble cavitation

A key assumption for many models is that the bubble remains spherically symmetric. However, this is invalidated when a non-uniform pressure field develops around the bubble, resulting from the presence of solid or free surfaces or pressure gradients in the liquid [49]. These factors lead to deviations from sphericity during the collapse and the associated development of microjets. This behavior is typical in real-world applications, where bubbles frequently nucleate as part of clouds. Deformed cavitation bubbles have been extensively studied experimentally [48,43,40], and their dynamics

Figure 4



Snapshots of a single laser-induced bubble near a rigid boundary. The red regions in the left halves of the frames represent numerical simulations performed using a BIM, seamlessly superposed onto the experimental data. The simulations accurately capture the dynamics of the bubble, including the formation of the microjet. The white line indicates the 2-mm scale. Illustration from Ref. [45].

can be accurately simulated. Numerical simulations based on the Boundary Integral Method (BIM) and the Volume of Fluid method [38,45,20] provide a more sophisticated alternative to overcome the spherical symmetry assumption (Figure 4).

Nevertheless, numerical models still need improvements. Capturing the formation of sharp gas–liquid interfaces often requires more sophisticated modeling approaches [39]. Additionally, higher-order deviations from sphericity can arise due to the onset of gas–liquid interface instabilities. These instabilities present significant challenges for both experimental visualization and numerical modeling due to their characteristic length scales being several orders of magnitude smaller than the bubble radius. Furthermore, deformations have been observed to affect collapse convergence, negatively impacting sonochemical reaction rates [34].

On the experimental side, one challenge of multibubble cavitation is related to the stochastic nucleation of bubbles as a result of the randomness of the nuclei spatial distribution into the liquid. Excessive nucleation is also an undesirable phenomenon, especially for ultrasound-induced cavitation, as bubbles in the proximal region are responsible for shielding sound energy at the distal region, thereby reducing collapse intensity [8]. Moreover, quantifying cavitation and tracking the bubble dynamics within a cloud is practically impossible. In this case, macroscopic investigations of features such as the spectrum of the noise emissions prove useful for cavitation analysis [47].

Recent advances in cavitation and sonochemistry

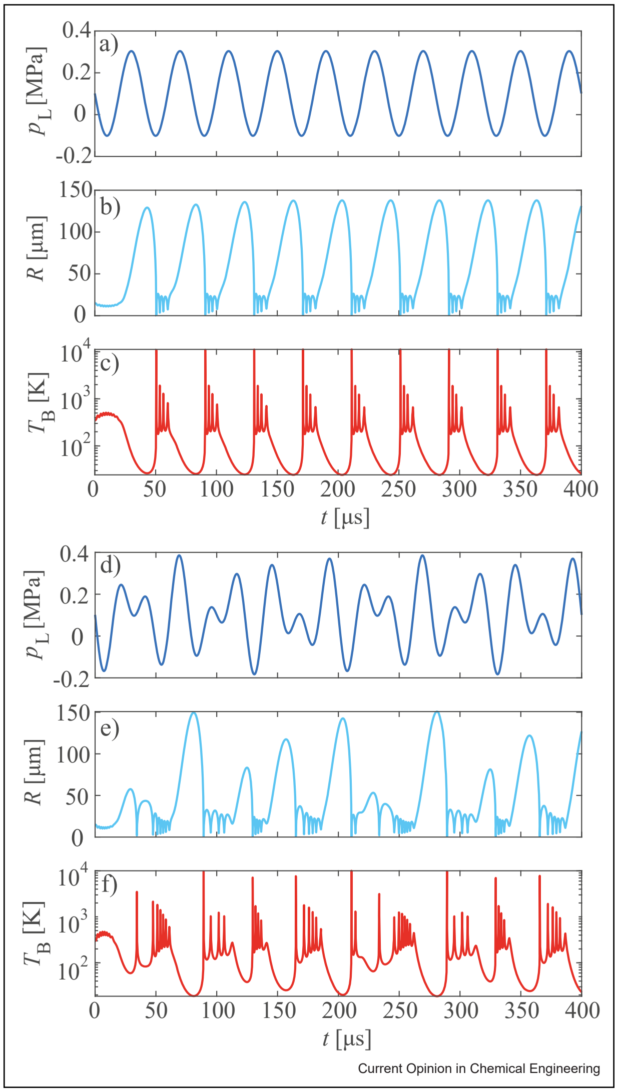
As mentioned, the past few decades have witnessed significant progress in mechanical processes related to cavitation. The most recent advancements in the field of cavitation have shed light on various phenomena. Novel indirect methods have been employed to understand bubble contents [6,21,52], and a plasma-based technique was used to initially attempt the direct measurement of the bubble contents [24,53]. The latter technique was applied to millimeter-sized laser-induced cavitation bubbles, as their position relative to the plasma initiation site could be precisely controlled. Studies have also offered new insights into the development of thin supersonic microjets in bubbles collapsing near different surfaces [37,43]. In parallel, a synchrotron-based X-ray back illumination was employed to obtain optical access to subsurface details of cavitation bubbles. X-ray imaging, whose temporal resolution has considerably improved over the last years (order of magnitude of the MHz), revealed intricate details about the development of sharp interfaces, such as liquid jets, which are hardly captured with conventional techniques [2]. Efforts to tackle the stochastic nature of cavitation nucleation have also progressed. Ultrasound-responsive agents such as microbubbles, nanodroplets [41], gold nanoparticles [9,42], and gas-stabilizing nanoparticles [19] have emerged as effective tools. These agents enable on-demand cavitation generation, with precise control over the number, size, and location of the bubbles, facilitating a wide range of applications.

A significant theoretical development introduced a unifying framework to describe single cavitation bubbles [61]. The model incorporates boundaries, bubble interactions, ambient flow fields, gravity, bubble migration, fluid compressibility, viscosity, and surface tension. From a numerical perspective, a notable milestone is the parallel simulation of a cloud containing 12,500 collapsing cavitation bubbles [36]. Models have also improved in simulating chemical reactions, achieving greater accuracy in predicting reaction products and kinetics [15,18].

Lastly, progress has been made in developing more efficient sonochemical reactors [35,59], though much of this remains confined to laboratory-scale systems. Most advancements are related to experimental work, through which diverse sonochemical reactors have been investigated. Recent advancements in reactor design have aimed to enhance cavitation effects. While focused ultrasound reactors are more complex and difficult to scale up, they have proven to be more efficient than unfocused reactors. This suggests that reactor design plays a crucial role in improving sonochemical efficiency [59]. Additionally, when operating in burst mode, the number of cycles in ultrasound-induced cavitation becomes a key factor influencing the performance of sonochemical reactors [58]. Besides, hydrodynamic cavitation is also considered a promising technology for process intensification, offering high energy efficiency, cost-effective operation, and a great potential for scalability [62]. Numerical tools such as COMSOL Multiphysics have advanced the analysis of these complex systems [10,57], although these tools still cannot fully account for cavitation-related phenomena, often treating cavitation as a 'black box.'

It is also particularly noteworthy that multi-frequency ultrasonic technologies have demonstrated superior performance compared to single-frequency systems in various research domain. Numerical studies revealed that dual-frequency ultrasound reduces the inertial cavitation threshold [11,23], enhances mass transfer at the bubble–liquid interface [56], and allows for larger maximum bubble expansion [22]. Experimental validation, while limited, supports these findings [46,50]. In sonochemical reactors, fine-tuning frequencies in dual-frequency mode boosted the total radical yield compared to single-frequency excitation set at the lower frequency [25]. Although dual-frequency excitation resulted in lower collapse temperatures, there was an increase in sonochemical activity [17]. The collapse temperature dynamics for single- and dual-frequency excitation (Figure 5) can be examined using the adiabatic [16] model in the absence of chemical reactions. As expected, single-frequency excitation led to more frequent peak-temperature events, but dual-frequency mode produced a higher number of collapses in the same time frame.

Figure 5



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(a) Liquid pressure p_L oscillation due to oscillating acoustic pressure, **(b)** single bubble radius R, and **(c)** bubble internal temperature T_B at collapse as a function of time t. **(d)**, **(e)**, and **(f)** are the corresponding data with excitation in dual-frequency mode, respectively. All inputs for the simulations were consistent with those used in Figure 1. The simulation with dual-frequency excitation was at 25 kHz and 40 kHz.

Conclusion and perspectives

This article has explored the current challenges in cavitation and their implications for technological and scientific advancements, with a particular focus on sonochemistry. Overall, a broader comprehension of cavitation-related phenomena and a more accurate control of the bubble collapse was identified as the primary need for the continued development of medical, biomedical, and chemical technologies. Numerical modeling has emerged as a pivotal tool in bridging knowledge gaps, despite being constrained by limited computational power and the scarcity of experimental data. These limitations highlight the pressing need for parallel advancements and integration of experimental techniques and numerical methods. To this end, a major obstacle to overcome in numerical modeling is the implementation of multibubble systems with three-dimensional spatial resolution capable of accurately accounting for all cavitation-related phenomena. Experimentally, overcoming the short spatial and temporal scales of cavitation bubbles remains a significant technological challenge, with the aim of being able to clarify fundamental unknowns of cavitation bubbles related to their dynamics and internal composition. These limitations emphasize the importance of fostering interdisciplinary research within the scientific community, which is critical for advancing cavitation research across applications with diverse needs. From a sonochemical perspective, there is an urgent need to develop real-time cavitation monitoring techniques, more energy-efficient systems, and scalable reactor designs in order to expand the application of sonochemical reactors beyond laboratory settings.

Commercial applications of sonochemical technologies are most likely on the horizon, and a widespread industrial deployment will be possible as long as research continues on this path. A clear correlation exists between the insights provided by existing literature and their translation into contemporary industrial practices. The mechanical effects of collapsing bubbles, which have been extensively studied and understood over the past few decades, are already being exploited in chemical extraction, wastewater treatment, and biomedical technologies. Given this trajectory, it is reasonable to anticipate that sonochemical processes will achieve broader industrial adoption within the next decade, further solidifying the role of cavitation in advancing modern technologies.

Ethical approval

Not required.

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Data Availability

Data will be made available on request.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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- • of special interest
- •• of outstanding interest

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