Intense Cavitation at Extreme Static Pressure

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1 Cavitation is usually performed at hydrostatic pressures at or near 0.1 MPa. Higher static
2 pressure produces more intense cavitation, but requires an apparatus that can build high
3 amplitude acoustic waves with rarefactions exceeding the cavitation threshold. The absence
4 of such an apparatus has prevented the achievement of intense acoustic cavitation,
5 hindering research and the development of new applications. Here we describe a new high-
6 pressure spherical resonator system, as well as experimental and modeling results in water
7 and liquid metal (gallium), for cavitation at hydrostatic pressures between 10 and 150 MPa.
8 Our computational data, using HYADES plasma hydrodynamics code, show that, under
9 these conditions, the formation of dense plasma reaches peak pressures of about three to
10 four orders of magnitude greater than the hydrostatic pressure in the bulk liquid and
11 temperatures in the range of 100,000 K. Passive cavitation detection (PCD) data validate
12 both a linear increase in shock wave amplitude and the production of highly intense
13 concentrations of mechanical energy in the collapsing bubbles. High-speed camera
14 observations show the formation of bubble clusters from single bubbles. The increased
15 shock wave amplitude produced by bubble clusters, measured using PCD and fiber optic
16 probe hydrophone, was consistent with current understanding that bubble clusters enable
17 amplification of energy produced.

Keywords: High intensity cavitation; Ultrasound; Acoustic resonator; High pressure;
Bubble dynamics; Shock wave

1. Introduction

Acoustic cavitation is the growth and collapse of cavities in a liquid driven by an acoustic wave: the growth is caused by the negative pressure, and the subsequent inertial collapse is driven by the combination of the positive-pressure phase of the wave and hydrostatic pressure [1]. Even at a hydrostatic pressure of 0.1 MPa, the pressure inside the collapsing bubbles can rise to more than 100 MPa and the temperature can reach several thousand degrees [1-8]. These conditions enable the cavitation bubbles to act as micro reactors with rapid heating and cooling rates to initiate, speed up, or enhance chemical reactions and provide a range of capabilities including producing allotropic transformations and synthesizing novel materials [5-11]. It is reasonable to expect that these transformational capabilities will be increased with more intense cavitation. Previous studies have shown that cavitation intensity can be increased by producing cavitation at high static pressures [12-19]. Here we describe a system to produce cavitation at extreme static pressures and provide illustrative experimental and numerical results of this system between static pressures of 10 and 150 MPa.

2. Materials and methods

2.1. Acoustic resonators

To achieve cavitation at extreme static pressures, we significantly changed our previous designs [12-19] of high-quality-factor spherical resonators that, with relatively small consumption of power, produce high-amplitude standing acoustic waves. Resonators have varied between 6-inch (15.24 cm) and 15-inch (38.1 cm) outer diameter (OD), with larger ODs operating at lower frequencies for stronger cavitation. The state-of-the-art is the 15-inch OD resonator operated at 16 kHz. The quality factor of the resonators is affected by the number, position and design of the access ports (e.g., for filling and draining, inserting hydrophones and other probes), windows and acoustic drivers. All of these components were significantly changed following numerous tests and modeling with COMSOL multiphysics.

The resonators were pressurized using high-pressure generators (HiP, High Pressure Equipment Company), which are essentially manually operated piston screw pumps. For the experiments above 30 MPa static pressure, we motorized the HiP generator and controlled the static pressure using a program written in IGOR Pro (IGOR Pro 6, WaveMetrics, USA). In these experiments, static pressure was monitored using two calibrated pressure transducers (PX91N0-60KSV, OMEGA Engineering, USA and AST47HPX60000P2D0123, American Sensor Technologies, USA) with digital panel meters M20020DCV1 (Micron Meters, USA). In the experiments below 30 MPa, we used analog gauges (10,000 psi, ACCO Helicoid Gauge, USA), as shown in the bottom right of Fig. 1, as well as digital gauges (MP40A series, Micron Instruments, USA).

Fig. 1. Schematic of a typical experimental configuration and a photograph of a 9.5-inch windowed resonator used for experiments with a high-speed camera (top right).

The resonators were driven by piezoelectric transducers, connected to RF power amplifiers (1140LA, Electronic Navigation Industries, USA). The amplifiers were fed by a function/arbitrary waveform generator (33120A, Agilent, USA) controlled by a computer using a program written in IGOR Pro.

The resonators were filled with filtered and degassed liquids. In selected experiments, argon or deuterium gas was added to the system. For experiments from 30 to 150 MPa the resonators were filled with a liquid metal, gallium, maintained at temperature around 40°C. The photography and high-speed imaging data are from water-filled, windowed resonators operated at room temperature at 10 to 30 MPa static pressure, which is the safety limit of the sapphire windows required for visual observations.

2.2. High-speed imaging and shadowgraph photography of shock waves

High-speed videos were recorded using a Phantom v710 camera (Vision Research, USA) with 16 GB memory. The images were recorded either at 210,526 frames per second (fps) with 128 x 128 pixel resolution or at 397,660 fps with 128 x 64 resolution. In one sequence, the camera captures about 342,000 consecutive frames at 210,526 fps or 685,000 frames at 397,660 fps, producing videos of about 1.6-1.7 seconds.

The images were taken using a Nikon 50 mm F1.8 lens and 13 mm extension tube. Illumination (Fig. 1, right panel) was provided using a 250 W gooseneck photo light (North Star, USA) for side lighting and a 50 W light with a diffusion disc for backlighting. Shadowgraph photography of shock waves was performed using a parallel beam of light produced by a 5 mW laser (532 nm,

LightVision Technologies) and a 10X beam expander (Melles Griot). Propagation speeds of shock waves from collapsing bubbles were determined by measuring the radial expansion of shock waves seen as expanding shadowgraphic circles between the consecutive frames. To reduce the uncertainty of the measurements, we used average values of circle diameters measured at different angles.

2.3. Measurements of absolute pressure

Absolute pressure was measured using a self-calibrated fiber-optic probe hydrophone (FOPH-2000, RP acoustics, Germany) [20]. The sensitivity of the hydrophone was determined by measuring a DC photodetector signal at normal atmospheric pressure without acoustic signal and using an internal light scattering factor of the fiber-optic system that was measured at regular intervals. The photovoltage-pressure relation (provided by the manufacturer) was derived using isentropic Tait parameters for water and Gladstone-Dale relation between refractive coefficient and density, as well as correction factors for compression of glass and water temperature. Since the FOPH shows the absolute pressure, we also checked FOPH readings of static pressure against calibrated pressure gauges (20,000 psi, McDaniel Controls, Inc., USA) in 0.1-138 MPa pressure range. The readings agreed with an accuracy of 5%.

The FOPH uses a 140- μm diameter glass fiber with a 100- μm diameter sensitive core, making the hydrophone virtually omnidirectional up to MHz range. The FOPH fiber was inserted in high-pressure resonators through a 3/8-inch NPT port using 1/16-inch OD stainless steel tubing. To minimize acoustic interference from the tube, the fiber tip typically extended about 3 cm beyond the end of the tube. Recorded traces were deconvoluted with the FOPH impulse response provided by the manufacturer and processed using programs written in LabVIEW (National Instruments, USA).

2.4. Passive cavitation detection

Passive cavitation detection [21,22] was performed using polyvinylidene fluoride (PVDF) sensors (LDT0-028K/L, Measurement Specialists, USA). The PVDF sensors are 28 μm piezoelectric films with rectangular (10 mm by 14 mm) sensitive elements and silver ink screen-printed electrodes, laminated to a 125 μm polyester layer with crimped-on solder tabs for electrical connections. The PVDF sensors were mounted on the outer surface of the resonators and had the same spherical curvature as the resonator shell, thereby focusing the sensors to the cavitation zone at center of the resonator (Fig. 1). This focusing ensured that shock waves radiated by cavitation bubbles reached different parts of the sensor at the same time, allowing us to maximize signal detection.

2.5. Laser nucleation

In some experiments, bubbles were nucleated using a Q-switched Nd:YAG laser (Minilite 1, Continuum) with a wavelength of 532 nm, pulse duration of 3-5 ns, and energy adjustable up to 12 mJ. The laser beam was focused through a sapphire window into the center of a high-pressure spherical resonator. To adjust the firing time of the laser relative to the phase of the acoustic wave, we designed gate and phase adjuster electronics.

2.6. Numerical model

Cavitation bubbles were modeled as spherically symmetric objects using a HYADES hydrocode (Cascade Applied Sciences) [23]. HYADES is a radiation hydrodynamics and dense plasma code with the following main assumptions: 1) all energy transport is modeled using the diffusion approximation; 2) the radiation is coupled only to the electron fluid; and 3) the electron and ion components are treated separately in a fluid approximation and are loosely coupled to each other, each in thermodynamic equilibrium and described by Maxwell-Boltzmann statistics. HYADES does not model gas diffusion, so the amount of gas in the bubble is determined by the initial radius of the bubble. For simulations discussed in this report, we used argon bubbles in water and deuterium bubbles in gallium with an initial radius from 5 to 50 μm . The initial temperatures were 297 K for water and 300 K for gallium. The thermodynamic properties were simulated using the following equations of states (EOS) from the SESAME database (Los Alamos National Laboratory): EOS 315 for water, EOS 93 for argon, EOS 552 for gallium, and EOS 151 for deuterium. We modeled cavitation with 16-64 zones in the bubble and 1000-1500 zones in the liquid, restarting the code twice with decreasing zone sizes for finer resolution near the collapse. Since one-dimensional HYADES code can model only a single, spherically symmetric bubble and cannot model three-dimensional bubble clusters observed in subsequent acoustic cycles, we limited the modeling to one acoustic cycle. The frequency of the acoustic wave was 25,982 Hz in water and 50,443 Hz in gallium, corresponding to the spherically symmetric (0,6) mode of the 9.5-inch resonators. For this mode, the standing wave amplitude at the center of the resonator is nearly 11 times higher than at the resonator wall. The wave amplitude at the resonator wall was typically set to 10% of static pressures, producing a net negative pressure near the center of the resonator to drive the growth of bubbles.

3. Results and discussion

3.1. Numerical results

Our modeling of cavitation under extreme static pressures shows high temperatures and pressures inside and in close proximity to a collapsing bubble. Video 1 and Fig. 2 show numerical results obtained with the HYADES hydrocode for an argon bubble in water at 20 MPa static pressure. The bubble grew to a maximum radius of about 0.834 mm with the growth-collapse cycle lasting about 14.5 μs (not shown). Upon collapse, the pressure at the center of the bubble spiked to 250 GPa for about 30 ps (not shown) and remained, for about two nanoseconds, above 175 GPa (Video 1), which is four orders of magnitude greater than the hydrostatic pressure in the bulk liquid. The temperature at the center of the bubble reached about 85,000 K (second frame, Fig. 2). High temperatures and pressures ionized the gas, producing plasma with density almost five times the density of the surrounding water.

Video 1. Numerical modeling of pressure, density, and temperature during the collapse of a cavitation bubble at 20 MPa static pressure.

After reaching the maximum temperature and pressure, the bubble continued to implode until the pressure wave, originated at the center of the bubble, hit the bubble wall, producing a supersonic shock wave in the surrounding liquid. For example, the speed of the shock wave between the fourth and fifth frame in Fig. 2 was 6550 m/s, which is 4.3 times faster than sound in water at 20 MPa static pressure. Due to absorption, the spherical shock waves are attenuated faster than $1/r$, where r is the radial distance from the bubble. For example, increasing the distance 1.69 times, from 46.9 μm (frame 4, Fig. 2) to 79.3 μm (frame 5, Fig. 2), resulted in an attenuation of the shock by 1.84 times (from 28.2 GPa to 15.3 GPa). Despite this attenuation, the shock waves in close proximity to the bubbles are still very intense—approximately three orders of magnitude greater than the hydrostatic pressure in the bulk liquid. Similar calculations were previously reported for a bubble filled with water vapor [13].

Conditions at higher static pressures are more intense. For instance, at 30 MPa static pressure, the pressure at the center of the collapsing bubble spiked to 4,400 GPa for about 35 ps duration and remained, for a nanosecond, above 800 GPa, which is about four times greater than at 20 MPa. The maximum density in the bubble reached ~ 8 g/cc. The temperature reached its maximum at the center of the bubble of about 200,000 K. The bubble radiated a shock wave that has an amplitude of about 660 MPa at 0.7 mm and 43 MPa at 7 mm from the bubble. Hence, a 10 times increase in distance was associated with ~ 15 times reduction of shock wave amplitude. That is, in this range, this spherical shock wave attenuated as $(1/r)^{1.18}$ due to nonlinear absorption.

At 110 MPa static pressure, the bubble grew to a maximum radius of about 1.76 mm. Upon collapse, the pressure at the center of the bubble spiked to $\sim 8,300$ GPa for 30 ps, and remained above 1500 GPa for about a nanosecond. The density reached ~ 20 g/cc, and the temperature $\sim 380,000$ K. The radiated shock wave had an amplitude of ~ 5.6 GPa at 0.7 mm from the bubble and 146 MPa at 7 mm from the bubble, that is, a 10 times increase in distance was associated with ~ 38 times reduction of shock wave amplitude, attenuating as $(1/r)^{1.58}$. Comparison of the shock wave amplitudes at 30 MPa and 110 MPa show that at 0.7 mm from the bubble the shock at 110 MPa was about 8.5 times greater than at 30 MPa, while at 7 mm shock amplitudes differ about 3.4 times. This difference in shock wave amplitude continued to decrease, albeit to a lesser extent, as the shock waves propagated further from the bubbles [24,25].

Fig. 2. Selected frames from Video 1 showing temperature (thin solid line), pressure (thick solid line), and density (dotted line) inside and in close proximity to a collapsing bubble at 20 MPa static pressure. The vertical axis shows pressure, density and temperature normalized by their maximum values: $P_{\max}=228$ GPa, $\rho_{\max}=5.24$ g/cc and $T_{\max}=84,900$ K. The horizontal axis shows radial distance from the bubble center. Arrows show directions of motion of the gas-liquid interface (vertical line) and shock wave (SW, last two frames). Zero time corresponds to the minimum radius, $R_{\min}$, of the bubble (third frame).

Numerical simulation in gallium showed a linear increase of shock amplitude with static pressure (Fig. 3). The slope of the curve is determined by the cavitation threshold—the driving acoustic amplitude that produces spontaneous cavitation in the liquid. This result is obtained under an assumption that the cavitation threshold grows linearly with static pressure [16,17]. For this simulation, the driving amplitude at the resonator wall was set to 10% of static pressure, as was done in the above simulation in water.

Fig. 3. Shock amplitude as a function of static pressure in gallium at 0.7 mm from the bubble. Trend line equation and coefficients of determination, R^2 , are shown bottom right on the chart.

3.2. Bubble dynamics

Cavitation starts spontaneously when the acoustic field ruptures the liquid creating a bubble at or near the center of the resonator. Alternatively, a bubble can be deliberately nucleated, e.g., by a laser pulse [15,18,25-27], as in Fig. 4, or by a neutron source [12,28]. Regardless of the nucleation mechanism, the bubble is expanded by the negative pressure (frames 1-4, Fig. 4) and continues to grow under the positive pressure due to inertia of the liquid (frames 5-6, Fig. 4). Then, forced by the positive-pressure phase of the acoustic wave and high static pressure, the bubble rapidly collapses (frame 8, fast moving water/air interface appears blurred). Upon collapse, the bubble reaches high pressures and temperatures, emitting sonoluminescence (SL, frame 8) [2-4,6,12,18,19,29], and produces a micro jet [2,19] (traceable in video 2), generating cavitation nuclei that become visible under the negative pressure of the next acoustic cycle (Fig. 4, frame 10). These daughter bubbles grow and coalesce (frames 11-15), forming a cluster that collapses and produces, in this instance, a cloud of ~20 micro bubbles (frame 19). Bubble proliferation [30] continues through subsequent cycles of the acoustic wave, adding more bubbles to the bubble cluster.

Fig. 4. Cavitation bubbles during three acoustic cycles at 10 MPa static pressure (top). This high-speed camera sequence was recorded using backlight so that bubbles appear black. The bottom graph shows the timing of high-speed camera frames relative to the phase of the driving acoustic wave (illustrated as a sinusoid at the driving frequency of ~ 26 kHz). The first bubble was laser nucleated at the beginning of the negative-pressure phase of the driving acoustic wave (laser pulse can be seen in frame $-2.51 \mu\text{s}$ in video 2).

[Video 2. Backlight video of growth and collapse of cavitation bubbles for three acoustic cycles at 10 MPa static pressure.](#)

Bubbles at their maximum expansion for 12 acoustic cycles are shown in Fig. 5. A spontaneously nucleated bubble (first frame) gives birth to a cluster of bubbles that grows in size as the number of bubbles increases from cycle to cycle. Bubble dynamics for 27 acoustic cycles are shown in Video 3. After a few tens of growth-collapse cycles, cavitation ceases. We postulate that this is because cavitation bubbles exhaust the energy of the standing acoustic wave and detune the resonator due to changes in density and compressibility caused by the presence of bubbles and heat generated from collapses. Then, typically within hundreds of milliseconds (depending on driving power), the standing acoustic wave regains its strength, repeating the above process of growth and collapse of cavitation bubbles.

Fig. 5. Cavitation bubbles during 12 acoustic cycles (~ 26 kHz, 12 MPa static pressure). This high-speed camera sequence was recorded using both back and side illumination. Bubbles are shown at their maximum expansion; growth and collapse are shown in Video 3.

[Video 3. Sidelight video of growth and collapse of cavitation bubbles for 27 acoustic cycles at 12 MPa static pressure.](#)

3.3. Radius-time curve from the HYADES code and experimental data

The laser nucleated bubble at 10 MPa static pressure (Fig. 4 and video 2, frames 0-17.6 μ s) was used to measure radius-time curve and compare it with the HYADES hydrocode (Fig. 6). For HYADES simulations, we used bubbles with 5 μ m and 50 μ m initial radii. The amplitude of the driving acoustic wave ($f=25,987$ Hz) was set to 1.1 MPa at the resonator wall, which for this mode of the resonator results in about 12 MPa acoustic amplitude at the center of the resonator. At this driving amplitude, the 5 μ m bubble grew to a maximum radius (R_{max}) of 0.63 mm, and the 50 μ m grew to 0.72 mm, while the experimental value for R_{max} was 0.66 mm.

Fig. 6. Bubble radius versus time from the HYADES code and experimental data (crosses) at 10 MPa static pressure. Solid curves show HYADES simulation for 5 μ m and 50 μ m bubbles. Vertical lines show the measurement uncertainty of bubble radius. Horizontal lines show the 2.1 μ s exposure time of the high-speed camera images. The 2.5- μ s step high-speed camera sequence of this bubble is shown in Fig. 4 and video 2 (frames 0-17.6 μ s).

3.4. Absolute pressure

Absolute pressure in high-pressure spherical resonators was measured by the fiber-optic probe hydrophone positioned at approximately 11 mm from the center of the resonator. At this distance, the amplitude of the driving acoustic wave is about 22% lower than at the center of the resonator. This can be seen in the radial distribution of acoustic pressure calculated for the (0,6) mode of the resonator [12-19] and shown in the top right inset of video 4 (two frames are shown in Fig. 8).

Fig. 7 shows a hydrophone trace at 20 MPa static pressure for two cavitation events. The first cavitation event (top-left enlargement in Fig. 7 and video 4) decreased the acoustic wave so that the rarefaction stopped producing net negative pressure, ceasing the cavitation. Starting from 0.04 s, the growing standing wave started producing net negative pressure, but cavitation did not occur until ~ 0.123 s. This second cavitation event (top-right enlargement, Fig. 7) occurred when the acoustic wave amplitude exceeded the cavitation threshold.

At the above static pressure, the cavitation threshold was greater than 30 MPa. Although this threshold also depends on other experimental conditions (such as presence of cavitation nuclei, amount of the dissolved gas, type and purity of the liquid) [16,17,30], these observations reinforce the notion that the cavitation threshold grows with static pressure. Furthermore, Fig. 7 shows that the standing acoustic wave grows only until it ruptures the liquid and creates cavities. Postponing the onset of cavitation by using high static pressure enables the generation of higher amplitude standing acoustic waves, therefore achieving more intense cavitation.

Fig. 7. Absolute pressure measured by fiber-optic probe hydrophone positioned at 11 mm from the center of the resonator at 20 MPa static pressure during two cavitation events.

3.5. Validation of the numerical model

While spectroscopic analysis of sonoluminescence has traditionally been used to assess the temperature at the outer boundary of a bubble, this method is not suitable for measuring the temperature of the optically dense plasma at the core of the collapsing bubble [3] or in opaque liquids. In contrast, evolution of pressure waves originating at the core of the bubble can be measured and compared with the numerical model. For example, the model predicts that a bubble with a maximum radius of ~ 0.8 mm at 20 MPa static pressure radiates a shock wave that has an amplitude of ~ 18 MPa at 9.2 mm from the bubble. In experiments, the fiber-optic probe hydrophone measured 17 ± 2 MPa at that distance (movie 3). This shock wave was radiated by a spontaneously nucleated bubble that grew to a maximum radius of 0.7 ± 0.1 mm (frame -7.54 μ s, video 4).

Furthermore, this shock wave was captured by high-speed shadowgraphs at two radial distances from the bubble: 1.9 ± 0.1 mm (Fig. 8, left) and 6.2 ± 0.1 mm (Fig. 8, right). The ratio of the distance travelled (4.3 ± 0.2 mm) to the time interval between the frames ($2.515 \mu\text{s}$) gives the average velocity of the shock wave of 1710 ± 80 m/s, which is about $12 \pm 5\%$ faster than the sound speed (1525 m/s at 20 MPa static pressure). For comparison, the model predicts a speed of 1750 m/s, which is about 13% faster than sound at 20 MPa static pressure. This correlation of measured and calculated values for shock speed and amplitude validates the numerical model.

Fig. 8. Two consecutive frames of video 4 (left and right) showing shadowgraphs of a shock wave (top) radiated by a single cavitation bubble spontaneously nucleated at 20 MPa static pressure. The absolute pressure is shown in the middle (one acoustic cycle) and bottom (~ 31 acoustic cycles) panels. The absolute pressure was recorded by a fiber-optic probe hydrophone positioned at about 11 mm from the center of the resonator (the tip of the hydrophone is faintly visible at the top right corner of the shadowgraphs). The high-speed shadowgraphs were recorded $2.515 \mu\text{s}$ apart.

[Video 4. High-speed shadowgraph imaging, shock waves, and absolute pressure during cavitation at 20 MPa static pressure.](#)

3.6. Bubble clusters and returning shock waves

Video 4 shows simultaneous recordings of high-speed shadowgraph images, shock waves, and absolute pressure for cavitation at 20 MPa static pressure. A single cavitation bubble was spontaneously nucleated near the center of the resonator during the maximum of the negative pressure phase of the driving acoustic wave (absolute pressure during one period of the driving acoustic wave is shown in the middle panel and highlighted yellow at the bottom). The bubble became visible in the next frame ($-12.57 \mu\text{s}$), reached the maximum radius of 0.7 ± 0.1 mm (frame $-7.54 \mu\text{s}$), and collapsed at $-2.51 \mu\text{s}$, radiating a shock wave that propagated from the bubble (seen as the circles in shadowgraphs at $-2.51 \mu\text{s}$, $0.00 \mu\text{s}$, and $2.51 \mu\text{s}$). During the collapse the bubble produced a microjet and moved in the direction of the jet (from right to left in these images), generating daughter microbubbles (faintly visible) that served as cavitation nuclei for the next acoustic cycle. The daughter microbubbles started to grow upon arrival of the negative-pressure phase of the driving acoustic wave at $17.60 \mu\text{s}$, forming an almost spherical cluster, which is apparent in images with higher magnification (Figs. 4 and 5). The cluster reached the maximum size at $32.69 \mu\text{s}$, and collapsed at $40.24 \mu\text{s}$, radiating stronger shock waves (frames $40.24 \mu\text{s}$ and $42.75 \mu\text{s}$).

Radiated shock waves, reflected from the resonator wall, converge back to the center of the resonator, nucleating new clusters of bubbles. For example, the shock wave radiated by the first bubble at $-2.51\ \mu\text{s}$ returns back to the center of the resonator at $130\ \mu\text{s}$ (video 4), the convergence of this returning shock wave is visible in frames $123.22\ \mu\text{s}$ and $125.74\ \mu\text{s}$. It formed a new cluster of bubbles at $128.25\ \mu\text{s}$. Since this cluster was formed during the positive-pressure phase of the driving acoustic wave, it collapsed almost immediately (between frames) radiating a shock wave at $130.76\ \mu\text{s}$, but then started to grow at $133.28\ \mu\text{s}$ driven by the negative-pressure phase of the acoustic wave, reaching a larger size and collapsing at $158.43\ \mu\text{s}$. This shows the importance of the arrival time of the returning shock waves.

The bubble clusters grow larger, live longer, and therefore collapse later than solitary bubbles (video 4). As a result, clusters of certain sizes radiate shocks that return back during the negative-pressure phase of the driving acoustic wave. These returning shocks produce significantly larger clusters of bubbles. For example, the shock emitted by the cluster at $40.24\ \mu\text{s}$ is seen to converge back to the center of the resonator in frames at $163.46\ \mu\text{s}$, $165.97\ \mu\text{s}$, and $168.49\ \mu\text{s}$, producing a larger cluster of bubbles (171.00 - $183.57\ \mu\text{s}$, video 4). The cluster starts to collapse at about $186.09\ \mu\text{s}$ radiating numerous shock waves as observed in the shadowgraphs and recorded by the fiber-optic probe hydrophone (frames $188.6\ \mu\text{s}$ - $203.69\ \mu\text{s}$).

Growth and collapse of bubble clusters show that the clusters start to collapse from the periphery to the center [31], radiating shock waves that intensify the collapses of the central cavities (video 4, Fig. 9). As a result, the cavities at the center of the cluster emit the brightest light (last frame, Fig. 9) and the strongest shock waves (video 4). These stronger shocks propagate with higher velocities and catch up to the less intense shock waves, often evolving into one larger shock. These combined shocks reflect off the resonator wall and converge at or near the center, producing bubble clusters in subsequent acoustic cycles (video 4).

Fig. 9. Bubble cluster dynamics during one acoustic cycle ($\sim 26\ \text{kHz}$, $30\ \text{MPa}$ static pressure).

3.7. Transient and stable cavitation

Since bubble cluster cavitation typically lasts on the order of a few tens of acoustic cycles (video 3), we have reported them as transient cavitation under these conditions [12]. In addition to the transient cavitation, we also observed stable cavitation, when one or several small bubbles oscillated with the driving acoustic field for many acoustic cycles. Stable cavitation bubbles were typically two orders of magnitude smaller than the clusters in transient cavitation.

Stable cavitation was typically observed in liquids with large concentrations of dissolved gases and at lower driving acoustic field amplitudes than typically required for transient cavitation.

Stable cavitation was also seen between transient cavitation events, when a few small bubbles continued to oscillate while the acoustic field grew to produce the transient cavitation. Stable cavitation could spontaneously evolve into transient cavitation, at times, without obvious reasons. We postulate that this transition from stable to transient cavitation was caused by the constructive interference of the returning shock waves emitted by stable cavitation bubbles. This rationale is evidenced in the previous section that showed the significance of timing and amplitude of the returning shock waves for generation of bubble clusters in transient cavitation.

3.8. Cavitation intensity at different static pressures

Extreme static pressures produced substantially more intense cavitation. Video 5 shows transient cavitation at 7 MPa (left) and 30 MPa (right) static pressures. This video shows every fourth acoustic cycle, skipping the time needed for the shocks to return (~ 3.5 acoustic cycles at this mode of the resonator) and drive the growth of bubbles. Two frames of the video 5 are shown in Fig. 10. The bubble cluster at 30 MPa static pressure (Fig. 10, right) was about six times larger in size, ~ 200 times in volume, than the cluster at 7 MPa static pressure (Fig. 10, left). Hence, a four-fold increase in static pressure (from 7 to 30 MPa) increased the potential energy (pressure times the maximum volume of the cluster) by three orders of magnitude, which is evidenced by the stronger shock waves (section 3.6). This shows that the ability to cavitate at extreme static pressures has the potential to release previously unachievable levels of energy for chemical and physical reactions.

Fig. 10. Cavitation bubble clusters at 7 MPa (left) and 30 MPa (right) static pressures. Higher static pressure (right panel) generated larger clusters of bubbles (top frames show clusters at maximum expansion) that produced stronger collapses with brighter sonoluminescence (bright spots at the center of the clusters, bottom images). Growth and collapse of these cavitation clusters are shown in Video 5.

331 [Video 5. High-speed video of bubble clusters at 7 MPa and 30 MPa static pressures.](#)

3.9. Illustrations of strength of cavitation at high static pressures

332 The intensity of high static pressure cavitation is illustrated in Fig. 11. At 10 MPa static pressure,
333 cavitation severed a 0.9-mm diameter stainless steel wire within a few seconds (Fig. 11a),
334 demonstrating the high energy density of our system. Transient cavitation at 20 MPa severed the
335 wire in less than half a second (Fig. 11b and Video 6). Even the stable cavitation at extreme static
336 pressures was strong enough to sever the wire in about a minute at 30 MPa static pressure. In
337 contrast, cavitation from a conventional ultrasonic horn, operated at the same (~200 W) electric
338 power for ten minutes at normal atmospheric pressure (0.1 MPa), produced only a small amount
339 of pitting faintly detectable using a 10X microscope.

340 [Video 6. High-speed video of a 0.9-mm diameter stainless steel wire severed by cavitation at](#)
341 [20 MPa static pressure in less than half a second.](#)

(a)

(b)

Fig. 11. (a) Stainless steel wire severed by high-pressure cavitation at 10 MPa static pressure. (b) Growth and collapse of bubble cluster during one acoustic cycle at 20 MPa static pressure. Cavitation at this static pressure severed the wire within 0.5 s (selected frames are shown in Video 6).

3.10. Cavitation at 30-150 MPa static pressure

Acoustic cavitation at 30-150 MPa static pressure was reliably achieved at various driving frequencies in the 16-50 kHz range using non-windowed spherical resonators of three different sizes: 9.5-, 12- and 15-inch OD. This cavitation was produced in gallium with and without adding argon and deuterium gas. By adding the gas into the system and adjusting the driving amplitude, we were able to control generation of both transient and stable cavitation regimes (section 3.8), evidenced by signals from passive cavitation detectors (PCD). Typical PCD signals from transient cavitation events are shown in Fig. 12. An increase of static pressure was associated with the increase of number and amplitude of detected shock waves. For instance, in the example shown in Fig. 12, the PCD signal at 30 MPa had only about seven strong spikes with the peak-to-peak amplitude growing up to about 0.8 V (top left), while at 150 MPa there were about 25 strong spikes with the peak-to-peak amplitude growing up to 2.2 V (top right).

PCD data also shows a four-cycle periodicity of the radiated shock waves (Fig. 12), beginning when cavitation is spontaneously initiated and the collapsing bubble emits a shock wave. This emitted shock wave reflects off the shell of the resonator and returns 3.5 acoustic cycles later, producing a bubble cluster that collapses, emitting a shock wave at the fourth cycle from the original shock wave (Fig. 12, bottom). Subsequent returning shock waves nucleate bigger bubble clusters that, in turn, radiate new, stronger shock waves four cycles later (Fig. 12).

As bubble clusters collapse, emitting shock waves, they also produce cavitation nuclei (Section 3.2). These nuclei can dissolve, or substantially diminish in size, especially under extreme static pressures, to the extent that they themselves are not sufficient to produce cavitation in the next acoustic cycle (Fig. 12, bottom). Eventually, however, bubble clusters are present to collapse every acoustic cycle, most likely because, as bubble clusters grow larger, they collapse later during the positive pressure phase of the acoustic cycle (video 4, Section 3.7). As a result, the produced daughter microbubbles have less time to dissolve—increasing the probability that cavitation nuclei created upon collapse survive and form bubble clusters during the next negative-pressure phase of the acoustic cycle (Fig. 4). For example, in bottom left panel, shock wave B1 is emitted by the cluster generated from the nuclei remaining after the collapse A4. Similarly, C1 is emitted by the cluster produced from the nuclei remaining after B1. By the end of the time scale (bottom left), this resulted in shock waves emitted during three consecutive acoustic cycles: A5, B2, and C1.

In addition, when the returning shock waves nucleate bubble clusters during the positive pressure phase of the driving acoustic field, the produced bubble cluster undergoes a “first collapse” that also emits a shock wave (video 4, frame 130.76 μ s). This first-collapse shock wave then returns 3.5 cycles later and nucleates another bubble cluster that grows and collapses, emitting another shock wave (asterisk, bottom right) in addition to those occurring at the four-cycle period.

Fig. 12. Typical PCD traces at 30 MPa (left) and 150 MPa (right) static pressures. Each panel shows a recorded PCD trace and a 0.5-20 MHz band-pass filtered signal to suppress the ~50 kHz sinusoidal signal of the driving acoustic wave. The bottom plots are the enlargements of the first several collapses showing periodicity of emitted shock waves that, at first, radiate only every fourth acoustic cycle (e.g., A1-A4, bottom left). B1 is a shock wave resulting from nuclei remaining from the cluster generating A4. C1 is a shock wave resulting from the nuclei remaining from the cluster emitting B2. The curved arrows (bottom right) show the 3.5-cycle roundtrip time of flight for the reflected shock waves.

The observed periodicity differs from the periodicity reported by Johnston et al [21], in which periodic shock emission was observed from acoustically driven cavitation clouds at normal atmospheric pressure. In that case, there were no reflected waves and the period grew with the driving amplitude because bubble clusters were too big to collapse every acoustic cycle. The periodicity in high-pressure spherical resonators, in contrast, is determined by the time of flight of the reflected shock waves. Furthermore, cavitation bubbles, when they exist, were observed to collapse under the positive pressure during every acoustic cycle.

Fig. 13. Peak-to-peak amplitude of shock waves produced by first bubbles (left) and bubble clusters (right) as a function of static pressure. Each point shows an average and standard deviation for at least 25 shock waves. Trend lines (dashed) show an apparent linear increase of SW-amplitude with static pressure; equations and coefficients of determination, R^2 , are shown bottom right.

Statistical analysis of PCD data shows an apparent linear growth of peak-to-peak amplitude with static pressure for both first bubbles and bubble clusters (Fig. 13). These results are in agreement with the linear growth predicted for first bubbles by our numerical simulations (Fig. 3). A similar linear dependence on pressure was observed in water over the range from 0.1 to 30 MPa static pressure [19].

Bubble clusters (Fig. 13, right) produced about 20 times stronger shock waves than spontaneously nucleated first bubbles (Fig. 13, left), indicating significantly more intense conditions inside the collapsing bubble clusters under these conditions.

4. Conclusions

High-quality-factor spherical resonators under extreme static pressure can reliably produce intense, violent cavitation, especially transient cavitation with clusters of bubbles. Experimental data at 20 MPa static pressure validates numerical simulations that show bubble temperatures on the order of 100,000 K and pressure at the center of the collapsing bubble on the order of 250 GPa. Furthermore, experiments and modeling at static pressures up to 150 MPa show that acoustic cavitation at increasing static pressures increases the resulting physical effects. In conclusion, this high-pressure cavitation system provides a controlled range of cavitation intensity and has a potential for broad applicability, encompassing any process requiring repetitive application of high energy density with rapid high-intensity heating and cooling rates. We expect that the extreme pressure cavitation reported here will enable new chemical and physical applications in both research and industry, helping to create new materials and novel technologies.

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