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Measuring cavitation and its cleaning effect

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ABSTRACT

The advantages and limitations of techniques for measuring the presence and amount of cavitation, and for quantifying the removal of contaminants, are provided. After reviewing chemical, physical, and biological studies, a universal cause for the cleaning effects of bubbles cannot yet be concluded. An “ideal sensor” with high spatial and temporal resolution is proposed. Such sensor could be used to investigate bubble jetting, shockwaves, streaming, and even chemical effects, by correlating cleaning processes with cavitation effects, generated by hydrodynamics, lasers or ultrasound.

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1. Introduction: cleaning with bubbles

Bubbles are well-known for their cleaning potential. As stated by Prosperetti, thousands of papers have been devoted to the subject of bubbles [1]. Bubbles can be generated ultrasonically, by laser, hydrodynamic effects, or other techniques. However, the exact cleaning mechanism induced by bubbles has not yet been elucidated, and the contribution of jets, shockwaves and other phenomena is still under discussion. The aim of this article is to discuss possible methods for identifying the cleaning mechanism. Here we give a non-exhaustive list of the techniques used for measuring the presence and amount of cavitation, and to quantify cleaning, and finally on studies that have correlated the two.

Measurements of the ultrasonic cavitation intensity are used as indicators of the effectiveness of an ultrasonic cleaning system. The intensity is often being related to the speed and thoroughness of cleaning, and the distribution is related to the uniformity of surface cleaning [2]. Section 2 discusses techniques and concepts to evaluate the effects of the violent and short lived emptiness that a collapsing bubble represents.

It is important to define what ‘clean’ means. ‘Clean’ can be defined as the absence of contaminants, which can be any undesired substance on an object. Contamination can come from dust, polishing paste, production waste material, bacteria, or even our own cells or hair. It can be said that an object is clean when the

amount of contaminant has been reduced to an acceptable or detectable level. The *acceptable* level of contaminants is different for every application, and for some industries and health sectors this has been regulated by ISO norms, e.g. 15883 for medical instruments. The *detectable* level of contamination depends on the measurement technique; and an overview of methods for measuring the cleanliness of a surface is given in Section 3.

A technical challenge in elucidating the cleaning mechanisms is to correlate the cavitation activity to the cleaning performance. Some attempts in this direction are covered in Section 4, and these studies have given some insight into the cleaning mechanisms involved in those situation. However, the ideal sensor or setup is not yet available. An ideal sensor would be able to determine the events of a bubble at relevant time scales, while simultaneously quantifying the cleaning that the bubbles perform, at relevant space and time scales. Our view on this is given in Section 5. Fig. 1 shows the outline of the article.

2. Measuring cavitation

Cavitation is defined as the formation of a void within a liquid, and its subsequent behaviour [3]. Since a void is the absence of fluid, it cannot be detected directly, but there are indirect methods available for measuring cavitation.

The first known attempt to study cavitation bubbles by their erosion potential was by Rayleigh in 1917 [4]. More recent reviews on cavitation, characterisation techniques and physical effects can be found in scientific literature [5–7]. Regarding the chemical

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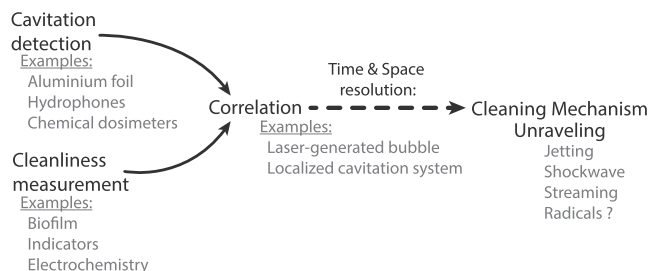


Fig. 1. General structure of the article. We begin with a discussion of the methods available for detecting cavitation bubbles and quantifying cleanliness. This is followed by an overview of previous attempts at correlating the two, in order to unravel the cleaning mechanism of bubbles. An ideal sensor will be required for this aim, which will be discussed at the end of the article.

effects of cavitation, there is equally an extensive body of literature around the generation of free radicals and biological effects [8–12]. Right from the start, researchers highlighted the difficulty in obtaining reproducible results.

A comprehensive review of different techniques to understand the cavitation activity distribution in chemical reactors, divided them in those that register primary effects and secondary effects [13], see Fig. 2. Primary effects include temperature pulse, pressure pulse, generation of free radicals inside the bubble, and micro-circulation in the vicinity of bubble at the collapse instant. Secondary effects involve quantification of chemical or physical effects after the collapse such as oxidation reactions, intensification of mass transfer coefficients, enhanced electrochemical effects, fluorescence, aluminium foil erosion, PIV, etc. Generation of free radicals is considered a primary effect, but measured mostly in chemical reactions after bubble collapse. The experimental information has often been complemented with theoretical modelling.

Exhaustive reviews on cavitation detection and measurement methods for high power ultrasonic fields have discussed their application in health care, sonochemistry and industrial ultrasonics [14,15]. The standardisation attempts of cavitation have been compared to that of standardising fire: its occurrence can be visualised and it can be controlled for practical uses, but only the

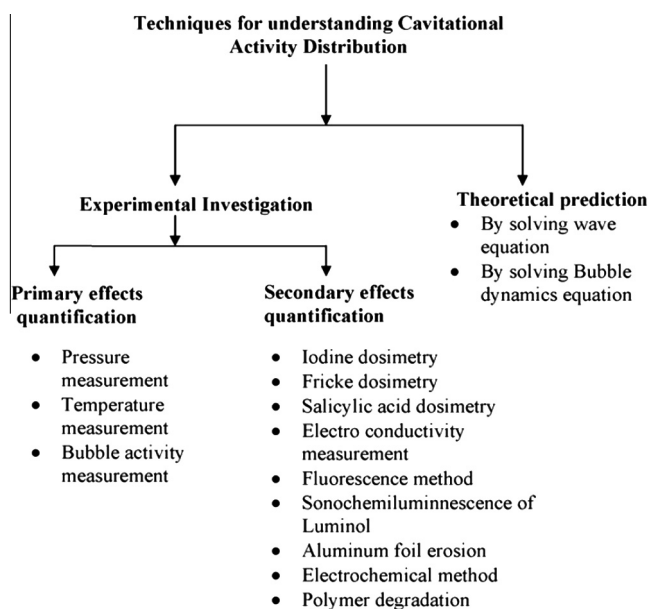


Fig. 2. Classification of different types of cavitation mapping techniques. The experimental techniques are divided into primary and secondary effects. Taken from [13].

time- and space-averaged phenomena of the flame (light emission, produced heat, oxidation, etc.) can be standardised. The same is true for cavitation.

2.1. Basic tests for detecting cavitation

Aluminium foils placed inside ultrasonic baths or close to ultrasonic horns are among the most basic tests to determine cavitation activity. A thin foil is eroded within minutes, and erosion patterns may be found corresponding to cavitation hot spots resulting in cleaning action concentrated in horizontal stripes (at pressure antinode regions). Bubbles can grow on the foil surface, translate and cluster. Streams of bubbles originate from certain regions on the surface and end in a bubble cloud “smoker” (or “streamers”), which has a strong erosive power [16].

It has been reported how streamers (multibubble structures) in standing waves are organised in planes parallel to the water surface. This resulted in cleaning action concentrated in horizontal stripes on the aluminium foil or painted glass surface [16]. This test is suitable for comparing the performance of an ultrasonic bath over time and obtaining a rough estimation of high pressure zones (hot spots), however the results are very sensitive to the placement of the foil, liquid temperature, dissolved gas and other variables. Ideally, this test would be standardised using materials with well-defined specifications, since the rigidity of the wall will affect the attraction of a bubble towards the boundary [17]. Automated analysis (e.g. image analysis or sample weighing) is another possible improvement [2].

The foil test is specified in the IEC/TR 60886 Technical Report, although it was concluded that there was no method suitable for standardisation. An alternative version of the foil test involves the erosion of pieces of lead under ultrasound exposure, or painted glass surfaces [2]. Another standard test uses carbon-coated ceramic rings, where the amount of removed carbon over time gives a measure of the mechanical cleaning activity inside an ultrasonic bath.

The distance from the bubble, or cluster of bubbles, to a wall as collapse occurs is defined as $\gamma = r/R_{max}$, where r is the distance from the wall and R_{max} the maximum radius attained by the bubble. This stand-off distance has a strong influence on the type of erosion effects. Fig. 3 shows how the laser-generated bubble collapses on aluminium foil leads to different erosion patterns for different values of γ [18]. The presence of defects in the surface, which can serve as nucleation sites, or simply pinning bubbles, have been observed to be accelerators of cleaning and erosion effects [19–21].

SonoCheck™ is a vial with a solution that changes colour within a few minutes due to ultrasound exposure, and can therefore be used as ultrasonic activity indicator. Its main ingredients are chloroform, buffer solution, and a pH-sensitive dye; its working principle appears to be based on ultrasound degradation of chloroform. Since the chloroform concentration is reduced by cavitation [22], the pH of the solution changes and therefore the colour of the vial solution changes. The SonoCheck™ also allows for monitoring the performance of an ultrasonic bath over time, but not to compare baths due to the underlying processes that depend on several factors, including ultrasound frequency and sample positioning.

2.2. Acoustic detection of cavitation

The onset of cavitation is characterised by an increase in the first subharmonic of the ultrasonic driving frequency. The origin of this phenomena lies in the onset of instabilities of large bubbles before they start to collapse [23]. Monitoring the subharmonic frequency component can therefore give an indication of the onset of

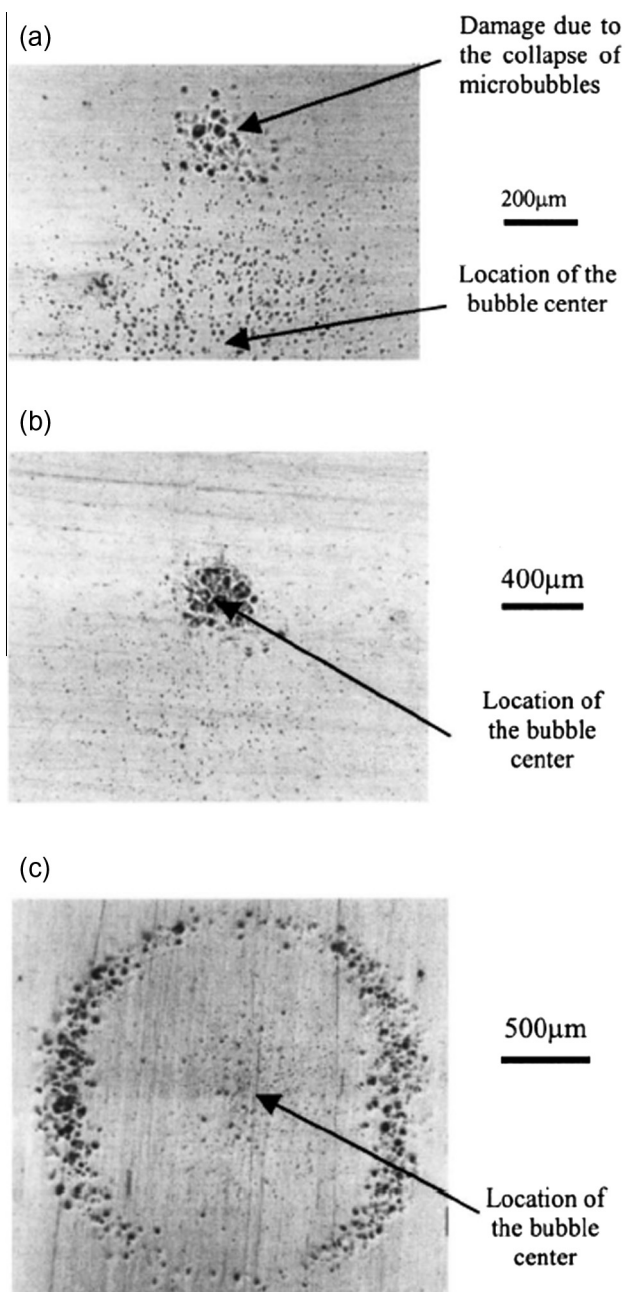


Fig. 3. Damage pattern observed on aluminium 1050A after creation of 600 bubbles for different values of γ . (a) $\gamma = 0.8$; $R_{max} = 1.3$ mm. (b) $\gamma = 1.1$; $R_{max} = 1.5$ mm. (c) $\gamma = 1.4$; $R_{max} = 1.4$ mm. Taken from [18].

cavitation, but does not give information on the amount or the precise location of cavitation.

Passive cavitation detectors are frequently used to detect the presence of cavitation in a liquid. Collapsing bubbles produce high-frequency acoustic signals (white noise), which can be captured using a suitable hydrophone [23]. The high-frequency component can be investigated using signal analysis. An increase in high-frequency signal compared to a reference measurement indicates the presence of cavitation. There are a couple of caveats to the acoustic measurement technique. The amplitude of the high-frequency component increases linearly with the number of bubbles only in the case of a small number of bubbles. High bubble densities lead to bubble clusters, in which the collapse of each bubble interferes with the others, also known as “shielding”. Such

interference induces a change in the acoustic emission, and the presence of bubbles may block the transmission of sound through the liquid to the hydrophone. Since the measuring principle is based on detecting the bubble collapse within the bulk liquid, near a surface, the acoustic signature may be different [24].

An array of acoustic detectors, together with the phase difference between received signals, has been used to reconstruct the location of a cavitation event inside an insonified volume of liquid. Using this technique, the authors made a map of the ultrasonic activity near an ultrasound transducer [25].

Several hand-held sensors have been developed to scan and map the cavitation activity inside an ultrasonic field, such as generated inside an ultrasonic bath. Some sensors are based on acoustic signal and other measure the pressure. The acoustic pressure does not directly give a measure of the amount of cavitation, but only predictions where cavitation is likely to occur.

There are several cavitation sensors available on the market, one being the CaviSensor and CaviMeter™ system. This system was developed by the National Physics Laboratory in the United Kingdom [26] and is now available at Onda Corp. The CaviSensor consists of a 30 mm diameter ring made of PVDF, which leaves the ultrasound field in an ultrasonic bath unattenuated but blocks frequencies in the low MHz range and up, coming from cavitation noise. Only the cavitation noise generated by bubbles inside the ring is detected, giving the sensor spatial resolution. The CaviMeter™ processes the signal and returns comparative values for the amount of ultrasonic driving signal, its subharmonic and cavitation noise.

Ultrawave's Hygea® Ultrasonic Activity Meter [27] is a hydrophone system that measures the frequency and acoustic pressure inside an ultrasonic bath, using a 15 mm diameter probe. It is an end-user targeted device that has a simple display for the ultrasonic frequency (5–50 kHz) and power (10–100%), making it suitable for comparative measurements over time.

Another pressure sensor is the Sonic Meter by SyncroCraft, marketed as Ultrasonic Cavitation Meter. A 8 mm probe measures the pressure in the frequency range 0–500 kHz and pressure range 0–900 kPa.

2.3. Optical

The small dimensions and fast dynamics of bubbles created by ultrasound do not allow confirmation of their presence by eye, although bubble clusters can often be seen in ultrasonic baths. To study bubble behaviour on more detail, high-speed imaging is required [28]. Frame rates on the order of 100,000 frames/s are typically required to accurately resolve the bubble dynamics. There have been studies using this technique to obtain valuable insight into cavitation, even using frame rates of several millions frames/s [29,30]. With the help of tracer particles (Particle Imaging/Tracking Velocimetry), the velocity distribution in the fluid around bubbles collapsing near a rigid wall have been resolved, for different standoff parameters [31]. A combination of shadowgraph techniques, short-pulsed lasers and high framing rates, allowed for the study of shock waves emission observed at minimum size of a laser-generated bubble [32,33].

The interaction of bubbles with nearby surfaces is another important phenomenon related to how bubbles can clean. With high-speed photography (200 million frames per second) and exposure times of 5 ns, and spatial resolution a few micrometers it was possible to record two types of shock wave emission. One was attributed to the free collapse of a bubble within the cloud by the ambient pressure in the fluid; the other was a result of the interaction of the cloud-collapse-induced shock wave with existing microbubbles close to the collapse site of the cloud. This secondary shock wave emission was reported to have amplitudes

around 0.5 GPa [34,35]. With similar experiments it was observed how bubbles adjacent to salt crystals break particles due to their rapid dynamics [36].

A simultaneous combination of laser-beam deflection probe (BDP) and shadow photography of laser-generated bubbles was proposed as an alternative to high-speed photography [37].

2.4. Chemical and physical methods

Several sonochemical methods have been developed to detect and quantify the effects of cavitation [38–40]. The chlorine release test is based on the decomposition of carbon tetrachloride dissolved in potassium chloride, that releases chlorine when exposed to ultrasonic cavitation. The chlorine in turn reacts with the potassium yielding iodine; its light absorption can be analysed spectroscopically as the concentration increases. This test was popular for some time, as it was reproducible under standardized conditions, but had the flaw that at higher frequencies (greater than 40 kHz) it still worked chemically, but no corresponding cleaning results were found [2].

Chemical tracers released by a bubble have been used to study the flow around it; particularly sulphur particles as a result of CS₂ decomposition [41].

The light emitted by collapsing bubbles (sonoluminescence) [42] can be measured using sensitive detectors such as photo multiplier tubes (PMT) and cameras. Single bubble sonoluminescence (SBSL) has no practical interest for ultrasonic cleaning, yet many studies have been done due to the easier conditions to observe and measure the effect of one bubble compared to a bubble cluster.

PMT's have the highest sensitivity and temporal resolution, however they give no spatial information. Extremely sensitive, cooled cameras have poor time resolution, but do offer spatial resolution. A method to map regions where collapsing bubbles exist is by using chemicals such as luminol, which react with the OH[•] radicals that form upon bubble collapse (sonochemiluminescence) [43–45]. In appropriate dark conditions, today's consumer photo cameras are sensitive enough to detect this light and give spatial information on the cavitation distribution in an ultrasonic reactor; the temporal resolution, however, is on the order of 10 s [46].

By studying the light emission from bubbles, it has been possible to gain insight into phenomena associated with cleaning. The luminescence of a laser-generated bubble collapsing near a single rigid boundary shows a smooth continuum spectrum. The luminescence spectrum of a bubble collapsing between two parallel rigid boundaries shows an emission band from the OH[•] radical at 310 nm; also observed in the multi bubble sonoluminescence (MBSL) spectra of water. The OH[•] band correlates to ultra high-velocity jets during the splitting of the bubble before the minimum-radius collapse point. The jet occurring for the case of a single wall is formed after the collapse point, when OH[•] is not present due to colder gas temperature. The spectral radiance of both scenarios was fitted with a blackbody curve at a temperature that decreases with the stand-off distance [47].

Other experiments comparing the light emission from normal and deuterated acetone, water, heavy water, and mixtures, at equivalent high operational static pressures yielded no difference in SBSL. Transient cavitation induced by neutron irradiation of the liquid, focused high intensity lasers, and spontaneous cavitation showed no significant difference in light or acoustic emissions [48].

An electrochemical approach to the detection of a sonochemical product (in this case hydrogen peroxide) together with the light output from multibubble sonoluminescence (MBSL) has been used to predict the conditions under which maximum sonochemical effect is expected [49].

Chemical dosimeters, such as terephthalic acid and luminol, have been reported not to be the ideal dosimeter for estimating sonochemical reactions, which depend on the mechanical effects of ultrasound [38].

2.5. Other cavitation measurement techniques

The cavitation meter currently being developed by Nanyang Technical University Singapore detects cavitation bubbles using a 1 mm diameter conductivity probe. Bubbles are recognisable as peaks in the conductivity signal, and can be analysed at rates up to 100 kHz.

The impact loads of bubbles collapsing against a piezo ceramic transducer have been measured using a hydrodynamic cavitation-liquid jet setup. The “simultaneous correlation” of impact load by collapsing cavitation bubbles and erosion damage was performed by volume loss. The authors assumed that the individual erosion indent was produced by a single pulse of cavitation bubble collapse, which is a debatable assumption [50].

Methods for calculating the impact energy and forces of a bubble or clusters of bubbles created with different techniques, and collapsing against a surface have been reported. An ultrasonic cavitation bubble cluster exerted a force of 20 μN on an AFM tip cantilever, which corresponds to pressures on the order of 50 atm [20]. Using a piezoelectric sensor, forces in the range 1–20 N were measured during hydrodynamic cavitation, and 1–10 N below an ultrasonic horn [51].

A passive p-type film of stainless steel can transform to an n-type semiconductor when exposed to cavitation [52]. This transformation of semiconducting behaviour of passive films can be useful for developing cavitation-resistant materials. The techniques used were polarisation curve determination, electrochemical impedance spectroscopy and capacitance potential measurement.

Magnetic resonance imaging (MRI) of a standing acoustic wave in water and surfactants solutions provide velocity spectra, kinetic energy maps, and the dispersion coefficient caused by cavitation bubbles, void fraction, as well as the influence of surfactants in the coalescence of bubbles. This method can measure various physicochemical parameters such as viscosity, velocity, diffusion, and molecular density, dynamics of dissolved gas. The advantages highlighted were spatial resolution by linear gradients of the magnetic field in three dimensions, non-invasive and capable of probing opto-acoustically opaque medium. On the downside, MRI can only provide statistically averaged information on the time scale of milliseconds and seconds, and in limited space resolution [53–55].

3. Measuring cleaning

3.1. Optical methods

In many industries and activities such as in jewellery, contact lens manufacturing, etc., cleaning is evaluated by visual inspection. Depending on the inspector's age and quality of sight, only contamination larger than 0.1 mm can be observed, and only on non-occluded surfaces. Magnifying glasses and microscopes provide increased optical resolution down to 0.5 μm, but at a reduced field of view. More details can be obtained using advanced microscopy techniques such as SEM, AFM and laser profilometry. However, these techniques require sampling of the surface that has been cleaned.

There are many different techniques available to assist in surface analysis, although they are mostly used in research laboratories. For example, surface material compositions can be detected

using X-ray photoelectron spectroscopy (XPS) [56] or hyperspectral imaging [57]. Surface reflection, glossiness, haze or colour can be analysed to detect differences related to contamination across the surface. In the cases where a transparent substrate is contaminated, the light transmittance change can also be used as a quantification method for cleaning. The contents of liquids (e.g. waste water) can be analysed using Total Organic Carbon (TOC) analysers, infrared spectroscopy (FT-NIR) or particle counters [58]. Waste water analysis using e.g. conductivity is often used to monitor the cleaning process, however it only determines the moment where no more contaminants are removed from an object. But this is not necessarily the point where the object is clean.

In many industrial cleaning situations, the cleanliness produced by cavitation bubbles cannot be seen, but verification is done by measuring process parameters including temperature, chemical concentration, conductivity, filter pressure drops or flow rate [2].

There are cleaning indicators available that are based on indirect effects: the effect of bubbles on an indicator material gives an indication of their presence and cleaning activity. Their application is to provide an easy and quick way for monitoring ultrasonic baths over time. These indicators do not provide insight into the cleaning mechanisms, nor on how many bubbles were present. They are also dependent on other factors like temperature and specific chemistries.

A step forward towards standardisation is provided by commercial ultrasonic cleaning indicators such as produced by *gke GmbH* and *Ultrawave*. They provide the cleaning indicators ready to be used, without user interference on applying the contaminant. The *gke* cleaning indicators are plastic strips on which an ink is printed in a reproducible method; four colours are available with increasing amount of binder added to the ink. The *Ultrawave* strips contain lipids and polysaccharides and a dye; the absence of colour indicates sufficient cleaning. For both commercial cleaning indicators, however, the analysis is still left to the subjective opinion of the user and is not quantitative.

The above cleaning indicators can be placed in a holder to mimic occluded surfaces of e.g. medical instruments. The test contaminants on these indicators are linked to ISO standards (15883-5 or HTM2030/HTM01-05) for cleanliness standards for medical instruments. However, a cleaned indicator does not mean that the actual objects are cleaned. It merely allows for a comparison of the cleaning efficiency compared to a previous validated instant, under the same conditions.

For the medical field, further validation of ultrasonic cleaning is provided by external companies that use a radionuclide method to measure the amount of contamination remaining in a medical instrument [59].

3.2. Biomaterial

In medical applications, cleaning often involves the removal of bacteria and cells that are grown on a surface. These biomaterials need to be both killed and removed from the surface in order to disinfect the object (e.g. a medical instrument or implant). Ultrasonic cleaning is often used to disrupt the cells and bacteria; in hospitals this is always followed by a sterilisation step such as steam disinfection.

The effect of cavitation on cells and bacteria has been evaluated by culturing a specific family of each into a container that is then exposed to cavitation generated by ultrasound, shockwave or laser. Among the first methods of investigating the mechanisms behind the biophysical effects of ultrasound, the “Harvey chamber” was used to perform qualitative observations [60]. The damage to human cells caused by the impact of the liquid jet developed during bubble collapse and the shock wave emitted during bubble rebound can be estimated by comparing the pressure values from

the jet and shock against the maximum stress the cell surface can withstand without breaking [61]. The damages on the microbial cells and degradation of polymer chain can be used as probes for obtaining values of the shear stress around the oscillating or collapsing bubbles. Particularly, the release rate of intercellular protein from yeast cells has been used to evaluate the physical (mechanical) effects of the ultrasonic field (see Fig. 4). The yeast cells are relatively rigid and may not be disrupted by the action of microstreaming. The rupture occurs when the yeast cells are in the vicinity of cavitation bubbles, resulting in protein release [62].

After the cleaning process, microscopy techniques in combination with dead/life staining give an indication of the cleaning/killing rate. Specific dyes are selected so that it is possible to check if dye molecules enter the cells after their membrane has been ruptured by cavitation. The uptake of calcein by cells reveals the cell membrane permeability; an increase in uptake indicates sonoporation. The calcein can be quantified using fluorescence techniques or flow cytometry. The latter technique has been used to correlate cellular bioeffects directly to the amount of acoustic cavitation [63].

A third method uses a reagent, MTS (*3-(4,5-dimethylthiazol-2-yl)-5-(3-carboxymethoxyphenyl)-2-(4-sulfophenyl)-2H-tetrazolium*), that is converted in metabolically active cells altering the light absorbance of the mixture. The absorbance value is directly proportional to the amount of viable cells after cavitation events; SEM images can provide confirmation of cell membrane rupture [64,65]. Recently, ultra high speed imaging of fluorescent dye uptake of cells being sonoporated by a bubble has provided new insight into the effects of stable bubbles on cells [66].

Another test, frequently used in industry, is to take a sample of a supposedly cleaned surface, e.g. using agar or a cotton swab, and place it in an incubator. The number of cells or bacteria (Colony Forming Units – CFU) grown after 24–48 h gives a representation of the cleanliness of the surface. A faster method is ATP bioluminescence, which allows for immediate analysis of the sampling swab [67]. Commercial devices are available for this test [68], which are often used in the food industry.

The disadvantage of taking a sample from a surface is that it is a single measurement at one location, and contamination may be present elsewhere. Logically the sample should be taken at the worst-case place in the surface or object. To find this place, the object of interest can be artificially contaminated, for example, with a fluorescent stain. The object goes through the standard washing procedure, after which the worst-case place still bearing stains can be detected.

Using living biomaterial such as cells and bacteria is a cumbersome task, since they have to be grown under controlled circumstances. Only with great care do researchers manage to make a reproducible contamination [69]. An additional problem is that bacteria grow into a biofilm, which is an extracellular polymer

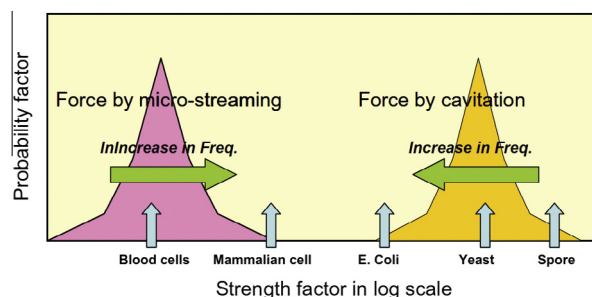


Fig. 4. Schematic representation of the relation between the forces in the cavitation field and which biomaterial is mostly affected by them. Taken from [62].

substance that gives a viscoelastic behaviour. This viscoelasticity is a mechanical and chemical defence adaptation, and strongly influences the cleaning mechanisms. It is difficult to have reproducible properties, since each biofilm can grow in a different way, and this makes biofilms difficult to include in cavitation research.

There are materials (phantoms) that mimic biological contamination and have been used for evaluating ultrasonic root canal cleaning [70,71]. The hydrogel introduced by Macedo et al. [71] displays viscoelastic behaviour similar to biofilms, and although its adherence to surfaces is weakly controlled, it allows for studying the interaction of cavitation bubbles with biofilm.

3.3. Semiconductor cleaning

The cleanliness of semiconductor wafers can be checked using automated particle counters, which detect particles down to a size of $300\ \mu\text{m}$ [72]. Another method for detecting (large) particles on wafers is by bonding a second wafer on top of the wafer of interest. Infrared imaging shows voids where particles are present on the wafer [73].

The semiconductor industry, which requires a very high level of cleaning to realise yet smaller features, has their own definition for numbers that assessing cleaning performance, such as Particle Removal Efficiency (PRE), Feature Damage Limit (FDL), and Feature Damage Probability (FPD); [74] which are relevant for megasonic frequencies.

4. Correlating cavitation and cleaning effects

The previous two sections have shown that there are many techniques available for measuring cavitation and for quantifying cleaning. Each method gives a different view on the phenomena during bubble collapse, and on how clean the surface actually is, but the methods don't conclude *how bubbles clean*. In this section we will highlight a few attempts in correlating the generation of cavitation bubbles with cleaning. At the root of this challenge lies the fact that there is no consensus of what is the ultimate reason why bubbles can clean. Since the effect can depend on the technique used to generate the bubble in the first place, is not easy to compare experiments done with ultrasound, laser-generated bubbles, or hydrodynamic cavitation, among others. Additionally, the spatial and temporal scales for bubble collapse and cleaning may be different, which complicates a direct comparison.

Laser-generated bubbles close to a surface have been reported to generate a shear boundary layer flow that is able to clean the surface from particles [75]. Micro-particle tracking velocimetry (μPIV) evidenced that forcing of particles is the strongest in a fraction of the bubble oscillation time. The force comes from a jet flow spreading radially and pulling away the particles. The study concluded that the cause for the cleaning effect is the induced jet flow. In an attempt to compare the laser-generated bubbles to ultrasonically generated ones, it was proposed that the same could happen for bubbles that grow to resonant size and jet towards the nearby wall, even though the equivalent frequency for the laser-generated bubble would correspond to few kilo-Hertz.

The cleaning mechanism may be different for acoustically or hydrodynamical generated cavitation bubbles. However, for those techniques, it is difficult to control cavitation in space and time for studying its cleaning mechanism. Generally, acoustic cavitation may be expected to occur at pressure antinodes, where cleaning by bubbles has been visualised (see Fig. 5) [76]. Recently, more controlled generation of acoustically generated cavitation bubbles has been achieved using micro-machined artificial nucleation sites that were driven ultrasonically [20]. A cloud of bubbles generated with this method was shown to remove various types of

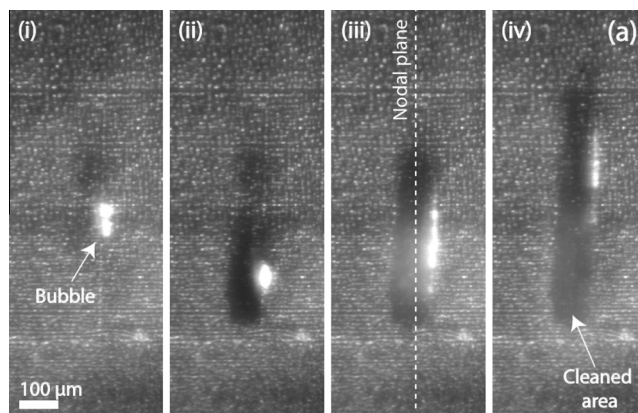


Fig. 5. Light scattering images showing the removal process of a layer of 100 nm gold particles from a glass substrate. The patchy grayish area is the particle layer. The dark regions are sections where the particles are removed. The bright spots originate from a bubble moving close to – or on the substrate. The vertical dashed line in frame (iii) marks the approximate intersection of a nodal plane with the substrate. Taken from [76].

contaminants from a nearby surface. The setup allowed for simultaneous visualisation of the generated bubbles and the cleaning process, but not to elucidate the cleaning mechanism. The time frame of cleaning was much slower (order of seconds) than the bubble oscillations (frequency of 200 kHz); see [Supplementary Supporting movies 1 and 2](#).

The combined study of high-speed imaging, sonoluminescence recordings, and surface cleaning tests helped identifying two distinct bubble populations in an ultrasonic bath [77]. Bubbles larger than linear resonance size group in planes parallel to the transducer surface at pressure minima spots. As bubbles rise they are followed by smaller satellite bubbles. The other bubbles below and near linear resonance size behave as “streamers” perpendicular to, and away from the transducer surface. Bigger bubbles did not sonoluminescence while the smaller bubbles in the streamers did when intersecting the planes of high driving pressure. Interestingly, both population of bubbles cleaned micro-particles attached to a glass substrate, and the mechanisms of particle removal, were suggested to be different: Bubbles sticking to the surface oscillate and jet against the surface, removing particles, and the streaming of the liquid carries them away. Streamer bubbles were observed to clean grooves and nearby to their trajectory as they moved. The authors concluded that the exact (microscopic) cleaning mechanism was impossible to elucidate. Slow recording and poor spatial resolution could not indicate if it was collapsing bubbles, jetting, or only effects of surface oscillations. The sonoluminescence measurements could only assure that volume oscillations were strong. In other studies, quantitative correlation between acoustic, physical and chemical parameters (sonochemical, sonoluminescence activity, and acoustical noise spectra) in the presence of small amounts of surfactants allowed to understand better influences in bubble population due to coalescence and other effects [78].

Other reports show comparisons of the broadband acoustic emission and aluminium foil erosion, assessing the spatial distribution of cavitation activity within ultrasonic cleaning vessels, as well as the inter-comparison of a large range of cavitation measurement methods. [79,80].

The broadband noise produced by inertial cavitation has been correlated with bio-effects acting as noninvasive feedback in real time [81]. The authors determined the kinetics of cavitation during sonication of Optison[®] contrast agent, and correlated that with cellular bio-effects predictable by cavitation dose. It was proposed

that this approach should correlate better with bioeffects than the ultrasound input parameters. Flow cytometry gave the fraction of cells containing intracellular calcein fluorescence, and cell viability loss with propidium iodide. A broadband piezo-polymer hydrophone was used as a passive cavitation detection system to monitor cavitation. The cavitation dose was calculated by summing the broadband noise on each frequency spectrum graph until the broadband noise decayed to a constant value [82]. The broadband noise is generated by both the initial rupture of stabilized Optison[®] as nucleation sites, and the destruction or dissolution of secondary bubbles generated over time. Additionally, bioeffects were correlated with sonoluminescence and free radical activity with invasive and complicated systems to measure cavitation inside the body [83]. Bioeffects generally increased with increasing pressure, increasing exposure time, increasing Optison[®] concentration, and decreasing frequency [63]. Unfortunately, the methods used in this study did not allow for temporal resolution of the cleaning.

Electrochemical methods have been used to study the erosion and corrosion activity at eight discrete frequencies within 20–150 kHz, where cleaning was reported to occur in similar ways as erosion. The electrode surface was damaged creating a corresponding electrochemical anodic current time transient. A spatial correlation was found between multi bubble sonoluminescence (MBSL) imaging, the acoustic pressure, and the erosion mapping at low frequencies (<50 kHz). For higher frequencies, the rate of the erosion activity decreased while high acoustic amplitudes and MBSL were measured. Fast imaging of a cluster of bubbles near the electrode surface presented dynamics correlated with detected erosion transients (see Fig. 6). A drawback of this method is that an erosive event may not always be detected; the magnitude of the effect can vary with time or bubble cluster location with respect to the electrode sensitive area [84].

The relationship between cavitation and its induced effects related to cleaning of large areas was studied with a multi-transducer device, designed and tested to clean the external hull of a boat [85]. The authors highlighted that the evaluation of washing efficiency was an important aspect of the dirt removal experiments.

5. The ideal cavitation-cleaning sensor

The studies described above illustrate why it is difficult to elucidate the cleaning mechanism of bubbles. Cleaning effects often take place at longer time scales than the bubble dynamics: similar to erosion, cleaning may have different periods of incubation [21]. Additionally, the area to be cleaned is typically much larger than the size of a single bubble, and there is a concerted effect of many individual bubbles. But simply measuring the amount of cavitation is not sufficient as an indicator for cleaning: the contaminant removal process has to be taken into account. Furthermore, the analysis can be flawed by subjective analysis of a human observer. Haptic techniques, e.g. an electronic finger, may provide a method to evaluate quantitatively and unbiased the result of cleaning of a surface, however such a sensor has not yet been reported in literature.

The challenge of designing, building and using an idealised sensor for cleaning effects given by cavitation is not a simple one; many of the difficulties and useful ideas on how to get this started have already been highlighted before [14]. The variety of different systems for generating cavitation as well as the various contaminants and cleaning methods make it difficult to compare the results from different studies.

A sensor that could fully correlate cleaning mechanisms and the bubble effects, to be used for ultrasonic cavitation, laser generated bubbles, hydrodynamic cavitation, etc. seems far from reachable in

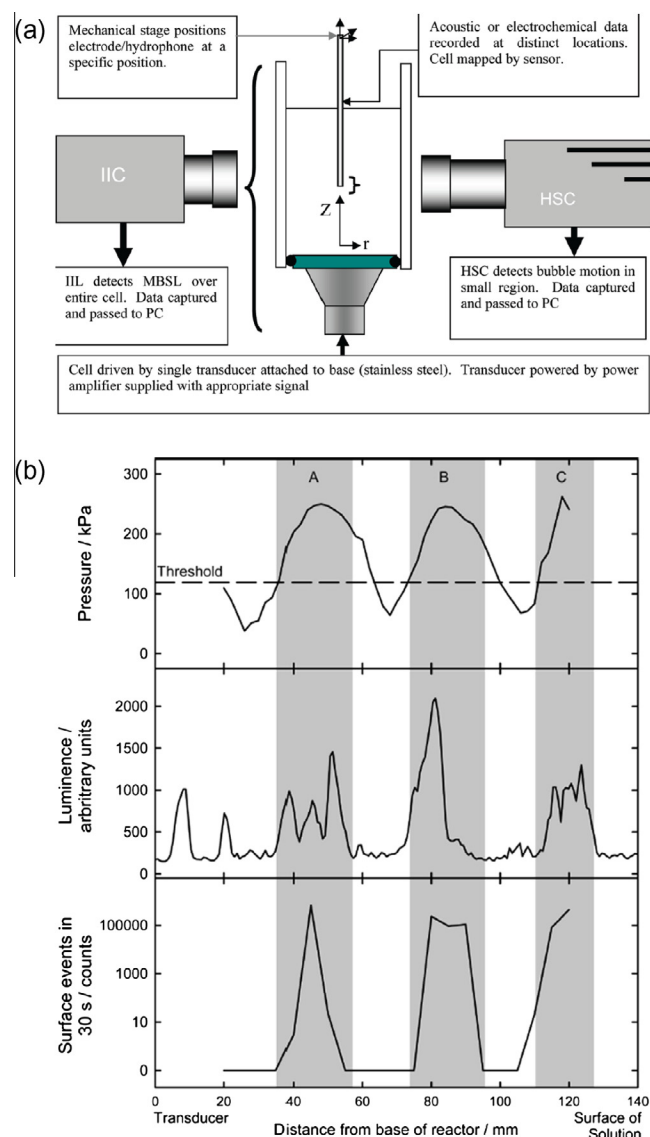


Fig. 6. (a) Schematic of the experimental setup used for erosion/corrosion, acoustic, imaging (both high-speed camera (HSC) and image-intensified camera (IIC)) of the cylindrical reactor. Note the HSC and the IIC are used under different lighting conditions. In addition the area imaged by each technique is different. The coordinate system (r - Z) is added for clarity. The open bracket shows the region addressed by the high-speed camera. (b) Plot comparing the central pressure scan and luminescence trace from reactor 4 driven at 23 kHz as a function of distance with three bands of increased activity identified (A, B and C). Taken from [84].

the near future. In practise, a combination of measurement techniques will be required, such as illustrated in Fig. 6, or reported elsewhere using cameras operating at different frame rates [66].

Ideally, such a sensor would be non-intrusive, and able to visualise the bubble dynamics and measure the physical-chemical effect of each individual bubble on a relevant contamination layer. Simultaneously, the contamination removal process should be quantified automatically at the time scale of the bubble dynamics, as well as the time scale relevant in the application. The sensing principle should be insensitive to any phenomena that may influence the bubble formation or contaminant removal, such as temperature or gas concentration changes. We have sketched such an ideal sensor in Fig. 7.

Besides the measurement technique, it is also important to look at the conditions under which bubbles are generated [2]. As noted before, bubbles generated acoustically can have a different

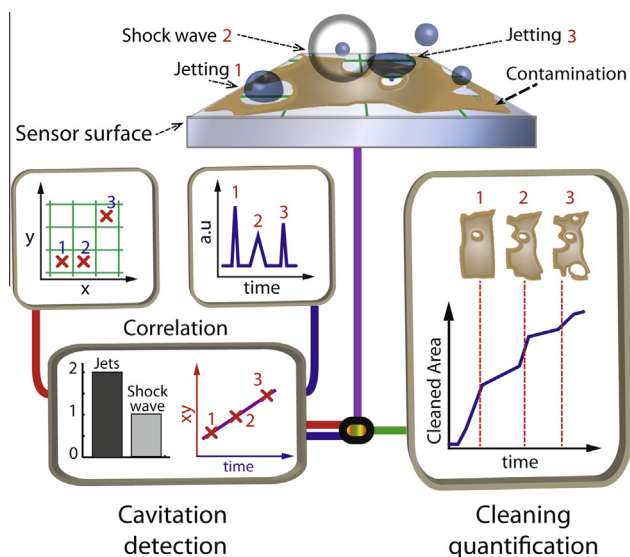


Fig. 7. Ideal sensor for the simultaneous detection of cavitation and cleaning quantification. We illustrate, on the lower-left side of the sketch how each cavitation event could be recorded, in space and time. The magnitudes in terms of intensity and duration could be correlated to identify if the event came from a jet or a shockwave. At the lower-right side, the quantification of cleaning can be directly associated to the cavitation events.

cleaning mechanism than those generated hydrodynamically or by laser. And even within one bubble generation technique, the control parameters can affect the results. In ultrasonic systems, the bubble dynamics are dependent on ultrasound parameters (frequency, power, sweep, etc.) and the properties of the liquid (fluidic properties, temperature, gas content, nuclei, etc.). For example, there is a frequency dependence on the effects exerted by cavitation bubbles, and the presence of an object in an ultrasonic bath affects the ultrasound field [2]. Mechanical effects dominate at low ultrasonic frequencies, whereas the contribution of chemical effects start to increase for frequencies above 100 kHz [86]. As mentioned before, chemical dosimeters, might not be the option for estimating sonochemical reactions that depend on the mechanical effects of ultrasound.

Water or liquids used in ultrasonic cleaning are almost impossible to use in its purest form; there are always contaminants that can behave as surfactant, i.e. lowering the surface tension [6,87,88]. This influences the measurement techniques discussed so far, e.g. the presence of additives such as particles or salt crystals were promote cavitation effects [13]. The dynamics of cavitation, such as bubble coalescence, clustering, and fragmentation, depend as well on physico-chemical aspects [89]. For example, the force transfer to a particle separated from a surface by a liquid film increases when decreasing the viscosity of the liquid. Additionally, the addition of organic solutions such as ethanol and acetone in water will lower the viscosity of the liquid, hence removing contaminants better [90].

Using “commercial” ultrasound test equipment for more scientific and refined or complex studies can lead to wrong conclusions. The overwhelming amount of experimental results found in literature cannot be readily summarised or compared due to the many differences in hardware, protocols, objectives of the studies. To overcome this limitation, the scientific community working in the fields mentioned in this article should reach a consensus on what type of solutions, physicochemical properties and parameters, materials and other relevant variables should be the most important for the detection of cavitation and cleaning effects.

6. Conclusions

This article has reviewed several techniques available for measuring the presence and amount of cavitation, and for the quantification of cleaning. Listing the advantages and limitations of such techniques we have found that attempts at correlating the cavitation and cleaning effects are not conclusive yet. In other words: clarifying a universal cause for the cleaning effects of bubbles needs further scientific research and poses several technical challenges.

We have shared our vision of an “ideal sensor” that is able to register all bubble events while quantifying contaminant removal, at the relevant spatial and temporal scales. Such a sensor is currently not available, but the increasing demands from industry and society for “cleaner objects” will require developments in this direction that will improve our understanding of the cleaning mechanisms of bubbles.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.ultsonch.2015.03.009>.

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