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Review

A parametric review of sonochemistry: control and augmentation of sonochemical activity in aqueous solutions

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A parametric review of sonochemistry: control and augmentation of sonochemical activity in aqueous solutions

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Abstract

In this review the phenomenon of ultrasonic cavitation and associated sonochemistry is presented through system parameters. Primary parameters are defined and considered, namely; pressure amplitude, frequency and reactor design; including transducer type, signal type, vessel-transducer ratio, liquid flow, liquid height, liquid temperature and the presence of a reflective plate. Secondary parameters are similarly characterised and involve the use of gas and liquid additives to influence the chemical and physical environments. Each of the parameters are considered in terms of their effect on bubble characteristics and subsequent impact on sonochemical activity. Evidence suggests that via parametric variation, the reaction products and efficiency may be controlled. This is hypothesised to occur through manipulation of the structural stability of the bubble.

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1. Introduction

Ultrasound, a sound field with frequency greater than ~20 kHz, has a plethora of applications in several actively researched areas. These include, but are not limited to, structural testing of materials [1, 2], underwater ranging and transmissions [3, 4], imaging and acoustic microscopy [5, 6], medical treatment [7-9], materials processing [10-13], cleaning [14] and more recently, wireless communications [15]. When ultrasound is incident upon a liquid it can facilitate the breaking of chemical bonds via the process of sonolysis, resulting in the formation of free radicals. The mechanisms of sonolysis include nucleation, cavitation, bubble dynamics / interactions, thermodynamic and chemical processes [16-18]. This process is useful for significantly increasing stoichiometric and catalytic chemical reactions [19], waste water treatment including degradation of organic pollutants [20], destruction of pharmaceutical waste [21], synthesis of nanomaterials and biomaterials [22] and potentially represents a method of production of fuels such as ethane, ethylene and acetylene [23].

However, ultrasound has an inherent problem of achieving an efficient process. This is due to inefficient energy transfer via impedance and secondary effects such as streaming, sound field attenuation, heating, bulk mixing, emitter erosion and sound emission [24]. The purpose of this review is to discuss the parameters that influence sonochemical reactions and consider how they may be implemented to achieve systematic optimisation. Particular attention is given to the effects of chemicals, changes in gas atmospheres and parameters that influence bubble surface instabilities.

2. Theoretical Background

Upon the application of ultrasound to a liquid, cavities are formed via nucleation that originate from pre-existing bubbles in the solution, stabilised against dissolution due to surface properties [25]. Nucleation sites may be found throughout the solution or at crevices on solid impurities (motes) or surfaces [25]. Following nucleation are cycles of oscillation; the sound field causing the liquid to experience cycles of high or positive pressure (compression) and low or negative pressure (rarefaction). The bubbles are also subject to these changes; the high pressure forcing the bubble to contract, the low pressure forcing the bubble to expand [25]. Since mass transport in and out of the bubble is dependent on the surface area, more 'material' will diffuse into the bubble than will diffuse out during the compression and rarefaction cycles. This is due to an increase in surface area during a long bubble expansion time compared to a decrease in surface area during rapid compression, hence overall cyclic growth occurs. [26, 27]. Since the bubble reaches a relatively large radius on expansion the collapse upon pressure change occurs very quickly and violently [28]. The diffusion rate is proportional to the gradient of concentration of dissolved gas localised around the bubble, described by Crum as a 'shell' effect [27]. On expansion, a liquid shell surrounding the bubble may be envisaged that becomes thinner and gas will be concentrated in this region. At the same time, inside the bubble the concentration decreases due to expansion. This change in concentration gradient causes the rate of diffusion from the shell into the bubble to occur more readily. On contraction, the shell becomes thicker and less concentrated, whilst the concentration in the bubble increases, therefore the diffusion does not readily occur. Hence the pressure induced oscillation cycles cause an overall growth in bubble size, generally known as growth by rectified diffusion.

If bubbles are below a critical size they are known to dissolve due to Laplace pressure in the bubble and as a consequence of surface tension [27]. However, if above critical size they resist dissolution and the size of the bubbles determines their position within the sound field. Small bubbles can move towards the pressure antinode via the primary Bjerknes force where they may grow to active (chemically producing) size through diffusion or coalescence [29-31]. If growth causes the bubbles to

be of a size greater than the resonance size (a size at which they expand dramatically and are able to repeatedly oscillate with pressure change) they will oscillate out of phase, move towards the nodes and become inactive due to the reduction in pressure variance [30]. These inactive bubbles are often referred to as degas bubbles and can oscillate at their natural frequency, without fragmentation [32]. Degas bubbles move to the surface of the liquid via buoyancy and the radiation force resulting in a reduction of gas in the fluid [25].

It should also be noted the role of the secondary Bjerknes force in the coalescence of neighbouring bubbles; bubbles of differing size will repel due to out of phase oscillation, bubbles of the same size will attract due to in-phase oscillation [25, 33, 34]. Kirkpatrick and Lockett studied the relationship between approach velocity and coalescence in the absence of ultrasound and considered that there are two competing effects [35]. An increased pressing between two bubbles causes film drainage (thinning) aiding coalescence, but an associated increase in the rate of contact area was found to circumvent the thinning effect. For higher approach velocities the rupture threshold may not be reached and the strain energy can reverse the bubble motion (rebound) and the film begins to thicken. At low approach velocities the rate of increase in contact area is slow enough to allow drainage and rupture [36]. In general, in the presence of ultrasound, an increase in frequency (at high frequencies) and the acoustic pressure increases the secondary Bjerknes force and the approach velocity [37]. Therefore at some point rebound will be more favourable as opposed to coalescence. However, coalescence is also influenced by the applied ultrasonic frequency that determines the time period available for it to occur [38, 39]. If the pressure amplitude has sufficient intensity and the sound field nears the resonance frequency, bubbles of similar size have been observed not to coalesce or rebound, remaining in contact and side by side [34]. In cases where there are surface-active materials in the liquid the thinning of the bubble wall may be controlled by viscous forces and not inertial forces as previously described. This is due to reduction in the mobility of the bubble surface and a surface concentration gradient that exists as two bubbles coalesce [40, 41]. Coalescence, in addition to rectified diffusion, represents another means of bubble growth and thus at high frequencies can be beneficial in producing bubbles of active size [42]. Rectified diffusion and coalescence may occur in approximately equal proportions and are important to consider when evaluating overall parametric effects in an ultrasonic system [43].

Two types of cavitation are generally considered; stable (or non-inertial) cavitation bubbles last over many cycles and may be considered weakly and symmetrically oscillating [43-45]. Transient (or inertial) cavitation bubbles grow extensively over a few cycles until the energy from the ultrasound can no longer be absorbed, then the forces acting on the bubble dominate causing instability and violent collapse. This in turn can lead to fragmentation into smaller bubbles that may repeat the process or dissolve [32, 46-50]. Accordingly, the concept has been visualised as 'dancing bubbles' as, upon fragmentation, a larger bubble ejects tiny bubbles and can be seen to dance by counteraction [51, 52]. Neppiras presented calculations for the threshold for the emission of these bubbles (often termed microbubbles) that were four times the threshold for bubble surface instabilities [53]. This would indicate that transient bubbles must become very unstable at their surface in order for fragmentation to occur [50].

Surface instabilities have been studied by several authors who theorise that they perpetuate in defined modes of vibration [54-58]. Further, the surface instabilities of a bubble have been said to be strongly coupled to bubble oscillations and translational motion [53, 59, 60]. In this regard they are often referred to as surface oscillations or surface waves [53, 61]. In this review it is acknowledged that surface oscillations at specific modes of excitation do occur, but for consistency the term surface instabilities is used to encapsulate the phenomena. Parameters which induce changes in bubble

oscillation may have a direct impact on fragmentation via surface instabilities. This is because there is a clear association between the degree of translational motion (caused by surface instabilities) and fragmentation [50]. It is generally accepted that fragmentation is the reason transient cavities are the ones that contribute significantly to sonochemical effects [32, 53, 62, 63]. This is because the smaller bubbles, upon dissolution, give opportunity for more disassociated gas products to interact in the liquid and at the bubble surface. The two main types of cavitation have been characterised by Leighton as fragmentary transient cavitation (now more commonly referred to as transient cavitation) and repetitive transient cavitation (high-energy stable cavitation) [25]. The former allows for new bubbles to be produced via fragmentation while the latter can remain stable and oscillate for hundreds of oscillation periods [44, 64]. With regards to the term 'transient', it is important to differentiate between two phenomena; an initial response to the applied ultrasound in which the bubble experiences a sudden change of state (to non-steady) which can result in large volume pulsations, or in the instance of the recent discussion, cavities that have a finite lifetime [25, 65].

The bubble size during cavitation is generally dependent on the pressure amplitude and frequency. However, to give perspective, during sonoluminescence several authors comment upon an equilibrium radius, $R_0 \sim 4 - 5 \mu\text{m}$, growth to a maximum radius of $10R_0 \sim 40 - 50 \mu\text{m}$ and collapse to a minimum radius of $R_0/10 \sim 0.4 - 0.5 \mu\text{m}$ [66-68]. The extremely rapid collapse precipitates almost adiabatic heating of the bubble contents due to negligible (if any) heat transfer to the bulk solution [66]. Since the surrounding liquid is cool the heat is highly localised to a 'hot-spot' that can reach temperatures and pressures of around 5000 K and 1000 bar respectively [19, 69]. The speed of collapse is apparent, observed at speeds above $\sim 1360 \text{ m/s}$ for a single sonoluminescing bubble [70]. Estimates for a multibubble system based on a variety of techniques place intra-bubble temperatures between 750 and 6000 K [43, 71-76]. The bubble temperature has been considered as a time and volume averaged temperature responsible for chemical reactions and a peak temperature associated with sonoluminescence [77]. However experimental evidence has also shown that an inverse relationship exists between the thermal conductivity of the gas and the measured sonoluminescence, which should not be apparent in an adiabatic process [78]. Gompf *et al.* observed that the sonoluminescence pulse width was identical in the red and UV part of the spectrum [79]. This would be inconsistent with an adiabatic compression model where the pulse width would be much larger in the red than the UV part of the spectrum.

Recent spectroscopic studies have shown a more complex situation with non-equilibrium plasma formation in the form of excited $\bullet\text{OH}$ radicals in different energy states [80]. In this instance the electron temperature (T_e) is much greater than the vibrational (T_v), rotational (T_r) and translational (T_g) temperatures in the order $T_e > T_v > T_r \approx T_g$ [81]. This moves away from an adiabatic model and indicates that, rather than considering the effects of temperature inside the bubble from the viewpoint of a single value, there is in fact a multi-temperature environment governed by differing particle energies and degrees of freedom. It was found that gas temperatures inside the bubble are much lower than electron temperatures and do not account for observed emissions. Lower ionisation potentials of the gas provide an increase in electron temperature concurrent with obtained spectra which supports formation of a non-equilibrium plasma [82].

A higher bubble temperature does not necessitate an increase in sonochemical activity; the experimentally determined temperature of the highly chemically active bubble surface region is lower than that of the interior of the bubble having a value of around 1900 K [73]. It should be noted that this temperature was determined using n-alkane solutions of metal carbonyls; reaction rates were determined and comparative rate thermometry used in approximating the temperatures involved. In another study that used ultrasound to degrade aqueous p-nitrophenol solutions the interface region

was the location of high temperature carbon nitrogen bond cleavage and radical production [83]. The effective temperatures for this process were suggested to be near to 900 K. Sufficiently high temperatures can destroy chemically active components within the bubble. This is concurrent with temperature dependant determination of an ionisation or disassociation process, which in turn can govern the number of reactive species produced [17, 25, 82, 84, 85]. For a multibubble sonoluminescence system the pressure inside the bubbles has been estimated to be in the order of 300 bar in argon saturated silicone oil, consistent with adiabatic compression [86]. For high energy stable cavitation a maximum pressure of 100 bar has been given with transient cavitation suggested to result in much higher pressures from 1000 to 70000 bar dependent upon applied pressure amplitude and frequency [87]. The high local temperatures and pressures achieved are said to provide effective means of driving chemical reactions under extreme conditions [16].

Disassociated products within the bubble, identified via spin trapping methods [88, 89], are radicals that interact and initiate chemical reactions. The majority of radicals produced within a repetitive transient bubble are confined (without fragmentation) to the bubble interior by the bubble surface [62]. However, some direct diffusion of highly soluble products from the bubble can occur [52]. The maximum lifetime of a bubble in a multibubble environment has been found to range from 0.35 ± 0.05 ms in a 213 kHz field and 0.10 ± 0.05 ms in a 1062 kHz field [90]. If the lifetime of active bubbles is shorter than the lifetime of the radicals produced, then the radicals are released into the liquid (via fragmentation or dissolution) where they may trigger chemical reactions [91-93]. If the bubble lifetime is longer than the radicals' lifetimes, then they will mainly recombine and there will be fewer reactions occurring within the solution [94]. This introduces frequency dependence for reaction pathways (discussed further in Section 3.2.). If the solution is water, a redox reaction occurs where $\bullet\text{H}$ and $\bullet\text{OH}$ radicals are produced inside the compressed bubble due to disassociation of the H_2O vapour [32, 95]. These radicals may then recombine and form H_2O_2 , while $\bullet\text{H}$ can react with oxygen to form $\text{HO}_2^\bullet$ [96, 97]. In fact, Yasui *et al.* for an air bubble consider 93 chemical oxidation and reduction reactions that involve the initiating species N_2 , O_2 and H_2O [98]. Of these, Gong *et al.* have summarised seven primary reaction mechanisms (1) - (7) [96].

The reactions are often described as an advanced oxidation process (AOP) based on a method of accelerated oxidation due to generation of oxidative species, particularly $\bullet\text{OH}$ and $\text{HO}_2^\bullet$ and H_2O_2 as a secondary product of radical combination [99]. The number of radicals formed via oxidation is often referred to as the sonochemical yield, though dosimetric measurements are generally based on the measurement of the hydroxyl radical formation [100].

Although there are several studies, there is general uncertainty concerning the prevalence of these reactions and some question (dependant on the individual sonochemical system) if some occur at all.

This could be due to the bubbles size and the time permitted for reactions to occur inside and outside the bubble i.e. a frequency based oscillation period determination. It may also be a question of temperatures achieved and reaction location. For a bubble with high water vapour content the main oxidant is $\bullet\text{OH}$, however in bubbles with less water content, temperature dependence becomes apparent. When the bubble temperature is in the range of 4000 – 6500 K, H_2O_2 is the dominant product, whilst above ~6500K it is the O atom [101]. There are three reaction sites in sonochemical experiments; the gas phase region inside the bubble (pyrolysis reactions), the bubble surface (reactions occurring in pressure temperature gradients) and the bulk solution [100, 102-104]. Mason suggested a fourth site originating from a two site model that considers a liquid phase surrounding the hot bubble, that can either be a heated shell of liquid around the collapsing bubble or liquid droplets injected into the bubble by instabilities at the surface [100]. Of these sites the interfacial region is the greatest area of $\bullet\text{OH}$ population and recombination [20, 30, 105-110]. Merouani *et al.* through numerical simulation determined that of the numerous products, H_2 occurred most abundantly and more readily in the gas phase via reaction (8) [111].

Indeed, all sites to varying extents may play some role, particularly in terms of vapour pressure; high vapour pressure solutes will more readily enter the gas phase whilst those with lower values may react at the bubble surface or in the bulk solution [112].

Scavenging of radicals can also alter reaction pathways, for example, if $\bullet\text{OH}$ is scavenged then the formation of H_2O_2 will decrease [18]. This may occur particularly in the presence of hydrophobic solutes which tend to the bubble surface and scavenge $\bullet\text{OH}$ radicals [113]. Sonication in gas atmospheres, such as oxygen, can cause radical scavenging with O_2 reacting with $\bullet\text{H}$ to produce $\text{HO}_2^\bullet$ [23]. Gas atmospheres are further considered in this regard in Section 4.3.

The population of bubble types (transient or stable cavitation) is dependent upon the parameters that influence bubble surface instability. Therefore by controlling this phenomenon it is possible to control the sonochemical activity taking place. Several authors indirectly or directly have altered sonochemical effects through bubble surface instabilities, although it was not the primary focus of their work [51, 69, 114-117]. Bubble surface instabilities may be seen as unwanted due to asymmetric collapse which can reduce the temperatures and pressures achieved, however the required pressures and temperatures for disassociation can still be readily achievable in a transient cavitation field [69]. In addition, sonoluminescence which increases with increased bubble stability and higher interior bubble temperatures is still observed when shape instabilities are apparent [66, 118-120]. Since unstable bubbles are capable of instigating sonochemical reactions and offer a possible mechanism to define sonochemical activity, the mechanisms of bubble surface instabilities and their effect on sonochemistry is considered in the continuation of this review.

3. Primary Parameters

In this section three fundamental parameters are defined: pressure amplitude, frequency and reactor design. The term ‘primary’ may be attributed to these parameters as they provide a base to establish the working design of a sonochemical system.

3.1. Pressure Amplitude

In general an increase in pressure amplitude will relate directly to the power transferred to the liquid and, up to a point, increase the number of cavitation bubbles, the temperature of collapse and the

sonochemical yield [16, 95, 121]. There are upper and lower limits of pressure amplitude to the cavitation threshold [16]. Above the upper limit, coalescence and degassing occurs with limited active cavitation. Below the lower limit the amplitude of the sound field is too small to induce nucleation or bubble growth; bubbles more readily succumb to surface tension effects and dissolve, again resulting in limited active cavitation [122]. Between these limits, pressure amplitude impacts on both the magnitude of the collapse, bubble oscillation and bubble surface stability. Higher pressure amplitudes result in bubbles of larger size and the compression and rarefaction force increases. The collapse is more violent and can lead to increased temperatures and associated increase in disassociation or ionisation of the gas [122, 123]. The non-linear bubble and bubble surface oscillations related to high pressure amplitudes can cause bubbles to fragment producing new bubbles that can contribute to sonochemical activity [57, 124-128].

An increase in pressure amplitude effects bubble agglomeration, with an increase in field strength and an associated 'lock-in-place' effect on bubbles at the antinodes [122]. Therefore shrouds of bubbles may form at antinode regions or at the radiating surface that diminishes the penetration of the ultrasound causing an effective reduction in pressure amplitude and energy transfer efficiency [16]. Additionally, nonlinear oscillation increases with amplitude and these bubbles can form adjacent stably positioned pairs. Thus, rather than coalescing they cluster and accumulate in groups [129-132]. At higher pressures, the primary and secondary Bjerknes' forces increase and bubbles experience an increased driving force towards one another [133]. Bubble coalescence can also increase with pressure amplitude because of a rise in approach velocity [134]. This is only true up to the point where the rate of film drainage between bubbles in contact starts to decrease, upon which there is rise in critical thickness for rupture to take place and rebound is more likely to occur [35]. A further consideration is that an increase in pressure amplitude causes increased propagation of bubble-bubble oscillation and consequently the bubbles influence each other at increased distances [134].

Bubble motion, agglomeration and coalescence are also affected by quasi acoustic and acoustic streaming, influenced by pressure magnitude [128, 135]. Quasi acoustic streaming is caused by attenuation of the sound field, often influenced by the presence of bubbles causing the formation of an energy gradient in the direction of the fields propagation [135]. The radiation pressure as a result causes motion of the bubbles and an associated macroscopic flow (due to bubble motion and viscosity of the solution) in that direction [136-138]. Aside from influencing bubble dynamics, quasi acoustic streaming also reduces the degree of reflection of the sound field and distorts the standing wave component [117]. Acoustic streaming (also known as microstreaming) occurs near the individual bubbles and is due to power dissipation of the acoustic energy with influence from mass transfer and bubble surface instabilities [135, 139-142]. Any bubble deformation tending away from asymmetry causes translational motion, thus, where surface instabilities occur, acoustic streaming will be present [57, 143, 144]. In the absence of bubbles acoustic streaming can still occur due to the viscosity of the fluid, this is then further influenced by any fluid motion or acoustic nonlinearity [145]. Bubble surface motion in the vicinity of a boundary or hard surface causes the formation of microjets [146]. Microjets are high speed liquid jets that propagate from the proximal side of the bubble towards the nearby surface [147]. They are produced due to asymmetry of flow between the bubble and the hard surface which causes bubble surface instabilities and non-spherical collapse [16, 148].

In terms of sonochemical reactions the pressure amplitude has been shown to be influential with higher powers delivering an increase in the number of active cavitation bubbles [121]. However, although there is an increase in sonochemical yield at higher powers, there is a limit for excessive powers where there is a decrease in the number of active bubbles per unit volume [149]. This can be attributed an increase in bubble coalescence and degassing at higher powers. Rooze *et al.* took the

oxidation efficiency of potassium iodide to be the amount of I_3^- produced divided by the ultrasonic power [150]. Using low powers (1 - 6 W) they showed that at low frequencies (20, 41 and 61 kHz) the efficiency increased as a function of acoustic pressure. Henglein *et al.* considered higher powers at 800 kHz and found sonochemical yields increased up to a maximum at around 100 W and then decreased with further power increase [117]. The group also measured a decrease in the dissolved gas content as a function of pressure amplitude up to around 100 W. Above 100 W there was an increase in gas content due to agitation of the liquid, attributed to (quasi) acoustic streaming at high frequencies which was furthered by the use of higher acoustic pressures [117]. Therefore, reduction in sonochemical yield at excessively high acoustic pressures may be attributed not only to coalescence and degassing but also to the effects of increasing liquid agitation.

High pressure amplitudes increase the number of cavities available and these will experience high levels of structural instability due to increase in the force of collapse. Concurrently, due to bubble oscillation and surface instabilities, associated acoustic streaming can occur. The resulting translational and cavitational motion is further acted upon by the energy gradient of the sound field and facilitates quasi acoustic streaming which is increased with pressure amplitude. This can cause motion of the bubbles that reduces coalescence, increases bubble instabilities and augments rectified diffusion of species into the bubble. However, at very high powers, coalescence may still readily occur due to agglomeration in antinode regions with increased strength stability of the field. This agglomeration can attenuate the sound field and reduce its effectiveness. An increase in bubble size and the secondary Bjerknes force with pressure increase may also contribute to further coalescence. In summary, the effect of increasing pressure amplitude is to force bubbles into a structured positional and oscillatory regime which up to a point presents a mechanism for increased sonochemical activity.

3.2. Frequency

The frequency of the sound field influences the bubble population with respect to the number of bubbles, bubble size, distribution of bubbles and the amount of chemical activity. All these things are dependent on the reactor geometry, ambient pressure, and temperature, viscosity and gas composition of the liquid [42]. In general, higher frequencies will result in increased nucleation and production of bubbles that are comparatively small in size [106, 151-153]. This is because the resonance size of a bubble is inversely related to the applied frequency [25]. As frequency increases, the amount of vapour entering the bubble decreases due to the reduction in its expansion phase [32]. This increased rate of oscillation and reduction in growth per cycle allows small bubbles in steps to reach resonance size more efficiently than lower frequencies (under 100 kHz) where the bubbles have relatively long growth periods [42, 69]. It should be considered however that the cavitation threshold is increased at higher frequencies and as such higher powers are required to induce cavitation [154, 155].

A sound field consists of both standing and travelling wave components, with optimisation of sonochemical yields observed in the presence of travelling waves [42, 44, 156]. These waves are often referred to as a (quasi) acoustic streaming phenomenon with flow associated with the travelling part of the sound field [145, 157]. The ratio between standing and travelling waves changes as a function of frequency with travelling waves dominating at high frequencies [44, 156]. The frequency dependence of the nature of the bubbles, transient or repetitive transient, is difficult to define as it is reliant not only on the frequency but also the power and type of transducer (and transducer configuration) used [47]. This is discussed further in Section 3.3.1. Since sonochemical yields have been seen to increase during transient cavitation, the presence of travelling waves can increase sonochemical activity [42]. The effect of frequency on coalescence can also be considered in terms of its effect on bubble size. At low frequencies, coalescence causes bubbles to grow into larger degas bubbles, whereas at high frequencies, coalescence allows small inactive bubbles to grow to active

bubble size [158]. At higher frequencies there will be a reduction in the distance between antinode regions which can contribute to an increase in coalescence [159]. Additionally, at higher frequencies, the small bubbles in solution are closer to the resonance size, which induces a greater secondary Bjerknes force between them which can lead to coalescence [37]. But this force is proportional to the volume change and since bubble size decreases with increasing frequency, there will be a smaller volume change and the force will decrease, decreasing coalescence [160]. It has been shown for a two bubble system, that frequencies of 80 kHz and 100 kHz have similar approach velocities but for bubbles in lower frequencies it decreases [37]. Also, smaller bubbles have a shorter film drainage time which increases the likelihood of coalescence compared to larger bubbles [161]. Therefore, coalescence needs to be considered at low frequencies due to increased bubble size and high frequencies due to increase in the secondary Bjerknes force and reduction in drainage time [161-163].

An increase in the number of active bubbles and reactive species with an increase in frequency has been widely observed [42, 106, 121, 164]. At higher frequencies, collapse and fragmentation occurs more quickly due to a decrease in the oscillation period, thus more fragmentation bubbles are produced over a shorter space of time that can then grow to become active [106]. This could be a result of bubble surface instabilities which have been calculated to occur more readily in the presence of travelling waves that are inherent at high frequencies [165]. The surface-to-volume ratio at higher frequencies also increases and this leads to increased diffusion and sonochemical reactions [166]. However, at higher frequencies bubbles are smaller in size and more stabilised against fragmentation. Frequency also determines reaction pathways within the bubble. At lower frequencies, radicals inside the bubbles have more time to follow pathways analogous to flame reactions, whereas at high frequencies, diffusion into the bulk solution increases and more hydroxyl radicals are released into the liquid [167]. Spectral emissions from multibubble sonoluminescence in argon atmosphere have shown that vibrational excitations of the plasma inside the bubble can change with frequency [81]. A disparity in the population of vibrational levels can have significant effects on the reactions taking place for several sonochemical systems [80]. As frequency is increased bubbles experience a shorter growth time and sonochemical yield decreases as observed between 585 and 1140 kHz [106]. It has been shown that, for sonochemical yield, mid-range frequencies ~300 - 500 kHz are those that are most productive [69, 121, 167, 168], although some authors suggest values closer to 1 MHz [42, 169]. Characterisation of these types of results is often considered from the viewpoint of bubble size rather than applied frequency and, as discussed, the size of bubbles is also dependent upon the pressure amplitude [152, 170]. This demonstrates the difficulty in a generalisation of results, with additional dependencies on other parameters, as discussed in this review. However, there is a general upper limit at higher applied frequency for which active cavitation decreases. If the bubbles are not of sufficient size then the severity of collapse will be limited and fewer radicals will be produced [25].

The influence of frequency on bubble surface instabilities and association with the degree of sonochemistry can be inferred by consideration of two limits. At low frequencies, bubbles will be larger, collapse vigorously and more readily demonstrate surface instability. However, at lower frequencies there is also increased coalescence due to increased bubble size. At high frequencies, antinode regions are closer to one another which can lead to an increase in coalescence. However, the bubbles are smaller and will be subject to more intense quasi acoustic streaming which can cause nonlinearity of bubble motion and instigate surface instabilities. Furthermore, at higher frequencies there is a reduction in the production of reactive species due to shorter growth time and reduction in collapse intensity. Therefore, in line with observations that mid-range frequencies further sonochemical activity, it is suggested that bubble surface instability effects become more apparent between an upper and lower frequency limit.

3.3. Reactor Design and Operating Parameters

The distribution and type of active bubbles is dependent on numerous system design parameters including; transducer and signal type, geometry of vessel, artificial or intrinsic (acoustic / quasi acoustic streaming) flow, liquid height and the presence of an extended surface in or at the liquid surface.

3.3.1. Transducer Type

The type of transducer used to sonicate a liquid will determine the nature of cavitation for different frequencies and pressure amplitudes. Areas of active bubble congregation are localised in standing wave formations for plate type transducers or in regions of high energy density for horn type transducers [44].

In consideration of travelling and standing waves, a plate transducer at low frequencies will produce primarily stable transient cavitation due to standing wave formation, placing active bubbles at the antinodes [44]. At high frequencies the standing wave becomes attenuated and the travelling wave component increases. This causes cavitation bubbles to be forced in the direction of the propagating wave towards the liquid surface via quasi acoustic streaming, where they are trapped in standing waves [44, 163]. Standing wave formation occurs at the surface of the liquid due to a high concentration of bubbles in that region reducing the intensity of the travelling wave component [25]. In this groups' previous work it was postulated that mid to high frequencies of 376, 995 and 1179 kHz for a plate transducer increased transient cavitation due to a travelling wave dominant field [42]. Others have observed that there is a change in the nature of the bubbles from transient to high energy stable bubbles below frequencies of 200 kHz [47]. However these results can be entirely dependent on the system, stable bubbles that produce an increase in sonoluminescence have been reported at a frequency of 515 kHz [48].

An ultrasonic horn operating at 20 kHz consists of both travelling and standing waves [171]. There is a limited conical active cavitation zone near the horn tip due to the high energy density delivered at that point as opposed to the plate transducer which delivers the ultrasonic wave more diffusely [43, 99, 150]. A thin layer of bubbles is formed at the horn surface which impedes the sound field's propagation and causes a reduction in pressure amplitude beyond its formation. This has been confirmed with measurements of pressure amplitude when the bubble layer is the size of an acoustic half wavelength, in this instance the impedance suddenly drops [138]. It has been suggested that travelling waves are more prevalent for the horn and that transient bubbles dominate [44, 48].

An ultrasound horn that operates at low frequency produces primarily travelling waves, and is beneficial in creating larger bubbles that collapse violently and are unstable in nature. This contributes to both to an increase in radical production and bubble instability. However, there can also be increased impedance due to bubble agglomeration at the tips surface and, as such, can be an inefficient method of achieving sonochemical reactions. Conversely, a plate transducer operating at high frequency may be advantageous with quasi acoustic streaming and radiation forces causing increased motion of the bubbles and active cavitation occurring in standing waves at the surface. Quasi acoustic streaming hinders coalescence, and increases the rate of growth and mass transfer that contributes to further instability and fragmentation.

3.3.2. Signal Type

Pulsed, sweeping or bi-modal signals can increase active cavitation compared to continuous wave output [117, 171-180]. Continuous wave ultrasound becomes inefficient due to limited spatial distribution, coalescence-induced loss of applied pressure amplitude and bubble growth beyond the region of active cavitation [172]. Pulsed wave ultrasound has been shown to increase the

sonochemically active region, the degree of sonochemistry and reduce bubble growth (and associated degassing) [175, 176, 178]. Pulsed wave ultrasound consists of a power on time, P_{ONT} and a power off time, P_{OFFT} . Generally, it is based on a critical P_{OFFT} at which the desired effects (for example, sonoluminescence or sonochemical activity) begin to reduce and is in the range of tens to hundreds of milliseconds depending on the frequency used [175]. The shorter P_{ONT} the shorter the lifetime of the active bubbles once the signal is switched off. That is, if P_{ONT} is short enough a bubble may dissolve completely during P_{OFFT} before the next P_{ONT} pulse, or subsequent P_{ONT} pulses may not cause remaining bubbles to become active [117, 176, 179]. Conversely, a longer P_{ONT} time can result in the bubbles becoming too large to dissolve to an active size during P_{OFFT} . This may cause bubbles to become inactive degas bubbles primarily via coalescence [176, 181]. A P_{ONT} can be chosen such that more bubbles become active during successive pulses and there can be a substantial increase in radical production [178]. In the P_{OFFT} phase there is a clear decrease in bubble volume due to the action of surface tension. However the bubbles may not completely dissolve or become inactive during this phase, this is due to residual pressure amplitude greater than the cavitation threshold which is said to remain during a short P_{OFFT} [172, 176, 179]. If a P_{OFFT} is chosen that gives large inactive bubbles chance to dissolve to an active size there are two benefits; firstly, the bubbles are now active and secondly, a reduction in bubble size may cause a decrease in coalescence and inhibition of the generation of degas bubbles [172, 182]. Therefore, by choosing the correct P_{ONT} to P_{OFFT} ratio, the lifetime of the bubbles' activity can be increased [174].

Results include an observed five-fold increase in sonochemical efficiency and three-fold increase in the oxidation of potassium iodide with appropriate P_{ONT} to P_{OFFT} ratios [172, 178]. The increase in sonochemical activity may be due to the conditions at the start of sonication which deliver increased violent volume pulsations of the bubbles, therefore, repeated pulsing of this period can result in increased disassociation inside the bubble and aid in fragmentation [65, 179]. This is especially true of large degas bubbles that can be forced to fragment and produce new bubbles that may continue the cavitation cycle [176]. Also, several authors have suggested a pulsed signal causes bubbles to move away from one another and / or cause migration from antinodes, reducing coalescence, preventing reduction in the pressure amplitude and facilitating an increase in the area of active cavitation area [23, 175, 178, 183].

Frequency sweeping has seen both positive and negative effects on sonochemical activity. Using high intensity focussed ultrasound (HIFU) and frequency mixing, 3.5 MHz pulses were applied to a 250 kHz continuous wave [184]. During the sweep from 250 to 290 kHz there was reduction in the active bubble population, proposing that a bubble cannot readily enter active oscillation and frequency sweeping resulting in less violent collapse. Further research has shown that sweeping upward (positive) or downward (negative) frequency from 3.6 MHz to lower and upper limits of 3.4 and 3.8 MHz is able to both quench and enhance inertial cavitation respectively i.e. a negative sweep increasing sonochemical yield [173]. In these results it was suggested that the effects are due to the variations in resonance bubble size which changes with frequency.

Another method of frequency utilisation is a dual frequency (or bi-modal) signal. This was observed to significantly increase sonoluminescence intensity due to bubble oscillation entering a regime inaccessible to single frequency driving. This resulted in a change in the active bubble population and a potential increase in bubble collapse temperature [185, 186]. The sonochemical activity taking place under these conditions can result in a sonochemical yield greater than the individual sum of the single frequencies that are used [180]. The proposed reason for this is a reduction in the standing wave and thus more distributed cavitation activity coupled with an increase in applied energy efficiency [187].

3.3.3. Ratio of Vessel / Transducer Diameters

The size and shape of the vessel for plate transducers can determine the influence of applied parameters and the cavitation activity [97, 153, 188-190]. For larger vessels there is an increase of calorimetric power in the liquid with vessel diameter, however this does not necessarily translate to an increase in sonochemical activity [97]. A problem for reactors with increasing volume, with similar transducer sizes, is the existence of 'dead zones' [97, 188]. That is, an area beyond a point where no transient cavitation can be detected and only non-cavitation behaviour such as bubble motion, acoustic streaming, etc. occurs [189, 190]. Vessels with a diameter greater than 90 mm have been shown to exhibit a reduction in sonochemical activity for a 60 mm diameter transducer [97]. Others interpret a vessel-transducer ratio dependence which correlates closely; a value of 1.45 found to exhibit the highest sonochemical and acoustic energy transfer efficiency across a range of liquid heights [153]. Nikitenko *et al.* used an ultrasonic horn to study the sonochemical darkening of diphenylmethane (DPhM) in water [191]. They found that the size of the vessel had negligible effect on the absorption of acoustic energy. However, the sonochemical efficiency increased with reduction in the tip diameter of the horn. This was reasoned to be due to an increase in maximum bubble size and associated increase in intensity of collapse.

3.3.4. Liquid Flow

The utilisation of flow to influence the cavitation environment can be applied via mechanical stirring [42, 99, 149, 192], circulation pumping [116, 158] or through frequency-power dependant quasi and acoustic streaming [61, 115, 128]. Flow can prevent cavitation bubbles from coalescing and clustering by counteracting the primary and secondary Bjerknes forces [158, 193]. Without flow, coalescence can cause bubbles to grow and be expelled from the antinode region, becoming inactive. Dependent upon the rate, flow can allow release of a limited number of bubbles from the antinode region which, when controlled, can reduce coalescence and enhance sonochemical activity [158]. Additionally, increased flow can refresh the solution around the bubble, thus delivering increased gas concentration and increasing mass transfer [141]. The motion of the liquid and subsequent movement of bubbles can also disperse bubbles from dead zones such that they may become subject to active regions within the field and increase overall activity [158]. The nonlinear motion of bubbles caused by fluid flow can instigate structural instability, asymmetric oscillation and collapse, which can increase sonochemical activity [115, 116].

Both quasi acoustic streaming and acoustic streaming can be a source of liquid flow. Quasi acoustic streaming due to attenuation of the wave causes bubble motion and the bubbles are less able to collapse and oscillate symmetrically [115, 158]. Thus, it may be conducive to bubble fragmentation and an increase in the release of active radicals into the solution. Calvisi *et al.* reported that as the fluid accelerates, due to quasi acoustic streaming in the presence of travelling waves, there is a local drop in pressure across the bubble surface resulting in surface instabilities and bubble shape distortion [128]. Acoustic streaming localised around the bubble also increases and the rate of dissolution or growth of the bubble can be intensified up to twentyfold due to surface instabilities with bubbles becoming non-spherical in nature [61]. As discussed, quasi acoustic and acoustic streaming are more prevalent at higher frequencies and powers, therefore liquid flow associated with these mechanisms will increase with their increase [44, 156, 194, 195]. Quasi acoustic streaming has been found to cause liquid flow in the 100's of kHz range with travelling waves becoming apparent at 448 kHz and dominating at higher frequencies ~726 kHz [163]. In this groups previous work it was suggested that this flow causes an increase in sonochemical yield up to a point when the frequency becomes too high to be favourable to produce an increase in active bubble population [42, 116]. It can be said that

streaming represents a viable method to decrease coalescence and increase bubble surface instability that may be advantageous for sonochemical activity.

Results from experiments inclusive of artificially applied flow (stirring or circulation pumping) are generally varied depending on applied power, frequency and reactor configuration. Overhead stirring for example has been shown to increase sonochemical activity at frequencies 40, 486, 490, 613 kHz and decrease activity at frequencies 376, 1179 kHz [42, 99, 149, 192]. A large increase (up to fivefold) in sonochemical yield with overhead stirring (600, 900, 1300 rpm) was found at 40 kHz with a general reduction for mid to high frequencies; 376, 995, 1179 kHz [42]. This work supported the theory that coalescence is reduced at lower frequencies. However, it was found that this not apparent for higher frequencies where coalescence can aid in formation of bubbles of active size and where quasi acoustic streaming can already reduce coalescence [42]. Similar stirring experiments at 486 and 490 kHz have yielded different results with reaction rates and sonochemical efficiency being found to increase [149, 192]. At 613 kHz sonochemical production has been found to increase with mechanical stirring up to 370 rpm (and gas flow rate), this was attributed to more homogeneous bubble distribution [99]. For circulation flow, an increase in yield was demonstrated at 376 and 995 kHz at a flow rate of 34 ml min⁻¹ and 76 W L⁻¹ [116]. Hatanaka *et al.* showed that peak sonochemiluminescence intensities occur at higher powers when circulation flow is increased, but reduce with increased stirring for lower powers [158]. Further, they found an optimal flow rate exists for sonochemical reactions in relation to pressure amplitude and frequency. In general the effects of stirring and flow are dependent on the individual system and applied parameters; the shape, size and position of a stirrer for example can present a significant effect on the sound field potentially reducing the sonochemically active region [192, 196].

Relating to bubble structural instability, it may be implied that flow can cause surface instabilities to occur due to fluid motion and mass transfer. These instabilities act with the already present quasi acoustic streaming and induce further acoustic streaming. The translational motion of the bubble as a result of surface instabilities is acted upon by the pressure wave and unstable oscillation, collapse and fragmentation can occur. The effect of quasi acoustic streaming is thus to incite further streaming and bubble structural instability. If higher pressure amplitudes are used, bubbles may agglomerate at the anti-node regions, due to “locking” in place. Upon applying flow, it is suggested that not only can there be a favourable reduction in the number of bubbles in those regions, but the bubbles that remain will be subject to more intense deformations caused by the flow. The structural instabilities in that region can then lead to further nonlinear effects, and finally fragmentation. Also, since high powers may result in increased intensity of collapse and subsequent bubble instabilities, flow offers a viable mechanism to work in synergy with power and negate effects such as coalescence.

3.3.5. Liquid Height

Several authors have influenced sonochemical activity with the adjustment of liquid height [153, 197-200]. In particular, fitting the height to a distance of an odd multiple of the fourth of the wavelength increased cavitation activity [151]. By ensuring the water level falls at an anti-node point, the air / water interface reinforces the standing wave component. Asakura *et al.* showed that, by choosing the appropriate liquid height, low frequencies can deliver similar levels of sonochemical activity to that of higher frequencies where it would not ordinarily be seen [197]. However, each individual reactor has its own optimal liquid height for maximising sonochemical activity and this is often dependent upon the systems effective absorption of acoustic energy [153, 198].

3.3.6. Liquid Temperature

The effect of increasing the temperature of the liquid is to decrease the cavitation threshold, this is due to an increase in the liquid vapour pressure or a decrease in surface tension or viscosity [166].

Additionally, the gas solubility of the liquid will decrease at higher temperatures and as a consequence there will be fewer cavitation nuclei available [201]. If the liquid temperature increases so does its vapour pressure which has been found to reduce peak cavitation temperatures and the rate of sonochemical reactions [202]. This is especially true of more volatile reactants and is proposed to be due to more vapour entering the bubble at higher temperatures and in effect ‘cushioning’ the collapse [147]. A change in temperature can influence other parameters, frequency for example; an increase in liquid temperature to 40°C has been seen to cause a shift of maximum H₂O₂ production from the range 204 - 362 kHz to 613 kHz [99].

3.3.7. Reflective Plate

A reflective plate or surface stabiliser reinforces the standing wave component of the sound field resulting in an increase in pressure amplitude that can effect both sonoluminescence and sonochemiluminescence [42, 156, 196]. Zhou *et al.* proposed that although sonoluminescence is increased with surface stabilisation, sonochemiluminescence activity is lowered due to reflective reinforcement of the standing wave component [156]. However this is dependent upon the frequency; in this groups previous work there was shown a decrease in sonochemical activity of up to 69% with surface stabilisation for 376, 995 and 1179 kHz at low and high powers [42]. This indicates that for those frequencies the travelling wave component is the primary contributor to sonochemical activity. The region of active cavitation also changes, with substantial activity at the reflective plate which is maximised when positioned at the liquid surface [196, 203]. The action of a lid behaving as a surface stabiliser and reducing travelling waves is to reduce the effect of (quasi) acoustic streaming, which may similarly account for a reduction in transient cavitation and associated sonochemical activity [42].

The reinforcement of the standing wave via a reflective plate can be particularly effective at controlling the system, especially where high frequencies and flows have been implemented such that the standing wave is distorted and sonochemical activity reduced. Therefore, this lends to a theoretical method for increasing the structural instability of bubbles. That is, the bubbles can be strongly held in place by the reinforced standing wave, combined with a flow through system causing surface instabilities, nonlinearity oscillation and collapse.

3.4. Implications of Primary Parameters

From the discussions it is clear that the primary parameters represent a means of actuating and controlling the activity taking place within a sonochemical system. But even more apparent is their interlinked behaviour and effect on one another; by changing one parameter all others can be affected. Thus, this demonstrates the individualistic nature of sonochemical systems, a set of parameters that delivers high sonochemical yields for one system will not provide the same results when applied to another system. Add to this the chaotic and unpredictable nature of a multi bubble environment and the effect of parameters for different systems becomes even more elusive. However, general trends have been presented and they offer some understanding as to the mechanisms of individual parameters and their influence on each other. It is now of use to extend this knowledge to include parameters that can diversify the effects of these fundamental operating parameters.

4. Secondary Parameters

Upon the realisation of a working sonochemical system via primary parameters, augmentation may be achieved via the introduction of secondary parameters, namely; the introduction of solids, changes to the chemical environment and sonication in gas atmospheres.

A summary table of literature covering chemical and gas-liquid experimental results is given in Table 1 and 2.

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Table 1. Sonochemical Effects in Various Chemical Environments

Chemical	Freq. (kHz)	Power / Pressure Amplitude	Temp (°C)	Effects	Reference
Al powder (15mm mean size) in starch solution.	498	50 W	22	Enhanced liquid mixing. Decolourisation reaction rate (between I and Na ₂ HPO ₄) increased up to 70% with powder. Sonochemiluminescence maximised at 0.0034 g/L.	[204]
TiO ₂ powder (specific surface area 11 m ² /g) in water.	20	50 W	23 ± 3	29% increase in sonoluminescence for (0.02 g/ 100 ml solution). Thermo-mechanical heating excites charge carriers to catalyse dioxane decomposition. The formation of a TiO ₂ + •OH state that increases sonochemical activity is hypothesised. Oxygen vacancies in the structure suggested to causing increase in nucleation.	[205]
Zn, Ni, Cr, and Mo powder (~5 - 10 µm diameter) in C ₁₀ H ₂₂ solution.	20	50 W/cm ²	N/A	Inter-particle melting for Zn, Ni, Cr, and Mo indicating collision temperatures of between ~2800 K and 3600 K.	[103]
TiO ₂ powder (~30–50 nm, ~20 x 12 nm), ZrO ₂ powder (~20 nm), ThO ₂ powder (~5 µm, 4.5 µm, 5 nm)	20 362	0.34 W/mL 0.25 W/mL	22	Increase in hydrogen yield in argon atmosphere for nano-metal oxide suspensions (TiO ₂ , ZrO ₂ , ThO ₂) at 20 kHz and 362 kHz, due to enhanced nucleation. Additional mechano-chemical effects evidenced at 20 kHz in the presence of micro-particles.	[206]
DTAC in water.	22.31	27 ± 3 kPa	20 ± 3	Increase in bubble growth rate due to charge and size of the head group. Increase in bubble surface instabilities and acoustic streaming.	[141]

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Propan-1-ol, SDS, SDBS, AXC, DTAC, ZW3-10 solutions.	515	20 W	20 - 21	Decrease in total bubble volume indicating a reduction in coalescence due to electrostatic effects.	[38]
10% Surface-active agent, 40% ethylene glycol, 5% methanol and water.	19.5	N/A	25	Increasing surface tension with a wetting agent caused an increase in bubble size and the formation of bubble clusters. This, due to stabilisation of bubble growth reducing the potential for fragmentation.	[207]
CH ₃ OH, C ₂ H ₆ O, C ₄ H ₉ OH solutions.	20 (3.5ms pulsed) 515 (3.5ms pulsed)	0.09 – 0.2 W/cm ² 0.04 – 0.4 W/cm ²	20	Sonoluminescence quenching for high and low frequencies which increased with alkyl chain length. At 20 kHz increasing concentration has no effect. For 515 kHz sonoluminescence peaked at 2 to 3 mM. Quenching attributed to hydrophobic molecules entering the bubble decreasing intensity of collapse.	[48]
C ₆ H ₁₀ O ₇ , C ₆ H ₉ NO, (C ₄ H ₆ O ₂) _n , C ₂ H ₄ O) _x , C _{2n} H _{4n+2} O _{n+1} solutions.	1000	2 W/cm ²	N/A	Change in surface tension with polymer concentration indicating adsorption at bubble surface. Increase in the negative change of surface tension increased the rate of pyrolysis (CO production).	[113]
C ₂ H ₆ O ₂ solution.	23	0.8 bar	18	Experiments fit calculations very closely with bubbles in the viscous fluid residing within or close to the 'shape stable path unstable' phase.	[208]
Dissolved CO ₂ species in water.	928 (Pulse on 50 ms, pulse off 175 ms)	0.42 W/cm ²	20.5	Change in coalescence behaviour due to reduction in surface pH. Increase in cavitation activity with pulse off time and CO ₂ concentrations attributed to reduction in coalescence. The mechanism is suggested to be due to adsorption of ions at the bubble surface reducing the film drainage between bubbles.	[209]

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NaCl in water.	20	600 W	0 - 4	Increase in oxidising rates of aqueous pollutants due to an increase in the ionic strength of the solution. The result is to cause pollutants to move to the bubble surface where they can be subject to increased oxidation.	[210]
NaCl in water with He and Ar bubbling.	355 (4ms pulsed)	33 W	~22	Reduction in bubble size due to reduction in gas content by the salt.	[211]
NaCl, NaBr, KCl, NaClO ₄ , Mg(ClO ₄) ₂ , MgCl ₂ , AlCl ₃ , CH ₃ COONa, MgSO ₄ , Na ₂ SO ₄ and HClO ₄ solutions.	515 (3.5ms pulsed)	0.3 W/cm ²	22	With addition of the salts there is an increase in sonoluminescence intensity (with the exception of HClO ₄) until a limit is reached where it begins to reduce. The change in gas solubility of the liquid with salt is expressed as the most likely cause of the effects.	[212]
KCl, HCl and NaNO ₃ solutions.	515 (5.8 ms pulsed)	10 W	20	Coalescence reduced due to reduction in gas content also known as a "salting out" effect of the electrolyte.	[162]
MgSO ₄ , NaCl, KCl, HCl, H ₂ SO ₄ solutions.	213	25 W	25	The change in volume of the solution (a direct indication of the coalescence occurring) goes in the order MgSO ₄ > NaCl ≈ KCl > H ₂ SO ₄ ≈ HCl. Adsorption at the bubble surface cause surface tension gradients that effect the film drainage between bubbles.	[213]
Na ₂ SO ₄ in C ₇ F ₁₅ COOH solution.	40	150 W	Various	An increase in surface tension caused an increase in PFOA (surfactant molecules) to the bubble surface. Sulfate addition decreases surface tension however this was seen to stabilise and there was increase degradation of PFOA with sulfate concentration.	[214]

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Ketones and n-alkyl acetates in water.	20 515	0.1 – 0.4 W/cm ²	20 -25	Low concentrations of solutes give an increase in sonoluminescence suggested to be due to a reduction in coalescence. However in larger concentrations can quench sonoluminescence due to a build-up of concentration at the bubble surface and the solute entering the bubble.	[215]
Formation of amino acids in acetic acid solutions or under CH ₄ or CO ₂ with N ₂ .	355	70 W	~22 - 30	Amino acid formation for nitrogen and separately nitrogen with hydrogen, both in aqueous solutions with argon atmosphere, followed the order alanine > ethylglycine > glycine. Methane was the exception where for both nitrogen and hydrogen the rate of production followed the order glycine > alanine > ethylglycine.	[216]

Table 2. Gas-Liquid Sonochemical Studies

Experiment / Method	Freq. (kHz)	Power / Pressure Amplitude	Temp (°C)	Products	Results	Reference
Formation of H ₂ O ₂ in Ar/O ₂ , UV-Vis with Ti(IV) in 0.5M H ₂ SO ₄ .	20 204 362 613	40 – 43 W	20	H ₂ O ₂	Optimal at 204kHz (~1.4 μmol/kJ) - 362kHz (1.75 μmol/kJ), 613kHz Ar/20vol%O ₂ .	[99]
Formation of H ₂ O ₂ in O ₂ Ar N ₂ , UV-Vis with FeSO ₄ in acid solution (Fricke).	447	50 W	Room Temperature	H ₂ O ₂ , HNO ₂ , HNO ₃	H ₂ O ₂ yield follows order O ₂ > air > Ar > N ₂ and acid (nitrous and nitric) follows order air > N ₂ > Ar > O ₂ .	[91]
Formation of H ₂ O ₂ in Ar/O ₂ , UV-Vis with (NH ₄) ₂ MoO ₄ . CO ₂ . Degradation of HCOONa in Ar/O ₂ , GC.	300	N/A	Room Temperature	H ₂ , H ₂ O ₂ , CO ₂ , (CO ₂ H) ₂	Optimal (H ₂ O ₂ ~2.7x10 ⁻⁴ M min ⁻¹) at Ar/30vol%O ₂ .	[217]

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Degradation of CO ₂ in CO ₂ /Ar under sonophotocatalysis, titanium sulfate and GC.	200	200 W	20	CO, H ₂ , O ₂	Products: ~11 μmol H ₂ , ~111 μmol H ₂ O ₂ , ~16 μmol O ₂ . Maximum rate of CO in 2–3vol%CO ₂ in Ar.	[218]
Formation of H ₂ O ₂ in H ₂ /O ₂ , UV-Vis with 1 mM FeSO ₄ , 10 mM CuSO ₄ , 10 mM H ₂ SO (Fricke).	300	N/A	Room Temperature	•OH, HO ₂ , H ₂ O	Optimal rate (27 μmol/min) at H ₂ :O ₂ ratios of 1:3 and 2:1.	[219]
Product analysis in CO ₂ /Ar; UV-Vis with KI and Ti(SO ₄) ₂ in 9M H ₂ SO ₂ , and GC.	2400	15 W	25	H ₂ O ₂ , H ₂ O ₂	Varying product yields [49 μmol H ₂ O ₂ , 5.5 μmol H ₂ , 6.1 μmol O ₂ , 23 μmol CO] Optimal rate at Ar/2vol%CO ₂ .	[95]
Cavitation noise spectra in O ₂ and CO ₂ atmospheres. Acoustic spectral measurement.	928	Variable	20.5	N/A	Cavitation noise activity absent in solutions saturated with CO ₂ at power densities less than 1.7 W/cm ² . CO ₂ and O ₂ in variable combinations give a measurable cavitation noise signal at 420 mW/cm ² .	[209]
Sonochemical oxidation efficiency in Ar, He, N ₂ , O ₂ , CO ₂ atmospheres. UV-Vis with KI.	20 41 62	1 – 6 W	20	I ₃	At 20 kHz oxidation efficiency goes as Ar > O ₂ > Air At 40 kHz oxidation efficiency goes as Air > O ₂ > Ar Highest oxidation efficiency achieved at 60 kHz under air.	[150]
Production of H ₂ O ₂ and •OH under Kr, Ar, He and O ₂ . UV-Vis with KI.	20 40 80 500	Calorimetric	25 - 30	•OH, H ₂ O	Optimum rate of rates of H ₂ O ₂ (pH 7, 2.94 μM min ⁻¹) and •OH (pH 11, 0.391 μM min ⁻¹) under Kr at 500kHz. Lowest rates of H ₂ O ₂ (0.0508 μM min ⁻¹) and •OH (0.0310 μM min ⁻¹) under He at 20 kHz.	[220]
Product Analysis in CO ₂ /Ar and CH ₄ /Ar, UV-Vis	300	3.5 W/cm ²	20	C ₂ H ₆ , C ₂ H ₄ , C ₂ H ₂ , CO, CO ₂ , H ₂ O ₂	Varying product yields. Ar/CO ₂ : Optimal O ₂ at Ar/3vol%CO ₂ , optimal H ₂ at Ar/0vol%CO ₂ , optimal H ₂ O ₂ at Ar/5vol%CO ₂ , optimal CO at Ar/2vol%	[221]

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with KI and TiOSO ₄ · aq. GC and ion chromatography.					CO ₂ . Ar/CH ₄ : Optimal H ₂ at Ar/1% CH ₄ , optimal CO + CO ₂ at Ar/8vol%CH ₄ , optimal C ₂ H ₄ + C ₂ H ₆ at Ar/1vol%CH ₄ .	
Formation of N ₂ , O ₂ in Ar/N ₂ O. Degradation of N ₂ O in Ar/N ₂ O.	300	1.5 bar	Room Temperature	N ₂ , O ₂ , NO ₂ , NO ₃ ⁻	Optimal formation of N ₂ , O ₂ at Ar/20vol%N ₂ O. Optimal degradation at Ar/15vol%N ₂ O.	[222]
Degradation of CO ₂ in CO ₂ /Ar, GC.	200	200 W	Room Temperature	CO, H ₂ , O ₂	Optimal at Ar/3vol% CO ₂ .	[223]
Degradation of 1,4-dioxane. Formation of H ₂ O ₂ in Ar/O ₂ . UV-Vis with KI.	205 358 618 1071	128 W or 5.1 W/cm ²	20	H ₂ O ₂	Optimal decomposition rate at 355kHz Ar/25%O ₂ Optimal formation rate at 355kHz Ar/25%O ₂	[69]
Formation of H ₂ O ₂ in H ₂ O and HNO ₃ . In Ar and Ar/20%O ₂ atmosphere. Degradation of HNO ₃ . UV-Vis with TiOSO ₄ . Mass spectrometry.	20 207 615	Calorimetric	21	H ₂ O ₂	Formation rate H ₂ O ₂ more than double in Ar/20vol%O ₂ compared to Ar. Optimal H ₂ O ₂ rate for 20kHz in HNO ₃ Ar/20vol%O ₂ atmosphere. H ₂ O ₂ yield increases for higher frequencies in water and increases for lower frequencies in nitric acid. Optimal yield (1.56μmol kJ ⁻¹) at 615 kHz Ar/20vol%O ₂ in water.	[224]
Formation of amino acids in acetic acid solutions or under CH ₄ or CO ₂ with N ₂ .	355	70 W	~22 - 30	Alanine, Ethylglycine, Glycine	Amino acid formation for nitrogen and separately nitrogen with hydrogen, both in aqueous solutions with argon atmosphere, followed the order alanine > ethylglycine > glycine. Methane was the exception where for both nitrogen and hydrogen the rate of production followed the order glycine > alanine > ethylglycine.	[216]

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<p>Formation of tri-iodide in KI and H_2O_2 in water. Comparative analysis at 514 and 20 kHz in Ar and O_2 saturated liquid. H_2O_2 ammonium molybdate decomposition reaction in a 10% KI Tri-iodide UV-Vis with KI.</p>	<p>20 514</p>	<p>30 W</p>	<p>24</p>	<p>H_2O_2</p>	<p>Formation rate of I_3 in O_2: 514 kHz $\sim 1.75\mu\text{M}/\text{min}$ I_3 20 kHz $\sim 0.10\mu\text{M}/\text{min}$ I_3</p> <p>Formation rate of I_3 in H_2O_2: 514 kHz $\text{O}_2 \sim 7.80\mu\text{M}/\text{min}$ H_2O_2 20 kHz $\text{O}_2 \sim 0.20\mu\text{M}/\text{min}$ H_2O_2</p> <p>Formation rate of I_3 in Ar: 514 kHz $\sim 0.25\mu\text{M}/\text{min}$ I_3 20 kHz $\sim 0.25\mu\text{M}/\text{min}$ I_3</p> <p>Formation rate of H_2O_2 in Ar: 514 kHz Ar $\sim 6.50\mu\text{M}/\text{min}$ H_2O_2 20 kHz Ar $\sim 1.70\mu\text{M}/\text{min}$ H_2O_2</p>	<p>[167]</p>
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4.1. Solid Addition

The study of the presence of solids (or surfaces) in cavitation environments is extensive and originally stemmed from observations of cavitation damage to ship propellers in the late 19th Century [225]. More recent studies explore the reaction rate enhancement in both stoichiometric and catalytic reactions [103]. If there is an extended solid surface greater than 150 μm in the sonicated liquid it is able to cause nearby bubbles to experience non-spherical collapse with high speed jets forming on their proximal side which impact the surface [16, 128, 146, 226, 227]. For some metals the jet will perforate the surface at the point of collapse, exposing a highly heated chemically reactive surface [16]. In the presence of particles smaller in size, such as powders, the shockwave from bubble collapse causes particles near to one another to experience high velocity inter-particle collisions and melting [16, 103, 147]. It has been observed that for zinc, nickel, chromium and molybdenum interparticle melting can occur, however, this is not so for tungsten which indicates temperatures at collision sites must reach between 2800 K and 3600 K [226].

Powders can act as sonochemical catalysts via chemical, physical or combined mechanisms. Similar to extended surfaces, the surface oxide coatings of metal powders can be removed, improving rates of chemical reactivity and yields [103, 204]. Hydrogen yields in argon-saturated water with metal oxide particles was able to be increased at 20 kHz and 362 kHz attributed to increased nucleation sites in crevices at the particles surface [206]. At 20 kHz, additional increases in hydrogen yields with micron-sized oxides were accompanied by the fragmentation of particles, indicating an additional mechano-chemical contribution to the water splitting process. However at 362 kHz, micro-particles caused attenuation of the sound field, yet the smaller nanoparticles caused both increased nucleation and ultrasonic attenuation. The inherent molecular properties and defects of small particles can cause an increase in mixing and act as sources of nucleation. Zhou *et al.* noted an increase in bulk liquid flow and enhanced liquid mixing (via turbulent flows) upon addition of aluminium powder which resulted in increased sonochemical reactions [204]. TiO_2 powder, aside from causing excitation of charge carriers to improve sonochemical decomposition rates, has an intrinsic oxygen vacancy that has been suggested to contribute to structural defects which may act as sources of nucleation [205]. With regards to solids in ultrasound environments, although there has been extensive work in synthetic fields, such as synthesis of nanostructured inorganic materials or biomaterials [22, 103], there are few studies on the effect of powders on sonochemical activity.

4.2. Surfactants

Surfactants can influence the properties of cavitation bubbles, in particular their growth and surface stability, due to the charge and character of the head group [141, 228, 229]. By their nature, surfactants will be adsorbed at surface regions and therefore in a liquid environment where there are bubbles present they will gather at the bubble-liquid interface.

A bulky head group has been proposed to induce bubble surface instabilities and associated acoustic streaming that results in an increase in rectified diffusion [141]. However, surfactants with long chains have been observed to increase boundary resistance to mass transfer through the bubble surface [230]. This results from less mass transfer on contraction due to decrease in the bubble surface area and increase in surface surfactant concentration hindering outward diffusion [229]. On expansion there is less resistance to mass transfer as the surface area increases and the surface surfactant concentration is reduced. The resistance to mass transfer has been shown to be surfactant concentration dependant; 10 mM as opposed to 1 mM of DTAC presents an increase in resistance [141]. It has been found that as the alkyl chain length of the surfactant increases the amount of solute able to be adsorbed at the surface decreases [231]. Therefore the degree to which the surfactant is able

to be adsorbed at the surface of the bubble presents a limiting factor rather than the concentration of solute in the solution [215]. This has been attributed to surfactants aggregating into micelles above a certain concentration (critical micelle concentration) and once reached, growth rates become dependent on the increase bubble surface oscillations [229].

The ionic surfactant adsorption of a hydrophobic solute can cause a reduction in coalescence via steric and electrostatic repulsion particularly when surface-active solutes are charged [31, 90]. This is evident when salt, which acts to inhibit electrostatic effects, is present in dodecyl trimethyl ammonium chloride (DTAC) solutions; the associated electrostatic effects of the surfactant are no longer observed [141]. It has been found that the total volume of bubbles decreased in the presence of surface active solutes, and further, smaller bubbles and narrower size distribution resulted, both due to a proposed reduction in coalescence [38, 170]. Sodium dodecyl sulphate (SDS), a surface active solute, reduced coalescence by approximately 20% at 515 kHz due to electrostatic effects [38]. However, if electrostatic repulsion is the primary mechanism of reducing coalescence then it should be considered that the associated forces only dominate for small bubble oscillations and at higher pressure amplitudes the attractive Bjerknes forces dominates, reversing the effect [232]. At this point steric repulsion as seen for uncharged and zwitterionic surface-active solutes can play some role in reducing coalescence and associated effects [38]. It was suggested that by using a highly charged surfactant bubbles would not be able to reach minimum size due to repulsions at the surface, however this was not apparent during experimentation [229]. Alcohols (especially with a terminal hydroxyl group) act in a similar vein to SDS with their nonpolar and polar surface properties reducing coalescence and increasing the number of active bubbles [31, 47, 231]. Tronson *et al.* found that water solutions containing certain aliphatic alcohols quenched sonoluminescence at 515 kHz but indicated that by choosing the correct frequency it would be possible to increase sonochemical reactions [48]. However, sonochemical reactions are prevalent during transient cavitation and for substances such as acetone which does not have the ability to hydrogen bond transient cavitation can be quenched [215].

4.3. Changes in Liquid Properties

4.3.1. Surface Tension and Viscosity

If the solution has high surface tension or viscosity, either chemically or induced, there can be a direct effect on cavitation activity. When the surface tension is high, greater pressure is required for the nucleation of cavitation bubbles [233-236]. However, after nucleation, surface tension has less of an effect on the restriction of growth and provides bubble growth stabilisation, thus bubbles are more likely to grow to active size without breaking apart [207]. The overall effect of increased surface tension is to provide resistance to growth which is most prominent during the nucleation stage, this can result in a decrease in oscillation period [234]. For two moving (rising) bubbles with high surface tension, coalescence was determined to occur less readily due to the bubbles increased resistance to deformation and stretching between the bubbles in contact [237]. However for stagnant bubbles or bubbles in clusters there has been suggested a decrease in coalescence with decrease in surface tension [40]. In terms of reaction mechanisms, Henglein *et al.* found a distinct relation; if a polymer decreased the surface tension then the surface would be able to become more heavily loaded [113]. This would relate to surface concentration properties as previously discussed.

Acid or similar can be used to increase viscosity and influence bubble dynamics, without affecting the vapour inside the bubble. Phosphoric acid's vapour for example consists only of water molecules, with no significant trace of the acid molecules vapour at high temperatures [238]. An increase in viscosity due to reinforcement of the intrinsic bonding forces of the liquid requires a higher applied pressure to induce nucleation and cavitation [166]. This resistance to pressure amplitude is demonstrated when it is considered that oscillation period of the bubble increases with viscosity [234].

This is particularly true when the bubble approaches its maximum; the larger the bubble the more energy must be exerted on the surrounding liquid and hence the energy needed for expansion is greater [234]. Thus, an increase in pressure amplitude is required to achieve growth and furthermore, instigate bubble oscillation [50].

Calculations have demonstrated that in general the stability of the bubbles is increased with viscosity [239]. There are two proposed mechanisms that influence bubble structural stability at high pressures: Rayleigh-Taylor instability due to gas being strongly forced into the liquid, and bubble surface instability due to oscillatory effects [240, 241]. Brenner *et al.* proposed that increased viscosity would decrease bubble surface instabilities and the bubble would initially enter the Rayleigh-Taylor regime [242]. Further, since Rayleigh-Taylor instability acts so quickly, the bubble would have no time to stabilise and would fragment, as evident by the absence of sonoluminescence in highly viscous fluids. There has been observed an increase in the destabilisation of sonoluminescing bubbles' motion trajectory and surface stability in viscous liquids such as glycol, methylformamide, or sulphuric acid [208]. The elliptical pathways seen for higher viscosity fluids contribute to bubble surface instabilities due to inertial forces and can determine the upper boundary of pressure amplitude that facilitates high energy stable oscillation [85, 243].

The effect of viscosity on coalescence in a cavitation, multibubble environment is not easily determined. Aside from the above effects, in very viscous fluids, there is possibility of attenuation of the sound field and reduction in the areas of cavitation [244]. In the absence of ultrasound, Oolman *et al.* presented numerical values for the physical properties of liquids and related to bubble coalescence times [40]. For small values of viscosity (~1 - 3 cp) there is a varied relation to coalescence time, however for a large viscosity it is clear that the coalescence time increases. This could imply that highly viscous fluids inhibit bubble surface motion and film drainage between the bubbles in contact, which is known to reduce coalescence [213]. However, the time for coalescence to occur becomes relevant in an ultrasound environment due to time dependant forces on the bubbles [36]. Numerical analysis of a two bubble system suggests that increasing the viscosity of the liquid, in this case sulfuric acid, would decrease the secondary Bjerknes force, approach velocity and therefore increase contact time [245]. For low viscosity fluids it has been calculated that bubble structural instability is increased and the bubbles' approach to one another is inhibited [237]. This again may suggest reduction in coalescence is possible with an increase in solution viscosity.

4.3.2. Changes in pH

A change in the pH of the bulk solution during sonication is dependent on the solution used. For water in air atmosphere, in the short time after application of ultrasound there is an initial decrease from pH 7 to ~pH 4 [246]. This decreasing trend is also seen for oxygen and argon atmospheres, however for nitrogen there is an initial small increase followed by a gradual increase [91]. The effect of decreasing pH with sonication in air is due to the •OH radicals oxidising nitrogen, resulting in the formation of nitrous and nitric acids [247]. However, in air-saturated aqueous NaOH (pH 12) sonication resulted in the formation of the peroxyxynitrite anion, likely to be produced from the superoxide radical formed in the basic conditions [248]. In terms of sonochemical effects, an increase in the production of H₂O₂ has been seen to occur with a change in pH, however there is little to suggest there is a direct relation [246]. In degradation experiments with perfluorooctanoic acid there was considered to be no direct influence in the change in pH on chemical reactions taking place [214].

Price *et al.* considered that equilibrium adsorption of volatile solutes at the bubbles surface may not be readily achieved for active cavitation bubbles [215]. However, when the pH is changed to alter the solute's hydrophobicity this will effect where it gathers in the solution. For example, when molecules are changed to their more volatile version using pH, they are evaporated into the bubble and are able

to quench sonoluminescence. However, in the ionic form, the molecules remained in the solution and did not quench sonoluminescence, although they were still subject to radical degradation reactions [64, 215, 231]. Thus, pH may influence chemical dynamics of the system such that it may act to control the reactions taking place.

The change in pH will also affect the charged species adsorbed at the surface of the bubble. In general, at low pH values, positive charge groups (H^+) will be adsorbed at the surface [249], whereas at higher pH values there is negative charge (OH^-) adsorption [250]. The change in surface adsorption of species can influence coalescence via surface tension or steric mechanisms [251]. Lorentz, in early work, suggested surface tension independence from pH 1 to pH 13, citing that absorption of hydroxide ions is always balanced by equilibrium conditions in the form of similar absorption of hydronium ions [252]. However, there is an unaccounted for dynamic surface tension in this context. That is, adsorption at the surface of the bubbles causes a change in concentration of the solute in bulk solution and there is an associated change in the surface tension [251]. If the change in surface tension is positive then this indicates negative surface excess concentrations, if it is negative then there is positive excess surface charge adsorption [253]. In addition, the variation of charged molecules at the surface can cause a shift in the isoelectric point (a point where there is no net charge) of the surface of the bubbles which may induce electrostatic repulsion, not only this, but adsorbed species at the surface of the bubbles can contribute to steric repulsion (Pauli repulsion) between bubbles [209, 254].

When CO_2 is dissolved in water it offers a particularly unique means of demonstrating these behaviours, this is because it may form several complex species as shown in equations (9) – (12) [255].

The adsorption of these species is suggested to occur at the bubble surface due to a change in surface pH and reduction in surface tension of the liquid [209, 256]. CO_2 for example can change the surface tension of water from 72 mN m^{-1} to 57 mN m^{-1} as its pressure changes from 1 to 11 bar [257]. This relates to an increase by approximately a third in the cavitation threshold [235]. Brems *et al.* proposed that a reduction in pH at the surface indicates surface activity of hydronium ions and could have some influence on coalescence [209]. In the absence of ultrasound it has been shown that CO_2 bubbles do not coalesce below pH 6, this behaviour comes from the observed difference in surface charge for different pH values [254].

4.3.3. Salt Solutions

Seymour *et al.* found an increase in oxidation reactions occurring at the bubble surface with the use of sodium chloride [210]. The group suggested that there was an increase in the ionic strength of the liquid which drove organic pollutants to the region. Furthermore, it was seen that there was a decrease in surface tension at the bubble surface due to the presence of the excess hydrophobic molecules. Similar observations were observed for hydrophobic solutes which have the effect of driving hydrophobic ions to the bubble surface region where they may react with $\bullet OH$ radicals formed in the bubbles [18]. This effect is not electrostatic in nature as it has been shown that salt reduced the

strength of electrostatic repulsion when used in conjunction with head groups that induce surface charge [38].

Several authors have recognised the ability of electrolyte solutions to influence the size of bubbles during sonication, the severity of collapse and coalescence [162, 211-213, 251]. There are two suggestions as to the electrolytes' effect. Firstly, ion absorption at the bubble surface that influences coalescence via reduction in film drainage between bubbles in contact and steric repulsion [213]. Secondly, a reduction in the gas content of the liquid due to a "salting out" effect that both reduces the amount of nucleation, clustering of bubbles and would decrease attenuation of the sound field [162, 211, 251]. Sonochemical yields, aside from the above observations, have been suggested to increase due to less vapour entering the bubbles causing an increase in the severity of collapse [212].

4.4. Effect of Gases

In the presence of gases, the intrinsic nature of the gas will determine the physical and chemical reactions that occur during sonication. For noble gases an increase in sonoluminescence goes in the order $\text{He} < \text{Ne} < \text{Ar} < \text{Kr} < \text{Xe}$, in agreement with theoretical temperatures for the gas inside the bubble [78, 258]. Similarly, at 20 and 513 kHz (10 W/cm^2 and 1.5 W/cm^2 respectively) the zero order rate of production of H_2O_2 increases in the order of $\text{He} < \text{Ar} < \text{Kr}$ [220]. Pflieger *et al.* simulated the spectral emission from inside the bubble during sonication for Ar and Xe in consideration of electron (T_e), vibrational (T_v), rotational (T_r) and translational (T_g) temperatures contributing to non-equilibrium plasma formation [259]. Xe showed a significant increase in vibronic temperatures (T_v, T_e) compared to Ar due to its lower ionising potential. This indicates that for Xe, the conditions inside the bubble are more energetic, which correlates with the order of increase in sonoluminescence.

The energy developed and associated temperature on collapse can be approximated assuming adiabatic compression of an ideal gas and is directly related to the adiabatic ratio of the gas content [78]. However, the higher the thermal conductivity of the gas the less adiabatic the process [258]. Generally, a more conductive gas will lower the maximum temperature reached, this is because the heat will dissipate more quickly from the areas of cavitation. The addition of a polyatomic gas will act to reduce the bubble temperature due to its higher heat capacity [260]. If the vapour of the liquid has a lower adiabatic ratio than the gas it can also influence the temperature upon collapse [261]. That is, for a liquid with a low vapour pressure, less vapour will be able to enter the bubble and cushion the collapse, therefore it occurs more violently, increasing temperature [147]. Indeed, as a result of solubility, there is a direct influence on the size of the bubbles during cavitation which has been shown to go $\text{air} > \text{Ar} > \text{He}$ [162]. Highly soluble gases also result in the formation of an increased number of cavitation nuclei and lower the threshold for nucleation [262]. This is likely due to a reduction in the surface tension of the liquid and can cause the mass transfer process to occur more readily [263].

When the liquid is saturated with different gases, the chemical reaction cascade, and subsequent product formations can change [32, 221]. Suslick *et al.* suggest that for high sonochemical yields, reactants should be highly volatile such that more vapour can enter the cavitation bubbles [147]. This is confirmed by increased $\bullet\text{OH}$ yields observed for gas atmospheres in the order $\text{Kr} > \text{Ar} > \text{O}_2 > \text{He}$ [220]. Henglein provides an example of the concentration dependence of products under argon-methane atmospheres; at low concentration of methane a 2:1 ratio exists of hydrogen formation to methane loss, however at higher concentrations, methane is more readily consumed [10]. Also at low concentrations, a greater amount of unsaturated hydrocarbons were formed, however at high concentrations formation is reduced. Consideration of gas-liquid sonochemical systems may be

separated into overall gas effect on sonochemical reactions, including gas mixtures and the effect on products from gas-liquid interactions.

4.4.1. Gas Saturation Effect

A solution saturated with argon is known to enhance pyrolytic reactions due to the high temperatures that can be achieved [23, 73]. It has also been suggested to contribute to electron excitation energy that is transferable to the water molecules [264]. Since it allows for enhanced reactions inside the bubble, radical induced reactions in the liquid and bubble surface, and is relatively inexpensive, it is the choice for many researchers [171]. In terms of sonochemical reactions, a frequency dependence for potassium iodide oxidation was found; 20 kHz gave greater efficiency for argon, whilst 60 kHz was found to be more efficient for air [150]. More recently, frequencies above 515 kHz have been calculated to decrease $\bullet\text{OH}$ production rate in the order $\text{Ar} > \text{O}_2 > \text{air} > \text{N}_2$, while decreasing frequency to 213 kHz the order becomes $\text{O}_2 > \text{air} \sim \text{N}_2 > \text{Ar}$ [84]. This may be reflective of bubble size and the change in available time for reactions to occur which is dependent upon the applied frequency.

Oxygen's disassociated products have a direct effect on the reaction scheme. This is because they form reactive species that may scavenge hydrogen atoms and reduce recombination of $\bullet\text{OH}$ and $\bullet\text{H}$ at the bubble surface region [167]. Their ability to do this stems from $\bullet\text{O}$'s high reactivity and as such is a long lived species that has time to move to the bubble surface region [69]. If no O_2 is present there is formation of $\bullet\text{H}$ that produces H_2 and $\bullet\text{OH}$ recombines to give H_2O_2 , whereas in the presence of O_2 there are $\bullet\text{OH}$ and $\bullet\text{O}$ radicals and recombination will result in less H_2O_2 being produced [217, 224]. Reactions with O_2 include (13) – (15) [99, 167].

Akin to argon, there is also a frequency dependence for oxygen; at high frequencies more $\bullet\text{OH}$ radicals are available due to increased escape from the bubble, at low frequencies $\bullet\text{H}$ has time to react with O_2 which may produce the hydroperoxyl radical, as shown in reaction (16) [167].

Although CO_2 results in the formation of large, stable bubbles it is known to inhibit free radical production due to a reduction in active cavitation [150, 265]. CO_2 has been found to decrease the number of cavities in water by preventing their activation due to the acidic nature of products formed upon dissolution [265]. In ultra-pure water no measurable acoustic noise spectra was detected for CO_2 indicating no cavitation events, conversely for O_2 a signal was found to be present [209]. Since CO_2 is highly soluble, a greater concentration is able to dissolve into the liquid, this causes an increase in diffusion into the bubble during expansion. Since the bubble collapse is very rapid the large amount of gas inside the bubble is unable to escape at the same rate, this precipitates a cushioning effect, reducing the heat obtained and disassociation inside the bubble [266]. Returning to the idea of a change in pH influencing adsorption of species at the surface, the reduction in coalescence of bubbles in CO_2 atmosphere has been suggested due to change in surface charge at the bubbles surface and steric mechanisms [209]. This is intrinsic to weak acids, i.e. CO_2 upon dissolution changes the pH which causes charged species to move to the bubble surface [267]. Similarly, a coalescence influence

was observed for inert gas bubbles that were able to coalesce between pH 3 and 7, yet CO₂ preventing coalescence below pH 5 [254].

4.4.2. Gas Mixtures

For O₂/Ar gas mixtures the production rate of H₂O₂ increases with O₂, this occurs until the volume reaches 20% at which point H₂O₂ decreases with further increase [96]. However, this is entirely dependent on the system; Pflieger *et al.* found that (with mechanical stirring) intensive Ar/O₂ bubbling significantly improved H₂O₂ yields, maximised at 70-80% argon and 20-30% oxygen. They also realised a frequency dependence finding frequencies between 204 and 362 kHz were ten times more efficient for sonochemical production of H₂O₂ compared to 20 kHz [99]. Up to a fivefold increase (in comparison to solely Ar or O₂ atmospheres) in H₂O₂ production has similarly been observed at 300 kHz and 615 kHz [217, 224]. The maximum chemical reaction rate for 1,4-dioxane in was found at 358 kHz in mixed O₂/Ar gas atmospheres [69]. Similar ratios have been observed; Hart and Henglein investigated the degradation and formation of N₂O to N₂, O₂⁻, NO₂⁻ and NO₃⁻ respectively, finding a maximum rate to occur at 80-85% argon and 15-20% N₂O [222]. These observations may indicate that the amount of radical production and recombination rate during the process is favourable at these volumes ratios.

For large amounts of CO₂ there is inhibition of radical production, however small amounts in Ar, He, O₂, N₂ have been found to improve sonochemical rates [95, 221, 223, 268]. Sonochemical oxidation (of I⁻ ions) reduces significantly above 3% CO₂ in air, as does the sonochemical reduction of the carbon dioxide present which in atmospheres other than air follows the order Ar > He > H₂ > N₂ [223, 268]. Similarly, a maximum rate of CO formation in argon with 2- 3% CO₂ has been observed at 300 kHz [218]. Henglein in an earlier paper also considered that CO₂ amounts greater than 3% resulted in decreased sonochemical yields [221]. In another other study, it was concluded that the effect of small amounts of CO₂ in argon at 200 kHz and 100 W can lead to an increase I⁻ production up to seven fold [95]. These findings may be understood by considering CO₂ to be a radical scavenger, that is, if CO₂ quenches •H, then •OH will remain and oxidise the reactants. This is shown in reactions (17) and (18) which are those that occur under Ar in water, reactions (19) and (20) show how •OH contributes to oxidation while •H is consumed by the CO₂ [95].

As discussed, CO₂ may also reduce coalescence due to surface tension properties. Therefore through these observations, it may be inferred that the introduction of small amounts of CO₂ does not substantially act to prevent cavitation as would be seen with large amounts of the gas, furthermore that via both quenching and surface tension mechanisms small amounts of CO₂ can aid in increasing sonochemical oxidation of iodide. Further details of the aforementioned gas ratios and their effects can be found in Table 2.

4.4.3. Products from Gas-Liquid Reactions

The products resulting from sonication of water in the presence of gas and gas mixtures have potential for industrial implications; however there is little research in this area. The sonolysis of carbon dioxide, nitrous oxide and methane in aqueous solution was investigated by Henglein in 1985 [221].

At 300 kHz, 3.5 Watt/cm² and 20 °C it was shown there is a decrease in the concentration of hydrogen with increased mole fraction of carbon dioxide in an Ar-CO₂ atmosphere. Optimal formation (7×10^{-3} M) of hydrogen is shown at 0.08 mole fraction of methane in an Ar-CH₄ atmosphere and optimal formation (11.5×10^{-4} M) of hydrogen at 0.3 mole fraction of CO₂ in an Ar-CO₂ atmosphere.

Sonication of methane in water under an argon atmosphere brings about the products hydrogen, ethane, ethylene, C₃ and C₄- hydrocarbons and carbon monoxide. In addition, the concentration of gas dissolved in the solution directly influences the products and reaction rates [171]. If parameters can be manipulated to favour certain products, there is increased scope for industrial applications.

Due to the bubble interaction complexity of a multibubble system, replication via mathematical or computer modelling, is, at present, primarily limited to single or two-bubble systems. However, more recently, for a single bubble system, Merouani *et al.* produced a model to determine the mechanism for hydrogen production [111]. It was found that the formation of hydrogen was dominant in the gas phase with recombination at the bubble surface region playing only a minor role. In a consecutive study, with various frequencies (140 – 1100 kHz) and powers (0.75 and 1 W / cm²) a range of bubble sizes and an optimal bubble radius for the production of H₂, were determined [269]. As the pressure amplitude increased and frequency decreased, both optimal bubble size and the range of bubble sizes for the production of hydrogen increased. These results illustrate that frequency determined bubble sizes have a direct influence on the products formed.

The use of alternate solutions to water can also determine different products, as demonstrated by the sonication of aliphatic aldehydes and carboxylic acids under argon [270]. For numerous 0.006 M aldehyde and acid solutions, differing proportions of CO, CO₂, CH₄, C₂H₆, C₂H₄ and C₂H₂ were formed [270]. Multiple short chained hydrocarbons of the configuration C_xH_yO_z were also detected. Others have observed the formation of glycine and other amino acids in nitrogen [216, 271, 272]. At 355 kHz and 70 W, amino acid formation for nitrogen and separately nitrogen with hydrogen, both in aqueous solutions with argon atmosphere, followed the order alanine > ethylglycine > glycine. Methane was the exception where for both nitrogen and hydrogen the rate of production followed the order glycine > alanine > ethylglycine [216]. These observations demonstrate the effective utilisation of ultrasound in the formation of products; by controlling the conditions under which they form it may be possible to control production in discrete amounts.

4.5. Implications of Secondary Parameters

The use of chemical additives to change aqueous solution properties can be advantageous in promoting chemical oxidising reactions, through both chemical and / or physical effects. Solids represent a mechanism for catalysis of chemical reactivity and bubble collapse can enhance reactivity of oxides via surface melting and erosion. Surfactants can instigate an increase in mass transfer into the bubble and, in the correct ratios, have been suggested to reduce coalescence via charge and steric properties. With increased surface tension and viscosity, bubbles can be more strongly forced for increased disassociation yet it is very difficult to make bubbles oscillate strongly. Increasing the viscosity can also provide possible reduction of coalescence due to inhibition of surface drainage. Additionally, the formation of radical species inside the bubble may be increased, however the degree of fragmentation will be reduced. By controlling the pH of the solution the dynamics of introduced species can be controlled. This may have a direct effect on the formation of products, and has been suggested to influence coalescence. Electrolytes may also offer some control in regards to molecular dynamics and have been shown to directly affect reactant population in regions, surface tension gradients and coalescence. However, it should be realised that changes in the chemical composition of the liquid can directly influence the effects of several applied parameters and that this could have an overall negative impact on the degree of sonochemical activity.

Sonication in gas environments can have significant effects on the sonochemistry taking place. Argon represents a means of enhancing high temperature reactions, however at certain frequencies it may not be as efficient as air in facilitating sonochemical reactions. Oxygen can directly change the reaction pathways taking place the disassociated products react dominantly at the surface of the bubble. Carbon dioxide can also act to determine recombination pathways as well as potentially reducing coalescence via its dissolved surface properties. However these reactions are only useful if they deliver the required effect to the individual system's needs. Furthermore, dissolved gases, such as carbon dioxide, previously discussed, have a direct influence on the surface tension of the solution influencing the cavitation threshold. An increase in surface tension is known to result in a more structurally stable bubble therefore gases that decrease surface tension may be beneficial, although it should be considered that a lower surface tension can influence coalescence as it indicates adsorption of surface active species. The introduction of parameters that decrease coalescence effects should be implemented for low surface tension inducing gases. Also, the frequency dependence of gas atmospheres in precipitating certain reaction pathways can be applied to product control. Argon and oxygen are particularly susceptible to changes in frequency and there is little to suggest other gases would not be affected in a similar manner.

In terms of structural stability of the bubbles, the presence of extended surfaces ($>150\text{ }\mu\text{m}$) and the use of powders in the solution have demonstrated an ability to induce bubble surface instabilities. The formation of jets for the former and the inter-particle collisions for the later cause the bubbles to become non-spherical and could potentially contribute favourably to bubble fragmentation. Less studied are chemical effects, however surfactants proffer a strong mechanism for inducing bubble surface instabilities via their intrinsic charge and dimension. The overall effect of increased viscosity and surface tension is to result in a more structurally stable bubble, thus it may be inferred that lowering their values could instigate surface instabilities. Electrolytes in reducing the gas content of the liquid may offer a method of increasing the magnitude of collapse and in doing so promote bubble instabilities. Although these two concepts have yet to be directly related in the literature, it is suggested that powders, chemicals and gases that induce chemical changes represent a niche in the field particularly with regards to bubble structural instability and sonochemical efficiency.

5. Discussion

An intrinsic problem of many parameters that increase bubble structural instability is coalescence that, at lower frequencies, can potentially lower the number of active bubbles and overall sonochemical activity. In this instance, it may be of use to combine parameters with a view to reduce coalescence and increasing effects of instability inducing parameters.

Mid to high pressure amplitudes have been considered to be favourable in achieving bubble structural instability due to a higher forcing collapse. Not only this, but the resulting acoustic streaming can cause translational motion of the bubble that can facilitate further nonlinearity. However, due to agglomeration and an increase in the field strength there may be inherent coalescence that is beyond prevention via intrinsic streaming. Similarly, the potential for bubbles to coalesce may be increased with bubble size due to an increase in the secondary Bjerknes force. The introduction of external flow could remove a favourable number of bubbles from the antinode region such that the remaining bubbles experience less coalescence to non-active size. Additionally, flow can be implemented such that there is an increase in bubble surface instabilities. Quasi acoustic streaming with increased frequency can be implemented in a similar vein to flow, although its rate is determinant upon the frequency and power used. This type of streaming can cause bubbles to move to the surface and produce sonochemistry in standing waves in that region. In this instance, via travelling waves, sonochemical activity has been seen to be significantly increased. Sonochemical activity may be

achieved in the standing wave pressure antinodes by establishing an effective pressure amplitude-flow ratio.

An alternative method to the use of flow to decrease levels of coalescence is the use of surfactants. By choosing a surfactant with the correct charge-size properties, coalescence and bubble surface instabilities can be controlled. Dissolved CO₂ species can also act as surfactants, dependent on pH due to the acid-base equilibria of the carbonates. The products of CO₂ dissolution have been suggested to reduce coalescence via both electrostatic and steric repulsion. A decrease in surface tension via CO₂, increase in temperature or introduction of other chemicals, will result in a less structurally stable bubble, although it may also lead to an increase in coalescence. However surfactants have limited surface effects when the lifetime of the bubble is too short. This could be counteracted using pulsed ultrasound with its ability to increase a bubble's active lifetime. That is, P_{OFF}T may be applied such that there is a reduction in bubble growth beyond inactivity and coalescence. Additionally, the start of each pulse is a source of more violent pulsations that could also contribute to bubble surface instabilities and potentially increase the degree of fragmentation.

Bubble instability will also have an effect on the chemistry within the bubble and the bulk solution. For surface unstable bubbles there will be increased growth rates and enhanced diffusion into the bubble. An increase in fragmentation due to instability can also shorten the amount of time reactants spend inside the bubble and reduce reaction pathways. However the increase in fragmentation will also increase the dissolution of sonochemical products into the bulk solution. Additionally, where chemicals are introduced into the system to control physical aspects in a chemical system, such as surfactants, the interactions between those chemicals and the radicals present should be also considered.

An increase in viscosity of the solution via temperature change or by the introduction of viscous fluids may be an option to reduce coalescence. However, this may introduce both attenuation of the sound field or a change in the chemical reaction scheme (depending on the solvent). Similarly, electrolyte solutions have been suggested to reduce film drainage between bubbles due to uneven concentration gradients at the bubbles' surfaces. Adsorption at the surface can also contribute to steric repulsions. An associated effect of coalescence is the formation of larger bubbles and clustering that attenuates the sound field. In this case a reflective plate and selection of the appropriate liquid height can act to reinforce the standing wave.

Thus, the formation of degas bubbles from coalescence can be controlled while allowing a determinable amount of active cavitation to occur at the antinodes that undergo heightened surface instabilities. It is suggested that methods of localising bubbles in specific regions whilst subjecting them to bubble surface instability inducing parameters is highly beneficial to sonochemical reactions. The bubbles, in this instance, can be said to exist in a state of 'highly localised ordered disorder'.

6. Conclusions

A sonochemical system configured through bubble structural instability offers a method to increase or control sonochemical activity. Several authors indirectly or directly have supported this idea, although it was not the primary focus of their work. Increases in sonochemical efficiency in various ratios of mixed gas atmosphere and via changes in the chemical and/or physical environment have been reviewed. Through these parameters' effects, the literature suggests that the sonochemical system may be controlled with respect to efficiency and product selectivity. It is proposed that bubble surface instabilities represent a mechanism to augment this concept.

References

- [1] J. Krautkrämer, H. Krautkrämer, *Ultrasonic testing of materials*, Springer Science & Business Media, 2013.
- [2] E. Statnikov, V. Muktepavel, A. Blomqvist, Comparison of ultrasonic impact treatment (UIT) and other fatigue life improvement methods, *Weld. World*, 46 (2002) 20-32.
- [3] J.A. McKnight, L.M. Barrett, Ultrasonic range finding, in, Google Patents, 1989.
- [4] M. Sonnenschein, D. Wulich, E. Sonnenschein, Underwater communication apparatus and communication method, in, Google Patents, 2000.
- [5] A. Fenster, D.B. Downey, 3-D ultrasound imaging: A review, *Engineering in Medicine and Biology Magazine, IEEE*, 15 (1996) 41-51.
- [6] H. Wickramasinghe, Scanning acoustic microscopy: a review, *J. Microsc. (Oxford)*, 129 (1983) 63-73.
- [7] K.G. Baker, V.J. Robertson, F.A. Duck, A review of therapeutic ultrasound: biophysical effects, *Phys. Ther.*, 81 (2001) 1351-1358.
- [8] R.S. Cobbold, *Foundations of biomedical ultrasound*, Oxford University Press on Demand, 2007.
- [9] R.E. Cartee, B.A. Selcer, J.A. Hudson, S.T. Finn-Bodner, M.B. Mahaffey, P.L. Johnson, K. Marich, *Practical veterinary ultrasound*, Williams & Wilkins, 1996.
- [10] A. Henglein, Sonochemistry: historical developments and modern aspects, *Ultrasonics*, 25 (1987) 6-16.
- [11] A. Gedanken, Using sonochemistry for the fabrication of nanomaterials, *Ultrason. Sonochem.*, 11 (2004) 47-55.
- [12] T. Awad, H. Moharram, O. Shaltout, D. Asker, M. Youssef, Applications of ultrasound in analysis, processing and quality control of food: A review, *Food Res. Int.*, 48 (2012) 410-427.
- [13] G. Ruecroft, D. Hipkiss, T. Ly, N. Maxted, P.W. Cains, Sonocrystallization: the use of ultrasound for improved industrial crystallization, *Organic Process Research & Development*, 9 (2005) 923-932.
- [14] A. Shoh, Industrial applications of ultrasound - A review I. High-power ultrasound, *Sonics and Ultrasonics, IEEE Transactions on*, 22 (1975) 60-70.
- [15] Q. Zhou, J. Zheng, S. Onishi, M. Crommie, A.K. Zettl, Graphene electrostatic microphone and ultrasonic radio, *Proceedings of the National Academy of Sciences*, 112 (2015) 8942-8946.
- [16] K.S. Suslick, Sonoluminescence and sonochemistry, in: *Ultrasonics Symposium*, 1997. *Proceedings.*, 1997 IEEE, IEEE, 1997, pp. 523-532.
- [17] N. Serpone, P. Colarusso, Sonochemistry I. Effects of ultrasounds on heterogeneous chemical reactions—a useful tool to generate radicals and to examine reaction mechanisms, *Res. Chem. Intermed.*, 20 (1994) 635-679.
- [18] A. Henglein, C. Kormann, Scavenging of OH radicals produced in the sonolysis of water, *Int. J. Radiat. Biol.*, 48 (1985) 251-258.
- [19] K.S. Suslick, The chemistry of ultrasound, *Encyclopaedia Britannica*, Chicago, (1994) 138-155.
- [20] S. Nam, S. Han, J. Kang, H. Choi, Kinetics and mechanisms of the sonolytic destruction of non-volatile organic compounds: investigation of the sonochemical reaction zone using several OH monitoring techniques, *Ultrason. Sonochem.*, 10 (2003) 139-147.
- [21] M. Klavarioti, D. Mantzavinos, D. Kassinos, Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes, *Environ. Int.*, 35 (2009) 402-417.
- [22] K.S. Suslick, Y. Didenko, M.M. Fang, T. Hyeon, K.J. Kolbeck, W.B. McNamara, M.M. Mdleleni, M. Wong, Acoustic cavitation and its chemical consequences, *Phil. Trans. R. Soc. A*, 357 (1999) 335-353.
- [23] A. Henglein, Contributions to various aspects of cavitation chemistry, *Advances in sonochemistry*, 3 (1993) 17-83.
- [24] J. Löning, C. Horst, U. Hoffmann, Investigations on the energy conversion in sonochemical processes, *Ultrason. Sonochem.*, 9 (2002) 169-179.
- [25] T. Leighton, *The acoustic bubble*, Academic press, 2012.
- [26] R.F. Young, *Cavitation*, McGraw-Hill, 1989.

- [27] L. Crum, Acoustic cavitation series: part five rectified diffusion, *Ultrasonics*, 22 (1984) 215-223.
- [28] H. Flynn, Physics of acoustic cavitation in liquids, *Phys. Acoustics*, 1 (1964) 57-172.
- [29] T.J. Matula, S.M. Cordry, R.A. Roy, L.A. Crum, Bjerknes force and bubble levitation under single-bubble sonoluminescence conditions, *J. Acoust. Soc. Am.*, 102 (1997) 1522-1527.
- [30] H. Mitome, Action of ultrasound on particles and cavitation bubbles, in: *Proc. World Congress on Ultrasonics*, Paris, 2003, pp. 2342-2346.
- [31] M. Ashokkumar, J. Lee, S. Kentish, F. Grieser, Bubbles in an acoustic field: an overview, *Ultrason. Sonochem.*, 14 (2007) 470-475.
- [32] K. Yasui, Influence of ultrasonic frequency on multibubble sonoluminescence, *J. Acoust. Soc. Am.*, 112 (2002) 1405-1413.
- [33] L.A. Crum, Bjerknes forces on bubbles in a stationary sound field, *J. Acoust. Soc. Am.*, 57 (1975) 1363-1370.
- [34] P.C. Duineveld, The influence of an applied sound field on bubble coalescence, *J. Acoust. Soc. Am.*, 99 (1996) 622-624.
- [35] R. Kirkpatrick, M. Lockett, The influence of approach velocity on bubble coalescence, *Chem. Eng. Sci.*, 29 (1974) 2363-2373.
- [36] G. Jeffreys, J. Hawksley, Coalescence of liquid droplets in 2-component-2-phase systems. Part 2. Theoretical analysis of coalescence rate., *AIChE J.*, 11 (1965) 413.
- [37] J. Jiao, Y. He, S.E. Kentish, M. Ashokkumar, R. Manasseh, J. Lee, Experimental and theoretical analysis of secondary Bjerknes forces between two bubbles in a standing wave, *Ultrasonics*, 58 (2015) 35-42.
- [38] J. Lee, S.E. Kentish, M. Ashokkumar, The effect of surface-active solutes on bubble coalescence in the presence of ultrasound, *J. Phys. Chem. B*, 109 (2005) 5095-5099.
- [39] J. Jiao, Y. He, K. Yasui, S.E. Kentish, M. Ashokkumar, R. Manasseh, J. Lee, Influence of acoustic pressure and bubble sizes on the coalescence of two contacting bubbles in an acoustic field, *Ultrason. Sonochem.*, 22 (2015) 70-77.
- [40] T.O. Oolman, H.W. Blanch, Bubble coalescence in stagnant liquids, *Chem. Eng. Commun.*, 43 (1986) 237-261.
- [41] P. Murdoch, D. Leng, The mathematical formulation of hydrodynamic film thinning and its application to colliding drops suspended in a second liquid-II, *Chem. Eng. Sci.*, 26 (1971) 1881-1892.
- [42] M.J. Bussemaker, D. Zhang, A phenomenological investigation into the opposing effects of fluid flow on sonochemical activity at different frequency and power settings. 1. Overhead stirring, *Ultrason. Sonochem.*, 21 (2014) 436-445.
- [43] M. Ashokkumar, The characterization of acoustic cavitation bubbles—an overview, *Ultrason. Sonochem.*, 18 (2011) 864-872.
- [44] M. Ashokkumar, J. Lee, Y. Iida, K. Yasui, T. Kozuka, T. Tuziuti, A. Towata, The detection and control of stable and transient acoustic cavitation bubbles, *Phys. Chem. Chem. Phys.*, 11 (2009) 10118-10121.
- [45] J. Wu, W.L. Nyborg, Ultrasound, cavitation bubbles and their interaction with cells, *Adv. Drug Deliv. Rev.*, 60 (2008) 1103-1116.
- [46] T. Leighton, Bubble population phenomena in acoustic cavitation, *Ultrason. Sonochem.*, 2 (1995) S123-S136.
- [47] B. Gielen, J. Jordens, J. Janssen, H. Pfeiffer, M. Wevers, L. Thomassen, L. Braeken, T. Van Gerven, Characterization of stable and transient cavitation bubbles in a milliflow reactor using a multibubble sonoluminescence quenching technique, *Ultrason. Sonochem.*, 25 (2015) 31-39.
- [48] R. Tronson, M. Ashokkumar, F. Grieser, Comparison of the effects of water-soluble solutes on multibubble sonoluminescence generated in aqueous solutions by 20-and 515-kHz pulsed ultrasound, *J. Phys. Chem. B*, 106 (2002) 11064-11068.
- [49] T. Leighton, The principles of cavitation, M. J. W. Povey, & T. J. Mason (Eds.) ed., London: Blackie Academic & Professional, 1995.

- [50] D.F. Gaitan, L.A. Crum, C.C. Church, R.A. Roy, Sonoluminescence and bubble dynamics for a single, stable, cavitation bubble, *J. Acoust. Soc. Am.*, 91 (1992) 3166-3183.
- [51] S. Hatanaka, H. Mitome, K. Yasui, S. Hayashi, Single-bubble sonochemiluminescence in aqueous luminol solutions, *J. Am. Chem. Soc.*, 124 (2002) 10250-10251.
- [52] D. Lohse, S. Hilgenfeldt, Inert gas accumulation in sonoluminescing bubbles, *J. Chem. Phys.*, 107 (1997) 6986-6997.
- [53] E. Neppiras, Acoustic cavitation thresholds and cyclic processes, *Ultrasonics*, 18 (1980) 201-209.
- [54] M. Plesset, On the stability of fluid flows with spherical symmetry, *J. Appl. Phys.*, 25 (1954) 96-98.
- [55] M. Strasberg, Gas bubbles as sources of sound in liquids, *J. Acoust. Soc. Am.*, 28 (1956) 20-26.
- [56] E. Neppiras, Subharmonic and Other Low-Frequency Emission from Bubbles in Sound-Irradiated Liquids, *J. Acoust. Soc. Am.*, 46 (1969) 587-601.
- [57] A.I. Eller, L.A. Crum, Instability of the motion of a pulsating bubble in a sound field, *J. Acoust. Soc. Am.*, 47 (1970) 762-767.
- [58] A. Prosperetti, Viscous effects on perturbed spherical flows, *Q. Appl. Math.*, (1977) 339-352.
- [59] S.J. Shaw, Translation and oscillation of a bubble under axisymmetric deformation, *Phys. Fluids* (1994-present), 18 (2006) 072104.
- [60] S. Shaw, The stability of a bubble in a weakly viscous liquid subject to an acoustic traveling wave, *Phys. Fluids* (1994-present), 21 (2009) 022104.
- [61] R.K. Gould, Rectified diffusion in the presence of, and absence of, acoustic streaming, *J. Acoust. Soc. Am.*, 56 (1974) 1740-1746.
- [62] B.D. Storey, A.J. Szeri, Water vapour, sonoluminescence and sonochemistry, in: *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, The Royal Society, 2000, pp. 1685-1709.
- [63] S. Hatanaka, Sonoluminescence, sonochemistry and bubble dynamics of single bubble cavitation, in: *Nonlinear Acoustics State-of-the-Art and Perspectives: 19th International Symposium on Nonlinear Acoustics*, AIP Publishing, Tokyo, Japan, 2012, pp. 163-166.
- [64] M. Ashokkumar, P. Mulvaney, F. Grieser, The effect of pH on multibubble sonoluminescence from aqueous solutions containing simple organic weak acids and bases, *J. Am. Chem. Soc.*, 121 (1999) 7355-7359.
- [65] T. Leighton, Transient excitation of insonated bubbles, *Ultrasonics*, 27 (1989) 50-53.
- [66] M.P. Brenner, S. Hilgenfeldt, D. Lohse, Single-bubble sonoluminescence, *Rev. Mod. Phys.*, 74 (2002) 425.
- [67] S. Putterman, Sonoluminescence: The Star In A Jar, *Phys. World*, (1998).
- [68] G. Vazquez, C. Camara, S. Putterman, K. Weninger, Sonoluminescence: nature's smallest blackbody, *Opt. Lett.*, 26 (2001) 575-577.
- [69] M.A. Beckett, I. Hua, Impact of ultrasonic frequency on aqueous sonoluminescence and sonochemistry, *J. Phys. Chem. A*, 105 (2001) 3796-3802.
- [70] K.R. Weninger, B. Barber, S. Putterman, Pulsed Mie scattering measurements of the collapse of a sonoluminescing bubble, *Phys. Rev. Lett.*, 78 (1997) 1799.
- [71] V. Misik, N. Miyoshi, P. Riesz, EPR spin-trapping study of the sonolysis of H₂O/D₂O mixtures: probing the temperatures of cavitation regions, *J. Phys. Chem. (US)*, 99 (1995) 3605-3611.
- [72] V. Mišík, P. Riesz, EPR study of free radicals induced by ultrasound in organic liquids II. Probing the temperatures of cavitation regions, *Ultrason. Sonochem.*, 3 (1996) 25-37.
- [73] K.S. Suslick, D.A. Hammerton, R.E. Cline Jr, The sonochemical hot spot, *J. Acoust. Soc. Am.*, 89 (1991) 1885-1886.
- [74] E.J. Hart, C. Fischer, A. Henglein, Sonolysis of hydrocarbons in aqueous solution, *Int. J. Radiat. Appl. Instrum. C Radiat. Phys. Chem.*, 36 (1990) 511-516.
- [75] C. Sonntag, Sonolysis of tert-butyl alcohol in aqueous solution, *J. Chem. Soc. Perkin. Trans. 2*, (1999) 1129-1136.
- [76] E.B. Flint, K.S. Suslick, The temperature of cavitation, *Science*, 253 (1991) 1397-1399.

- [77] M. Ashokkumar, F. Grieser, A comparison between multibubble sonoluminescence intensity and the temperature within cavitation bubbles, *J. Am. Chem. Soc.*, 127 (2005) 5326-5327.
- [78] F.R. Young, Sonoluminescence from water containing dissolved gases, *J. Acoust. Soc. Am.*, 60 (1976) 100-104.
- [79] B. Gompf, R. Günther, G. Nick, R. Pecha, W. Eisenmenger, Resolving sonoluminescence pulse width with time-correlated single photon counting, *Phys. Rev. Lett.*, 79 (1997) 1405.
- [80] S.I. Nikitenko, R. Pflieger, Toward a new paradigm for sonochemistry: Short review on nonequilibrium plasma observations by means of MBSL spectroscopy in aqueous solutions, *Ultrason. Sonochem.*, 35B (2017) 623-630.
- [81] A.A. Ndiaye, R. Pflieger, B. Siboulet, J. Molina, J.F. Dufrêche, S.I. Nikitenko, Nonequilibrium vibrational excitation of OH radicals generated during multibubble cavitation in water, *J. Phys. Chem. A*, 116 (2012) 4860-4867.
- [82] S.I. Nikitenko, Plasma Formation during Acoustic Cavitation: Toward a New Paradigm for Sonochemistry, *Adv. Phys. Chem.*, (2014).
- [83] M.R. Hoffmann, I. Hua, R. Höchmer, Application of ultrasonic irradiation for the degradation of chemical contaminants in water, *Ultrason. Sonochem.*, 3 (1996) S163-S172.
- [84] S. Merouani, H. Ferkous, O. Hamdaoui, Y. Rezgui, M. Guemini, New interpretation of the effects of argon-saturating gas toward sonochemical reactions, *Ultrason. Sonochem.*, 23 (2015) 37-45.
- [85] H. Xu, K.S. Suslick, Molecular emission and temperature measurements from single-bubble sonoluminescence, *Phys. Rev. Lett.*, 104 (2010) 244301.
- [86] W.B. McNamara, Y.T. Didenko, K.S. Suslick, Pressure during sonoluminescence, *J. Phys. Chem. B*, 107 (2003) 7303-7306.
- [87] H. Flynn, Generation of transient cavities in liquids by microsecond pulses of ultrasound, *J. Acoust. Soc. Am.*, 72 (1982) 1926-1932.
- [88] K. Makino, M.M. Mossoba, P. Riesz, Chemical effects of ultrasound on aqueous solutions. Formation of hydroxyl radicals and hydrogen atoms, *J. Phys. Chem. (US)*, 87 (1983) 1369-1377.
- [89] C.M. Krishna, T. Kondo, P. Riesz, Sonochemistry of alcohol-water mixtures. Spin-trapping evidence for thermal decomposition and isotope-exchange reactions, *J. Phys. Chem. (US)*, 93 (1989) 5166-5172.
- [90] D. Sunartio, M. Ashokkumar, F. Grieser, Study of the coalescence of acoustic bubbles as a function of frequency, power, and water-soluble additives, *J. Am. Chem. Soc.*, 129 (2007) 6031-6036.
- [91] E.L. Mead, R.G. Sutherland, R.E. Verrall, The effect of ultrasound on water in the presence of dissolved gases, *Can. J. Chem.*, 54 (1976) 1114-1120.
- [92] Y.T. Shah, A. Pandit, V. Moholkar, Cavitation reaction engineering, Springer Science & Business Media, 2012.
- [93] A. Brothie, J. Schneider, R. Pflieger, D. Shchukin, H. Möhwald, Sonochemiluminescence from a Single Cavitation Bubble in Water, *Chem. Eur. J.*, 18 (2012) 11201-11204.
- [94] T.J. Mason, *Advances in sonochemistry*, AI Press Inc. , 1996.
- [95] H. Harada, Y. Ono, Improvement of the rate of sono-oxidation in the presence of CO₂, *Jpn. J. Appl. Phys.*, 54 (2015).
- [96] C. Gong, D.P. Hart, Ultrasound induced cavitation and sonochemical yields, *J. Acoust. Soc. Am.*, 104 (1998) 2675-2682.
- [97] B. Nanzai, K. Okitsu, N. Takenaka, H. Bandow, N. Tajima, Y. Maeda, Effect of reaction vessel diameter on sonochemical efficiency and cavitation dynamics, *Ultrason. Sonochem.*, 16 (2009) 163-168.
- [98] K. Yasui, T. Tuziuti, Y. Iida, Optimum bubble temperature for the sonochemical production of oxidants, *Ultrasonics*, 42 (2004) 579-584.
- [99] R. Pflieger, T. Chave, G. Vite, L. Jouve, S.I. Nikitenko, Effect of operational conditions on sonoluminescence and kinetics of H₂O₂ formation during the sonolysis of water in the presence of Ar/O₂ gas mixture, *Ultrason. Sonochem.*, 26 (2015) 169-175.

- [100] T.J. Mason, D. Peters, Practical sonochemistry: Power ultrasound uses and applications, Woodhead Publishing, 2002.
- [101] K. Yasui, T. Tuziuti, T. Kozuka, A. Towata, Y. Iida, Relationship between the bubble temperature and main oxidant created inside an air bubble under ultrasound, *J. Chem. Phys.*, 127 (2007) 154502-154502.
- [102] P. Riesz, T. Kondo, C.M. Krishna, Sonochemistry of volatile and non-volatile solutes in aqueous solutions: epr and spin trapping studies, *Ultrasonics*, 28 (1990) 295-303.
- [103] K. Suslick, S. Doktycz, E. Flint, On the origin of sonoluminescence and sonochemistry, *Ultrasonics*, 28 (1990) 280-290.
- [104] B. Yim, H. Okuno, Y. Nagata, R. Nishimura, Y. Maeda, Sonolysis of surfactants in aqueous solutions: an accumulation of solute in the interfacial region of the cavitation bubbles, *Ultrason. Sonochem.*, 9 (2002) 209-213.
- [105] K. Okitsu, K. Iwasaki, Y. Yobiko, H. Bandow, R. Nishimura, Y. Maeda, Sonochemical degradation of azo dyes in aqueous solution: a new heterogeneous kinetics model taking into account the local concentration of OH radicals and azo dyes, *Ultrason. Sonochem.*, 12 (2005) 255-262.
- [106] S. Merouani, H. Ferkous, O. Hamdaoui, Y. Rezgui, M. Guemini, A method for predicting the number of active bubbles in sonochemical reactors, *Ultrason. Sonochem.*, 22 (2015) 51-58.
- [107] M.H. Uddin, B. Nanzai, K. Okitsu, Effects of Na₂SO₄ or NaCl on sonochemical degradation of phenolic compounds in an aqueous solution under Ar: Positive and negative effects induced by the presence of salts, *Ultrason. Sonochem.*, 28 (2016) 144-149.
- [108] C.H. Fischer, E.J. Hart, A. Henglein, Ultrasonic irradiation of water in the presence of oxygen 18, 18O₂: isotope exchange and isotopic distribution of hydrogen peroxide, *J. Phys. Chem. (US)*, 90 (1986) 1954-1956.
- [109] M. Gutierrez, A. Henglein, J.K. Dohrmann, Hydrogen atom reactions in the sonolysis of aqueous solutions, *J. Phys. Chem.*, 91 (1987) 6687-6690.
- [110] S. Nikitenko, L. Venault, P. Moisy, Scavenging of OH radicals produced from H₂O sonolysis with nitrate ions, *Ultrason. Sonochem.*, 11 (2004) 139-142.
- [111] S. Merouani, O. Hamdaoui, Y. Rezgui, M. Guemini, Mechanism of the sonochemical production of hydrogen, *Int. J. Hydrogen Energy*, 40 (2015) 4056-4064.
- [112] K. Suslick, The site of sonochemical reactions, *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, 33 (1986) 143-147.
- [113] A. Henglein, M. Gutierrez, Sonolysis of polymers in aqueous solution. New observations on pyrolysis and mechanical degradation, *J. Phys. Chem. (US)*, 92 (1988) 3705-3707.
- [114] L.A. Crum, T.J. Mason, J.L. Reisse, K.S. Suslick, *Sonochemistry and sonoluminescence*, Springer Science & Business Media, 2013.
- [115] Y.T. Didenko, W.B. McNamara III, K.S. Suslick, Molecular emission from single-bubble sonoluminescence, *Nature*, 407 (2000) 877-879.
- [116] M.J. Bussemaker, D. Zhang, A phenomenological investigation into the opposing effects of fluid flow on sonochemical activity at different frequency and power settings. 2. Fluid circulation at high frequencies, *Ultrason. Sonochem.*, 21 (2014) 485-492.
- [117] A. Henglein, R. Ulrich, J. Lilie, Luminescence and chemical action by pulsed ultrasound, *J. Am. Chem. Soc.*, 111 (1989) 1974-1979.
- [118] K. Weninger, S.J. Putterman, B.P. Barber, Angular correlations in sonoluminescence: Diagnostic for the sphericity of a collapsing bubble, *Phys. Rev. E*, 54 (1996) R2205.
- [119] N. Harba, S. Hayashi, A glycerol drop in water boosts single-bubble sonoluminescence, *Jpn. J. Appl. Phys.*, 42 (2003) 716.
- [120] S.H. Naser H, Single Bubble Sonoluminescence in Inhomogeneous Medium, PACS reference: 43.35.HI Japan, (2008).
- [121] P. Kanthale, M. Ashokkumar, F. Grieser, Sonoluminescence, sonochemistry (H₂O₂ yield) and bubble dynamics: frequency and power effects, *Ultrason. Sonochem.*, 15 (2008) 143-150.

- [122] T.J. Matula, Inertial cavitation and single-bubble sonoluminescence, *Phil. Trans. R. Soc. A.*, 357 (1999) 225-249.
- [123] L. Stricker, Acoustic cavitation and sonochemistry, Universiteit Twente, 2013.
- [124] N. de Jong, L. Hoff, T. Skotland, N. Bom, Absorption and scatter of encapsulated gas filled microspheres: theoretical considerations and some measurements, *Ultrasonics*, 30 (1992) 95-103.
- [125] N. De Jong, R. Cornet, C. Lancee, Higher harmonics of vibrating gas-filled microspheres. Part one: simulations, *Ultrasonics*, 32 (1994) 447-453.
- [126] K. Wei, D.M. Skyba, C. Firschke, A.R. Jayaweera, J.R. Lindner, S. Kaul, Interactions between microbubbles and ultrasound: in vitro and in vivo observations, *J. Am. Coll. Cardiol.*, 29 (1997) 1081-1088.
- [127] K. Yasui, Fundamentals of acoustic cavitation and sonochemistry, in: *Theoretical and Experimental Sonochemistry Involving Inorganic Systems*, Springer, 2010, pp. 1-29.
- [128] M.L. Calvisi, O. Lindau, J.R. Blake, A.J. Szeri, Shape stability and violent collapse of microbubbles in acoustic traveling waves, *Phys. Fluids* (1994-present), 19 (2007) 047101.
- [129] A.A. Doinikov, Translational motion of two interacting bubbles in a strong acoustic field, *Phys. Rev. E*, 64 (2001) 026301.
- [130] A. Harkin, T.J. Kaper, A. Nadim, Coupled pulsation and translation of two gas bubbles in a liquid, *J. Fluid Mech.*, 445 (2001) 377-411.
- [131] N.A. Pelekasis, A. Gaki, A. Doinikov, J.A. Tsamopoulos, Secondary Bjerknes forces between two bubbles and the phenomenon of acoustic streamers, *J. Fluid Mech.*, 500 (2004) 313-347.
- [132] K. Yoshida, T. Fujikawa, Y. Watanabe, Experimental investigation on reversal of secondary Bjerknes force between two bubbles in ultrasonic standing wave, *J. Acoust. Soc. Am.*, 130 (2011) 135-144.
- [133] S. Hatanaka, K. Yasui, T. Tuziuti, T. Kozuka, H. Mitome, Quenching mechanism of multibubble sonoluminescence at excessive sound pressure, *Jpn. J. Appl. Phys.*, 40 (2001) 3856.
- [134] J. Jiao, Y. He, T. Leong, S.E. Kentish, M. Ashokkumar, R. Manasseh, J. Lee, Experimental and theoretical studies on the movements of two bubbles in an acoustic standing wave field, *J. Phys. Chem. B*, 117 (2013) 12549-12555.
- [135] H. Mitome, T. Kozuka, T. Tuziuti, L. Wang, Quasi acoustic streaming induced by generation of cavitation bubbles, in: *International Ultrasonics Symposium, IEEE. Proceedings 1997.*, IEEE, 1997, pp. 533-536.
- [136] A. Kumar, P.R. Gogate, A.B. Pandit, Mapping of acoustic streaming in sonochemical reactors, *Ind. Eng. Chem. Res.*, 46 (2007) 4368-4373.
- [137] L.K. Weavers, M.R. Hoffmann, Sonolytic decomposition of ozone in aqueous solution: mass transfer effects, *Environ. Sci. Technol.*, 32 (1998) 3941-3947.
- [138] B. Dubus, C. Vanhille, C. Campos-Pozuelo, C. Granger, On the physical origin of conical bubble structure under an ultrasonic horn, *Ultrason. Sonochem.*, 17 (2010) 810-818.
- [139] L. Zarembo, Acoustic streaming, in: *High-intensity ultrasonic fields*, Springer, 1971, pp. 135-199.
- [140] A.J. Szeri, B.D. Storey, A. Pearson, J.R. Blake, Heat and mass transfer during the violent collapse of nonspherical bubbles, *Phys. Fluids* (1994-present), 15 (2003) 2576-2586.
- [141] T. Leong, J. Collis, R. Manasseh, A. Ooi, A. Novell, A. Bouakaz, M. Ashokkumar, S. Kentish, The role of surfactant headgroup, chain length, and cavitation microstreaming on the growth of bubbles by rectified diffusion, *J. Phys. Chem. C*, 115 (2011) 24310-24316.
- [142] J. Lighthill, Acoustic streaming, *J. Sound Vibration*, 61 (1978) 391-418.
- [143] P. Saffman, The self-propulsion of a deformable body in a perfect fluid, *J. Fluid Mech.*, 28 (1967) 385-389.
- [144] Z. Feng, L. Leal, Translational instability of a bubble undergoing shape oscillations, *Phys. Fluids* (1994-present), 7 (1995) 1325-1336.
- [145] H. Mitome, The mechanism of generation of acoustic streaming, *Electron. Commun. Jpn. (Part III: Fundamental Electronic Science)*, 81 (1998) 1-8.

- [146] T.B. Benjamin, A.T. Ellis, The collapse of cavitation bubbles and the pressures thereby produced against solid boundaries, *Phil. Trans. R. Soc. A.*, 260 (1966) 221-240.
- [147] K.S. Suslick, G.J. Price, Applications of ultrasound to materials chemistry, *Annu. Rev. Mater. Sci.*, 29 (1999) 295-326.
- [148] N. Bourne, J. Field, Bubble collapse and the initiation of explosion, in: *Proc. R. Soc. A, The Royal Society*, 1991, pp. 423-435.
- [149] Y. Kojima, Y. Asakura, G. Sugiyama, S. Koda, The effects of acoustic flow and mechanical flow on the sonochemical efficiency in a rectangular sonochemical reactor, *Ultrason. Sonochem.*, 17 (2010) 978-984.
- [150] J. Rooze, E.V. Rebrov, J.C. Schouten, J.T. Keurentjes, Effect of resonance frequency, power input, and saturation gas type on the oxidation efficiency of an ultrasound horn, *Ultrason. Sonochem.*, 18 (2011) 209-215.
- [151] J.-L. Laborde, C. Bouyer, J.-P. Caltagirone, A. Gérard, Acoustic cavitation field prediction at low and high frequency ultrasounds, *Ultrasonics*, 36 (1998) 581-587.
- [152] A. Brothie, F. Grieser, M. Ashokkumar, Effect of power and frequency on bubble-size distributions in acoustic cavitation, *Phys. Rev. Lett.*, 102 (2009) 084302.
- [153] K. Thangavadeivel, K. Okitsu, G. Owens, P.J. Lesniewski, R. Nishimura, Influence of sonochemical reactor diameter and liquid height on methyl orange degradation under 200kHz indirect sonication, *J. Environ. Chem. Eng.*, 1 (2013) 275-280.
- [154] R. Esche, *Akustische Beihefte*, 2AB 202 (1952).
- [155] J. Sponer, Dependence of the cavitation threshold on the ultrasonic frequency, *Czech. J. Phys.*, 40 (1990) 1123-1132.
- [156] M. Zhou, N.S.M. Yusof, M. Ashokkumar, Correlation between sonochemistry and sonoluminescence at various frequencies, *RSC Advan.*, 3 (2013) 9319-9324.
- [157] W.P. Mason, *Physical Acoustics: Principles and Methods. Properties of Polymers and Non-linear Acoustics. Vol. 2B*, Academic Press, 1965.
- [158] S. Hatanaka, H. Mitome, K. Yasui, S. Hayashi, Multibubble sonoluminescence enhancement by fluid flow, *Ultrasonics*, 44 (2006) e435-e438.
- [159] J. Lee, M. Ashokkumar, Importance of Sonication and Solution Conditions on the Acoustic Cavitation Activity. In: *Handbook of Ultrasonics and Sonochemistry*, Springer Science + Business Media Singapore, 2016.
- [160] R. Mettin, I. Akhatov, U. Parlitz, C. Ohi, W. Lauterborn, Bjerknes forces between small cavitation bubbles in a strong acoustic field, *Phys. Rev. E*, 56 (1997) 2924.
- [161] M. Postema, P. Marmottant, C.T. Lancée, S. Hilgenfeldt, N. de Jong, Ultrasound-induced microbubble coalescence, *Ultrasound Med. Biol.*, 30 (2004) 1337-1344.
- [162] A. Brothie, T. Statham, M. Zhou, L. Dharmarathne, F. Grieser, M. Ashokkumar, Acoustic bubble sizes, coalescence, and sonochemical activity in aqueous electrolyte solutions saturated with different gases, *Langmuir*, 26 (2010) 12690-12695.
- [163] J. Lee, M. Ashokkumar, K. Yasui, T. Tuziuti, T. Kozuka, A. Towata, Y. Iida, Development and optimization of acoustic bubble structures at high frequencies, *Ultrason. Sonochem.*, 18 (2011) 92-98.
- [164] J.D. Seymour, H.C. Wallace, R.B. Gupta, Sonochemical reactions at 640 kHz using an efficient reactor. Oxidation of potassium iodide, *Ultrason. Sonochem.*, 4 (1997) 289-293.
- [165] S.A. Reddy A J, Shape stability of unsteadily translating bubbles, *Phys. Fluids* (1994-present), 14 (2002).
- [166] T.J. Mason, J.P. Lorimer, *Applied sonochemistry*, Wiley-VCH Verlag GmbH & Co. KGaA, 2002.
- [167] C. Petrier, A. Jeunet, J.L. Luche, G. Reverdy, Unexpected frequency effects on the rate of oxidative processes induced by ultrasound, *J. Am. Chem. Soc.*, 114 (1992) 3148-3150.
- [168] S. Koda, T. Kimura, T. Kondo, H. Mitome, A standard method to calibrate sonochemical efficiency of an individual reaction system, *Ultrason. Sonochem.*, 10 (2003) 149-156.

- [169] M.H. Entezari, P. Kruus, Effect of frequency on sonochemical reactions. I: Oxidation of iodide, *Ultrason. Sonochem.*, 1 (1994) S75-S79.
- [170] J. Lee, M. Ashokkumar, S. Kentish, F. Grieser, Determination of the size distribution of sonoluminescence bubbles in a pulsed acoustic field, *J. Am. Chem. Soc.*, 127 (2005) 16810-16811.
- [171] A. Henglein, Chemical effects of continuous and pulsed ultrasound in aqueous solutions, *Ultrason. Sonochem.*, 2 (1995) S115-S121.
- [172] T. Tuziuti, K. Yasui, J. Lee, T. Kozuka, A. Towata, Y. Iida, Mechanism of enhancement of sonochemical-reaction efficiency by pulsed ultrasound, *J. Phys. Chem. A*, 112 (2008) 4875-4878.
- [173] L. Hallez, J. Lee, F. Touyeras, A. Nevers, M. Ashokkumar, J.-Y. Hihn, Enhancement and quenching of high-intensity focused ultrasound cavitation activity via short frequency sweep gaps, *Ultrason. Sonochem.*, 29 (2016) 194-197.
- [174] D.M. Deojay, J.Z. Sostaric, L.K. Weavers, Exploring the effects of pulsed ultrasound at 205 and 616kHz on the sonochemical degradation of octylbenzene sulfonate, *Ultrason. Sonochem.*, 18 (2011) 801-809.
- [175] J. Lee, K. Yasui, T. Tuziuti, T. Kozuka, A. Towata, Y. Iida, Spatial distribution enhancement of sonoluminescence activity by altering sonication and solution conditions, *J. Phys. Chem. B*, 112 (2008) 15333-15341.
- [176] M. Hauptmann, H. Struyf, S. De Gendt, C. Glorieux, S. Brems, Evaluation and interpretation of bubble size distributions in pulsed megasonic fields, *J. Appl. Phys.*, 113 (2013) 184902.
- [177] H. Flynn, C.C. Church, A mechanism for the generation of cavitation maxima by pulsed ultrasound, *J. Acoust. Soc. Am.*, 76 (1984) 505-512.
- [178] D.J. Casadonte, M. Flores, C. Petrier, Enhancing sonochemical activity in aqueous media using power-modulated pulsed ultrasound: an initial study, *Ultrason. Sonochem.*, 12 (2005) 147-152.
- [179] C. Dekerkcheer, K. Bartik, J.-P. Lecomte, J. Reisse, Pulsed sonochemistry, *J. Phys. Chem. A*, 102 (1998) 9177-9182.
- [180] C. Zhu, R. Feng, Y. Zhao, Sonochemical effect of a bifrequency irradiation, *Chin. Sci. Bull.*, 45 (2000) 142-145.
- [181] M. Hauptmann, F. Frederickx, H. Struyf, P. Mertens, M. Heyns, S. De Gendt, C. Glorieux, S. Brems, Enhancement of cavitation activity and particle removal with pulsed high frequency ultrasound and supersaturation, *Ultrason. Sonochem.*, 20 (2013) 69-76.
- [182] W.-S. Chen, A.A. Brayman, T.J. Matula, L.A. Crum, M.W. Miller, The pulse length-dependence of inertial cavitation dose and hemolysis, *Ultrasound Med. Biol.*, 29 (2003) 739-748.
- [183] T. Leighton, M. Pickworth, A. Walton, P. Dendy, The pulse enhancement of unstable cavitation by mechanisms of bubble migration, in: *Proc. Inst. Acoust.*, 1989, pp. 461-469.
- [184] M.R. Bailey, Dual Frequency High Frequency Ultrasound To Control Bubbles, *International Symposium on Therapeutic Ultrasound: 2nd International Symposium on Therapeutic Ultrasound*, (2002).
- [185] J. Holzfuss, M. Rüggeberg, R. Mettin, Boosting sonoluminescence, *Phys. Rev. Lett.*, 81 (1998) 1961.
- [186] P.M. Kanthale, A. Brothie, M. Ashokkumar, F. Grieser, Experimental and theoretical investigations on sonoluminescence under dual frequency conditions, *Ultrason. Sonochem.*, 15 (2008) 629-635.
- [187] P.A. Tatake, A.B. Pandit, Modelling and experimental investigation into cavity dynamics and cavitation yield: influence of dual frequency ultrasound sources, *Chem. Eng. Sci.*, 57 (2002) 4987-4995.
- [188] G. Servant, J.-L. Laborde, A. Hita, J.-P. Caltagirone, A. Gérard, Spatio-temporal dynamics of cavitation bubble clouds in a low frequency reactor: comparison between theoretical and experimental results, *Ultrason. Sonochem.*, 8 (2001) 163-174.
- [189] P.R. Birkin, D.G. Offin, T.G. Leighton, Experimental and theoretical characterisation of sonochemical cells. Part 2: cell disruptors (Ultrasonic horns) and cavity cluster collapse, *PCCP*, 7 (2005) 530-537.

- [190] P.R. Birkin, D.G. Offin, C.J. Vian, T.G. Leighton, A.O. Maksimov, Investigation of noninertial cavitation produced by an ultrasonic horn, *J. Acoust. Soc. Am.*, 130 (2011) 3297-3308.
- [191] S. Nikitenko, C. Le Naour, P. Moisy, Comparative study of sonochemical reactors with different geometry using thermal and chemical probes, *Ultrason. Sonochem.*, 14 (2007) 330-336.
- [192] K. Yasuda, K. Matsuura, Y. Asakura, S. Koda, Effect of agitation condition on performance of sonochemical reaction, *Jpn. J. Appl. Phys.*, 48 (2009) 07GH04.
- [193] S. Hatanaka, K. Yasui, T. Kozuka, T. Tuziuti, H. Mitome, Influence of bubble clustering on multibubble sonoluminescence, *Ultrasonics*, 40 (2002) 655-660.
- [194] M.S. Longuet-Higgins, Viscous streaming from an oscillating spherical bubble, in: *Proc. R. Soc. A, The Royal Society*, 1998, pp. 725-742.
- [195] W.C. Boon, K. Petkovic-Duran, K. White, E. Tucker, A. Albiston, R. Manasseh, M.K. Horne, T.D. Aumann, Acoustic microstreaming increases the efficiency of reverse transcription reactions comprising single-cell quantities of RNA, *Biotechniques*, 50 (2011) 116-119.
- [196] J. Zhou, Y. Tada, Y. Kato, Y. Nagatsu, K. Yasui, Y. Sakaguchi, R. Takebayashi, K. Iwata, Effects of Reflection Plate on Ultrasonic Reaction in a Sonoreactor, *J. Chem. Eng. Jpn.*, 42 (2009) 570-575.
- [197] Y. Asakura, T. Nishida, T. Matsuoka, S. Koda, Effects of ultrasonic frequency and liquid height on sonochemical efficiency of large-scale sonochemical reactors, *Ultrason. Sonochem.*, 15 (2008) 244-250.
- [198] V. Renaudin, N. Gondrexon, P. Boldo, C. Petrier, A. Bernis, Y. Gonthier, Method for determining the chemically active zones in a high-frequency ultrasonic reactor, *Ultrason. Sonochem.*, 1 (1994) S81-S85.
- [199] M.-L. Doche, J.-Y. Hihn, A. Mandroyan, R. Viennet, F. Touyeras, Influence of ultrasound power and frequency upon corrosion kinetics of zinc in saline media, *Ultrason. Sonochem.*, 10 (2003) 357-362.
- [200] C. Little, M. El-Sharif, M. Hepher, The effect of solution level on calorific and dosimetric results in a 70kHz tower type sonochemical reactor, *Ultrason. Sonochem.*, 14 (2007) 375-379.
- [201] E.A. Hemmingsen, Cavitation in gas-supersaturated solutions, *J. Appl. Phys.*, 46 (1975) 213-218.
- [202] T. Mason, J. Lorimer, D. Bates, Quantifying sonochemistry: casting some light on a 'black art', *Ultrasonics*, 30 (1992) 40-42.
- [203] Y. Asakura, M. Maebayashi, T. Matsuoka, S. Koda, Characterization of sonochemical reactors by chemical dosimetry, *Electron. Comm. Jpn.*, 3, 90 (2007) 1-8.
- [204] J. Zhou, Y. Tada, Y. Kato, Y. Nagatsu, K. Iwata, K. Yasui, Y. Sakaguchi, R. Takebayashi, Enhancement of sonochemical reaction with the addition of aluminum powder in a sonoreactor, *J. Chem. Eng. Jpn.*, 43 (2010) 389-393.
- [205] A. Nakajima, H. Sasaki, Y. Kameshima, K. Okada, H. Harada, Effect of TiO₂ powder addition on sonochemical destruction of 1, 4-dioxane in aqueous systems, *Ultrason. Sonochem.*, 14 (2007) 197-200.
- [206] V. Morosini, T. Chave, M. Viro, P. Moisy, S.I. Nikitenko, Sonochemical water splitting in the presence of powdered metal oxides, *Ultrason. Sonochem.*, 29 (2016) 512-516.
- [207] Y. Iwai, S. Li, Cavitation erosion in waters having different surface tensions, *WEAR*, 254 (2003) 1-9.
- [208] R. Toegel, S. Luther, D. Lohse, Viscosity destabilizes sonoluminescing bubbles, *Phys. Rev. Lett.*, 96 (2006) 114301.
- [209] S. Brems, M. Hauptmann, E. Camerotto, A. Pacco, H. Struyf, P. Mertens, C. Gottschalk, S. De Gendt, The influence of dissolved carbon dioxide on cavitation intensity in ultrasound cleaning systems, *Jpn. J. Appl. Phys.*, 52 (2013) 066602.
- [210] J.D. Seymour, R.B. Gupta, Oxidation of aqueous pollutants using ultrasound: salt-induced enhancement, *Ind. Eng. Chem. Res.*, 36 (1997) 3453-3457.

- [211] R. Pflieger, J.T. Lee, S.I. Nikitenko, M. Ashokkumar, Influence of He and Ar Flowrate and NaCl Concentration on the Size Distribution of Bubbles Generated by Power Ultrasound, *J. Phys. Chem. B*, (2015).
- [212] M. Wall, M. Ashokkumar, R. Tronson, F. Grieser, Multibubble sonoluminescence in aqueous salt solutions, *Ultrason. Sonochem.*, 6 (1999) 7-14.
- [213] C. Browne, R.F. Tabor, D.Y. Chan, R.R. Dagastine, M. Ashokkumar, F. Grieser, Bubble coalescence during acoustic cavitation in aqueous electrolyte solutions, *Langmuir*, 27 (2011) 12025-12032.
- [214] J.-C. Lin, S.-L. Lo, C.-Y. Hu, Y.-C. Lee, J. Kuo, Enhanced sonochemical degradation of perfluorooctanoic acid by sulfate ions, *Ultrason. Sonochem.*, 22 (2015) 542-547.
- [215] G.J. Price, M. Ashokkumar, F. Grieser, Sonoluminescence quenching of organic compounds in aqueous solution: Frequency effects and implications for sonochemistry, *J. Am. Chem. Soc.*, 126 (2004) 2755-2762.
- [216] L. Dharmarathne, F. Grieser, Formation of Amino Acids on the Sonolysis of Aqueous Solutions Containing Acetic Acid, Methane, or Carbon Dioxide, in the Presence of Nitrogen Gas, *J. Phys. Chem. A*, 120 (2016) 191-199.
- [217] E.J. Hart, A. Henglein, Free radical and free atom reactions in the sonolysis of aqueous iodide and formate solutions, *J. Phys. Chem. (US)*, 89 (1985) 4342-4347.
- [218] H. Harada, C. Hosoki, M. Ishikane, Sonophotocatalysis of water in a CO₂-Ar atmosphere, *J. Photoch. Photobio. A*, 160 (2003) 11-17.
- [219] E.J. Hart, A. Henglein, Sonochemistry of aqueous solutions: hydrogen-oxygen combustion in cavitation bubbles, *J. Phys. Chem.*, 91 (1987) 3654-3656.
- [220] I. Hua, M.R. Hoffmann, Optimization of ultrasonic irradiation as an advanced oxidation technology, *Environ. Sci. Technol.*, 31 (1997) 2237-2243.
- [221] A. Henglein, Sonolysis of carbon dioxide, nitrous oxide and methane in aqueous solution, *Z. Naturforsch. B Chem. Sci.*, 40 (1985) 100-107.
- [222] E.J. Hart, A. Henglein, Sonolytic decomposition of nitrous oxide in aqueous solution, *J. Phys. Chem. (US)*, 90 (1986) 5992-5995.
- [223] H. Harada, Sonochemical reduction of carbon dioxide, *Ultrason. Sonochem.*, 5 (1998) 73-77.
- [224] E. Dalodière, M. Viot, P. Moisy, S.I. Nikitenko, Effect of Ultrasonic Frequency on H₂O₂ Sonochemical Formation rate in Aqueous Nitric Acid Solutions in the Presence of Oxygen, *Ultrason. Sonochem.*, (2015).
- [225] L. Rayleigh, VIII. On the pressure developed in a liquid during the collapse of a spherical cavity, *Philos. Mag.*, 34 (1917) 94-98.
- [226] S.J. Doicrycz, K.S. Suslick, Interparticle collisions driven by ultrasound, *Science*, 247 (1990) 1067-1069.
- [227] K. Suslick, M. Fang, T. Hyeon, M. Mdeleleni, Applications of sonochemistry to materials synthesis, in: *Sonochemistry and sonoluminescence*, Springer, 1999, pp. 291-320.
- [228] J. Lee, S. Kentish, M. Ashokkumar, Effect of surfactants on the rate of growth of an air bubble by rectified diffusion, *J. Phys. Chem. B*, 109 (2005) 14595-14598.
- [229] T. Leong, S. Wu, S. Kentish, M. Ashokkumar, Growth of bubbles by rectified diffusion in aqueous surfactant solutions, *J. Phys. Chem. C*, 114 (2010) 20141-20145.
- [230] V.K. La Mer, T.W. Healy, L. Aylmore, The transport of water through monolayers of long-chain n-paraffinic alcohols, *J. Coll. Sci.*, 19 (1964) 673-684.
- [231] M. Ashokkumar, R. Hall, P. Mulvaney, F. Grieser, Sonoluminescence from aqueous alcohol and surfactant solutions, *J. Phys. Chem. B*, 101 (1997) 10845-10850.
- [232] N. Segebarth, O. Eulaerts, J. Reisse, L.A. Crum, T.J. Matula, Correlation between acoustic cavitation noise, bubble population, and sonochemistry, *J. Phys. Chem. B*, 106 (2002) 9181-9190.
- [233] L. Crum, J. Gallemler, D. Nordling, Effect of Surface Tension on the Cavitation Threshold of Water, *J. Acoust. Soc. Am.*, 52 (1972) 151-152.

- [234] B. Han, B. Yang, Z. Shen, J. Lu, X. Ni, Numerical Investigation of the Influences of Liquid Viscosity, Surface Tension and Initial Bubble Gas Content on the Dynamic Properties of a Laser-Induced Cavitation Bubble, *Lasers Eng.*, 19 (2010).
- [235] L.A. Crum, Tensile strength of water, *Nature*, 278 (1979) 148–149.
- [236] M.C. Weinberg, Surface tension effects in gas bubble dissolution and growth, *Chem. Eng. Sci.*, 36 (1981) 137–141.
- [237] L. Chen, Y. Li, R. Manasseh, The coalescence of bubbles-a numerical study, in: *Proceedings of Third International Conference on Multiphase Flow: ICMF*, 1998.
- [238] A. Chakravarty, T. Georgiou, T.E. Phillipson, A.J. Walton, Stable sonoluminescence within a water hammer tube, *Phys. Rev. E*, 69 (2004) 066317.
- [239] Y. Hao, A. Prosperetti, The effect of viscosity on the spherical stability of oscillating gas bubbles, *Phys. Fluids* (1994-present), 11 (1999) 1309–1317.
- [240] S. Hilgenfeldt, D. Lohse, M.P. Brenner, Phase diagrams for sonoluminescing bubbles, *Phys. Fluids* (1994-present), 8 (1996) 2808–2826.
- [241] G. Taylor, The instability of liquid surfaces when accelerated in a direction perpendicular to their planes. I, in: *Proc. R. Soc. A, The Royal Society*, 1950, pp. 192–196.
- [242] M.P. Brenner, D. Lohse, T. Dupont, Bubble shape oscillations and the onset of sonoluminescence, *Phys. Rev. Lett.*, 75 (1995) 954.
- [243] A. Moshaii, M. Faraji, S. Tajik-Nezhad, Study of single bubble Sonoluminescence in phosphoric acid, *Ultrason. Sonochem.*, 18 (2011) 1148–1152.
- [244] P.R. Gogate, A.M. Wilhelm, A.B. Pandit, Some aspects of the design of sonochemical reactors, *Ultrason. Sonochem.*, 10 (2003) 325–330.
- [245] R. Sadighi-Bonabi, N. Rezaee, H. Ebrahimi, M. Mirheydari, Interaction of two oscillating sonoluminescence bubbles in sulfuric acid, *Phys. Rev. E*, 82 (2010) 016316.
- [246] H. Okawa, J. Yabuki, Y. Kawamura, I. Arise, M. Sato, Synthesis of FePO₄ cathode material for lithium ion batteries by a sonochemical method, *Mater. Res. Bull.*, 43 (2008) 1203–1208.
- [247] T. Ouerhani, R. Pflieger, W.B. Messaoud, S.I. Nikitenko, Spectroscopy of Sonoluminescence and Sonochemistry in Water Saturated with N₂–Ar Mixtures, *J. Phys. Chem. B*, 119 (2015) 15885–15891.
- [248] G. Mark, H.-P. Schuchmann, C. von Sonntag, Formation of peroxyxynitrite by sonication of aerated water, *J. Am. Chem. Soc.*, 122 (2000) 3781–3782.
- [249] M. Takahashi, ζ potential of microbubbles in aqueous solutions: electrical properties of the gas-water interface, *J. Phys. Chem. B*, 109 (2005) 21858–21864.
- [250] J.K. Beattie, A.M. Djerdjev, G.G. Warr, The surface of neat water is basic, *Faraday Discuss.*, 141 (2009) 31–39.
- [251] P.K. Weissenborn, R.J. Pugh, Surface tension of aqueous solutions of electrolytes: relationship with ion hydration, oxygen solubility, and bubble coalescence, *J. Coll. Interf. Sci.*, 184 (1996) 550–563.
- [252] J.K. Beattie, A.M. Djerdjev, A. Gray-Weale, N. Kallay, J. Lützenkirchen, T. Preočanin, A. Selmani, pH and the surface tension of water, *J. Coll. Interf. Sci.*, 422 (2014) 54–57.
- [253] P.C. Hiemenz, *Principles of colloid and surface chemistry*, M. Dekker New York, 1986.
- [254] R.F. Tabor, D.Y. Chan, F. Grieser, R.R. Dagastine, Anomalous Stability of Carbon Dioxide in pH-Controlled Bubble Coalescence, *Angew. Chem. Int. Ed.*, 50 (2011) 3454–3456.
- [255] J. Harper, Bubble rise in a liquid with a surfactant gas, in particular carbon dioxide, *J. Fluid Mech.*, 581 (2007) 157–165.
- [256] W. Yan, G.-Y. Zhao, G.-J. Chen, T.-M. Guo, Interfacial tension of (methane + nitrogen)+ water and (carbon dioxide + nitrogen) + water systems, *J. Chem. Eng. Data*, 46 (2001) 1544–1548.
- [257] S.D. Lubetkin, M. Akhtar, The variation of surface tension and contact angle under applied pressure of dissolved gases, and the effects of these changes on the rate of bubble nucleation, *J. Coll. Interf. Sci.*, 180 (1996) 43–60.
- [258] Y.T. Didenko, W.B. McNamara III, K.S. Suslick, Effect of noble gases on sonoluminescence temperatures during multibubble cavitation, *Phys. Rev. Lett.*, 84 (2000) 777.

- [259] R. Pflieger, A.A. Ndiaye, T. Chave, S.I. Nikitenko, Influence of ultrasonic frequency on Swan band sonoluminescence and sonochemical activity in aqueous tert-butyl alcohol solutions, *J. Phys. Chem. B*, 119 (2014) 284-290.
- [260] W.B. McNamara, Y.T. Didenko, K.S. Suslick, Sonoluminescence temperatures during multi-bubble cavitation, *Nature*, 401 (1999) 772-775.
- [261] J. Rooze, E.V. Rebrov, J.C. Schouten, J.T. Keurentjes, Dissolved gas and ultrasonic cavitation—a review, *Ultrason. Sonochem.*, 20 (2013) 1-11.
- [262] Y. Mori, K. Hijikata, T. Nagatani, Effect of dissolved gas on bubble nucleation, *Int. J. Heat Mass Transfer*, 19 (1976) 1153-1159.
- [263] S. Lubetkin, Why is it much easier to nucleate gas bubbles than theory predicts?, *Langmuir*, 19 (2003) 2575-2587.
- [264] M. Margulis, Sonoluminescence and ultrasonic chemical reactions, *Sov. Phys. Acoust.-USSR*, 15 (1969) 135-&.
- [265] V. Vinogradov, Depression of the cavitation centers in water under pulsed tension conditions, *Tech. Phys. Lett.*, 35 (2009) 54-56.
- [266] S. Kumari, M. Keswani, S. Singh, M. Beck, E. Liebscher, P. Deymier, S. Raghavan, Control of sonoluminescence signal in deionized water using carbon dioxide, *Microelectron. Eng.*, 88 (2011) 3437-3441.
- [267] V. Buch, A. Milet, R. Vácha, P. Jungwirth, J.P. Devlin, Water surface is acidic, *Proceedings of the National Academy of Sciences*, 104 (2007) 7342-7347.
- [268] H. Harada, Y. Ono, M. Oda, Decrease in the rate of sonochemical oxidation with introduction of CO₂, *Jpn. J. Appl. Phys.*, 53 (2014) 07KE10.
- [269] S. Merouani, O. Hamdaoui, The size of active bubbles for the production of hydrogen in sonochemical reaction field, *Ultrason. Sonochem.*, 32 (2016) 320-327.
- [270] S.B. Reifsneider, L.A. Spurlock, Chemistry of ultrasound. II. Irradiative behavior of aliphatic aldehydes and carboxylic acids in an aqueous medium, *J. Am. Chem. Soc.*, 95 (1973) 299-305.
- [271] M. Margulis, *Sonochemistry and cavitation*, Gordon and Breach Publishers, 1995.
- [272] I. Èl'piner, *Ultrasound: Physical, Chemical, and Biological Effects*, Consultants Bureau, (1964).

A Parametric Review of Sonochemistry: Control and Augmentation of Sonochemical Activity

Highlights

- Review of the parameters that control ultrasonic cavitation and sonochemistry.
- Primary and secondary parameters are defined and their influence determined.
- Discussed are changes in chemical and gas environment.
- Parameters are considered from the viewpoint of bubble structural instability.
- Bubble instabilities induce fragmentation effecting sonochemical efficiency.