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REVIEW

PRINCIPLES AND EFFECTS OF ACOUSTIC CAVITATION

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In the recent years, food industry has shown a real interest in ultrasound use because of its effect on physical, biochemical and microbial properties of food systems. In order to better understand how the acoustic cavity effects could be best applied in food industry, a review on acoustic cavitation and its effects was done. The present paper describes in detail the basic principles underlying the effects of ultrasounds on food processing applications. It also provides theoretical background on acoustic cavitation and ultrasound production method. Moreover, harnessing mechanic, optic, chemical and biological effects of acoustic cavitation in food industry were briefly highlighted.

Keywords: ultrasound, acoustic, cavitation, sonochemistry, sonoluminescence.

Introduction

Significant changes in the nature of food products as well as in their ingredients can be determined by acoustic bubbles and acoustic cavitation (Vilkhu et al., 2011). Moreover, the effects of acoustic cavitation which are linked to the hot spot, radical production, pressure changes can cause from mild to moderate changes in food product (Bhaskaracharya et al., 2009). Physical effects – stronger when close to the solid/fluid and fluid/fluid limit - are given a special interest in applications. Ultrasound was successfully used for certain operations specific to food industry and a succinct presentation of some applications of acoustic cavitation effects used in emulsifying, homogenization, filtering, altering viscosity was done. Thus, it was proven that ultrasound is extremely effective in improving emulsification and homogenization when the shock waves appearing due to cavitational collapse near the surface of the boundary layer of two immiscible liquids can produce an efficient mixture of these two liquids (Wu et al., 2001; Ramachandran et al., 2006; Bosiljkov et al., 2011). Furthermore, microstreaming and the shear forces, which may appear as cavitation effect, can be used to improve the filtering process as well as to clean the fouled membranes (Muthukumaran et al., 2007; Maskooki et al., 2010). The effects of acoustic cavitiation proved useful also for modifying the viscosity of starch solution as well as corn starch suspension (Iida et al., 2008; Jambrak et al., 2010). Even in the case of defoaming, ultrasound proved efficient as the energy transmitted by the acoustic beam during this application is powerful enough to destroy the foam without using chemical anti-foaming agents or mechanical breakers (De-Sarabia et al., 2006; Rodriguez et al., 2010).

As regards the biological effects of acoustic cavitation very little is known about the real effect of ultrasound on enzymes because there are contradictory studies on enzymes activation and inactivation (Rokhina et al., 2009).

In order to successfully use the ultrasound in different branches of food industry, the systematic knowledge of acoustic cavitation is necessary. Acoustic cavitation is a complex phenomenon which produces a series of effects in liquid, their consequences underlying most of the ultrasound applications. Thus, the main effects of acoustic cavitation, such as mechanic, optic, chemical, biological effects, were presented. Moreover, the method of producing ultrasound was briefly presented. For better understanding the effects of acoustic cavitation, the presentation of acoustic cavitation, namely bubble forming and movement in cavitational field, was necessary.

Acoustic cavitation

Ultrasound is similar to sound waves but has a frequency which cannot be perceived by human hearing. Being about the sound of waves, ultrasound can be propagated in any media (solid, gas and liquid) with elastic properties. Vibration movement is transmitted to the molecules of the medium and each of these transmits the movement to the neighbouring molecules before returning to the initial position. For liquids and gases, the particles oscillation happens in the direction of the wave and produces longitudinal waves. For solids, the particles movement is perpendicular on the direction of the waves leading to transversal waves. When a sound passes through a liquid, it generates compression and rarefaction regions (Mason and Lorimer, 2002). When the pressure during rarefaction is high enough, cavitation (or bubbles) is formed in the liquid. There is a difference between stabile cavitation characterized by the bubble's resistance during more cycles, and transient cavitation characterized by the growth of the bubble to collapse, whose consequence is fragmenting and disintegration into a mass of smaller bubbles (Leighton, 1995). In acoustic cavitation, many bubbles usually appear simultaneously and influence each other.

Bubble dynamics

At relatively low acoustic pressures, the radial movement of a bubble in a sonic field is linear. As the acoustic pressure has a sinusoidal form, oscillating between compression and rarefaction, this will be the movement of the bubble. During rarefaction the bubble increases in volume because of low pressure around the liquid and decreases in volume during compression because of high pressure. In this case, bubble motion is linear, being characterized by almost equal rates of expansion and contraction.

At high acoustic pressure, bubble motion becomes extremely nonlinear in rarefaction phase and the volume of the bubble increases in half a cycle. In compression phase due to high acoustic pressure, bubble motion is deviated and it continues to expand - even in the compression phase – up to a maximum radius. At that moment, the bubble collapses rapidly and undergoes a rapid decrease in volume to its initial radius, then reaching a minimum radius. When the bubble reaches the minimum radius, chemical reactions and light emission appear. After that, the bubble rapidly recovers and expands, undergoing a series of secondary compressions and expansions until it reaches the original radius (Suslick and Flannigan, 2008).

When the bubble is in the expansion phase, gas diffuses inside the bubble and when the bubble is in the compression phase, gas diffuses outside the bubble. The time of the bubble in the rarefaction phase is long compared to the time in the compression phase, thus after a complete cycle, more gas will diffuse inside the bubble than outside and will cause the bubble to grow. This phenomenon, called rectified diffusion, will be reduced if gas quantity in sonicated liquid is lower than the saturation level. Thus, reducing the concentration of the dissolved gas, it is possible to produce a stable cavity of only one bubble with big radii for each cycle (Crum, 1994). When more bubbles are simultaneously present in the acoustic cavitation, they interact and form structures. Images were recorded with high speed cameras, showing that bubbles aggregate into a filament structure (Lauterborn et al., 2008). The structure of acoustic cavitation formed in liquid at low ultrasonic frequencies is different. Thus, some types of structure, such as *streamer*, *filament*, *jellyfish*, *starfish*, are in tight connection with standing waves, others – *flare*, *sonotrode*, *cone bubble structure* (CBS) - with travelling waves in acoustic field. There are types of structure – *clusters*, *smokers* and *webs* - which can appear in all acoustic fields, in liquid or adherent to solid surfaces (transducers, submerged objects, walls). Their lifetime ranges from one or few cycles in *clusters* or *sonotrode* up to hundreds of cycles in streamers and filaments, the limiting factors of lifetimes being high bubble density, collision with neighbours (Mettin, 2005; Tervo et al., 2004).

Sonoluminescence

This effect of acoustic cavitation is the emission of light during cavitation process. The high temperatures and pressures generated in incondensable gases during cavitational collapse are considered to be responsible for luminescence, the emission of light. When this phenomenon appears in acoustic cavitation, it is called sonoluminescence, despite the proofs which show that it is the cavitation phenomenon which produces light emission rather than the sound (Brenen, 1995).

The first studies on sonoluminescence were focused on the light generated by the cloud of cavitational bubbles in ultrasonic field with an average frequency of 20 kHz to 2 MHz. Marinesco and Trillat first noticed this indirect phenomenon in

1933 but the fact that the light emission was produced during collapse was first noticed by Meyer and Kuttruff in 1959 (Brenen, 1995; Suslick and Flannigan, 2008). There are many theories regarding the origin of sonochemistry and sonoluminescence. One of the generally accepted explanations is the "hot spot" theory which states that the potential energy given to the bubble as it expands to maximum size is concentrated into a heated gas core as the bubble implodes (Suslick et al., 1999).

A recent theory of Yasui et al. (2008), showed that the light emission mechanism is explained by the light that results from an electron being accelerated by the collision with an ion or a neutral atom and which occurs in the weakly ionized plasma formed inside the heated bubble. Sonoluminescence can be produced in the case of a single bubble undergoing extremely nonlinear pulsations (SBSL) and also in the case of a field of bubbles undergoing cavitation, termed multibubble sonoluminescence (MBSL). There are theories supported by the presence of some experimental proofs regarding the existence of plasma inside the collapsing bubble (Flannigan and Suslick, 2005). It was proven the existence of a hot nucleus, energetically strong during SBSL, plasma generated during cavitation consisting of high energy atoms (Eddingsaas et al., 2008). The temperatures reached by the collapsing bubble depend on both the energy lost by sound emission at the collapse and on the energy used in internal processes such as vibrations, rotations, dissociation, ionization (Lohse, 2005).

Temperatures and pressures created at the end of bubble collapse can lead to forming interesting materials with unique properties. Therefore, in order to understand the intracavity conditions, researchers used kinetic and spectroscopic methods (Suslick and Flannigan, 2012).

In order to determine the temperature of the gasses inside the collapsing bubble, a correlation with the sonoluminescence emission spectra was done. Intensities, peak positions and profiles of emission lines from electronically excited atoms and molecules were used to quantify the temperatures and pressures during MBSL and SBSL. Thus, it was shown that during MBSL temperatures of ~5000K and pressures of ~300bar were recorded, while during SBSL temperatures of ~15000K and pressures higher than 1000atm were recorded (Flannigan et al., 2006; Suslick and Flannigan, 2008; Xu and Suslick, 2010; Suslick and Flannigan, 2012).

Sonochemistry

As a result of ultrasonic irradiation and the appearance of cavitational collapse, high temperatures and pressures appear, as well as extreme cooling and heating rates which generate a unique mechanism for generating chemical energy. Due to extreme conditions produced during cavitational collapse, it was noticed that there are two sites of sonochemical reactions: the first one is inside the bubble in a gaseous phase while the second is the liquid phase around the bubble. Chemical reactions involving free radicals can appear during collapse inside the bubble (because of extreme conditions the links break and free radicals are produced) as well as at bubble's interface (the temperatures in the thin layer of liquid which surrounds the bubble generate thermic reactions) (Suslick et al., 1999).

Apart from these two sites, the substances dissolved from the liquid outside the bubble can react with the free radicals inside the bubble (when the bubble breaks in the final phase of collapse and the unused free radicals are spread into the entire solution) or on the surface of the collapsed bubble (Leong et al., 2011). In the case of sonicated water solutions, due to the appearance of transient cavitation, hydrogen atoms and hydroxyl radicals are generated and they can recombine to form hydrogen and peroxide or can react with the substances dissolved in the gaseous phase.

When the water containing small gas nuclei is sonicated, due to high temperatures and pressures produced during cavitational collapse, water is dissociated into hydroxyl radicals and hydrogen atoms. In order to identify the hydroxyl radicals the "spin trapping" technique was used. This analytical technique used to detect free radicals in biological systems proved that hydroxyl radicals and hydrogen atoms are produced in sonicated water (Riesz et al., 1985).

Moreover, in order to measure the quantity of hydroxyl radicals generated during sonication the iodide method was used. By using different frequencies it was noticed that within the frequency interval 20-358 kHz, the amount of hydroxyl radicals generated increases by increasing frequency, but once the frequency keeps rising to 1062 kHz there is a quantitative decrease of hydroxyl radicals. Therefore, the amount of hydroxyl radicals generated was minimum at the frequency of 20 kHz which is characterised by transient cavitation of bubbles. When frequency is increased, cavitation becomes stable thus leading to an increase in the number of active bubbles and, implicitly, to a decrease in the quantity of hydroxyl radicals generated. Yet, this cannot explain the decrease in the quantity of hydroxyl radicals when frequency is raised up to 1062 kHz. In this case, when frequencies are quite high, the acoustic cycle becomes very short, thus restricting the quantity of water vapours which can evaporate in the bubble during the expansion phase of the acoustic cycle leading to a decrease in the quantity of hydroxyl radicals generated. The ultrasound frequency of 20 kHz seems to be an ideal frequency used in processing and extraction operations used in food industry as the amount of hydroxyl radicals generated is the lowest compared to other frequencies used. Higher frequencies can be used if hydroxyl radicals generated are minimized by using ascorbic acid or other methods. There is a potential of using hydroxyl radicals generated during sonication in order to improve the antioxidant activity by increasing the degree of hydroxylation of certain compounds such as flavonoids (Ashokkumar et al., 2008).

In solid-liquid heterogeneous systems, low acoustic intensities can be used in order to reduce the tension of the liquid at the liquid-solid interface. The use of ultrasound in order to accelerate the chemical reactions in heterogeneous systems has spread. When cavitation occurs in a liquid close to a solid surface, the dynamics of cavitational collapse is changed, cavitational collapse becomes asymmetric and generates high velocity jets which damage the shock wave on the solid surface. This phenomenon can produce erosions/corrosions and the cavitation and the shock waves produced during the ultrasonic irradiation of the liquid can accelerate the solid particles in powder suspensions, reaching high speeds. Thus, the microjet, the impact of the shock wave and the interparticle collisions have a substantial effect on the chemical and morphological composition of the solid which can improve its chemical reactivity (Suslick , 2001).

These high velocity interparticle collisions produce extreme heat at the impact point which can cause local melting or dramatic increase of the reaction rates in solid-liquid systems. A series of metallic powders was used in order to find out the temperatures reached and the interparticle collision velocity. The estimated collision velocity is close to half of the speed of sound in liquid and the real speed of particles during sonication depends on the dimension of the particles, particles smaller that 100 nano microns and bigger than 100 micro microns do not gather. Moreover, the cavitation effect for this phenomenon of interparticle collision is caused by the shock waves realeased into the liquid and not by the local temperatures, "hot spot", formed during caviational collapse (Prozorov et al., 2004).

Apart from the damages caused to solid surfaces the violent collapse of cavitational bubbles will generate noise as well, this being a consequence of momentary high pressures which appear due to strong bubble compressions. The cavitation beginning is often identified more with the noise rather than being visually perceived, several empirical methods suggesting that the estimation of material damages can be done by measuring the noise (Brennen, 1995).

Generation of ultrasound

The main elements of ultrasonic equipment used in generating and transmitting ultrasonic waves are an electrical power generator, a transducer and a sound emitter. The sound emitter most often used in food industry has the shape of a horn or a bath (Mason and Lorimer, 2002).

The ultrasound emitters are devices which transform the electric energy into acoustic energy and can be divided into two main groups: mechanical and electromechanical. Mechanical emitters are divided into aerodynamic and hydrodynamic, whereas the electromechanical ones are divided into electromagnetic, electrodynamical, magnetostrictive and piezoelectric. The emitter type used is chosen according to the frequency domain chosen, the propagation medium and technological process. Magnetostrictive and piezoelectric acoustic emitters are also called ultrasound transducers. The ultrasound generator represents the primary energy source (Tudose, 1997).

Magnetostrictive transducers are based on the fact that some ferromagnetic materials change the dimension when magnetized and when they are in a variable magnetic field they start to oscillate, becoming sources of acoustic waves. Piezoelectric transducers are more efficient than the magnetostrictive ones from the acoustic energy transfer point of view but they cannot last long at temperatures higher than 85°C. Piezoelectric materials are more efficient when converting electric energy into mechanical energy. They are cheaper and need lower tensions (Leadley and Williams, 2006).

Ultrasound equipment used in liquid media can have a whistle shape, ultrasonic baths or horn shape.

“Whistle” transducers use an ultrasonic mechanical source which is based on a jet of liquid which flows over a thin steel blade producing vibrations. This type of transducers is used in applications such as homogenising, mixing (Mason, 2000).

Ultrasonic baths are used for low intensities in 1-2W/cm^2 field. Transducers are fixed on the lower part of the tub, for small baths one transducer is enough, the number of transducers rising proportionally to the complexity of the system, thus increasing the power. This means that the frequency and power will depend upon the type and the number of transducers used.

“Horn” transducers use a horn-shaped device attached to a transducer in order to amplify the sonorous signal. A proper form of the horn will increase the vibrations amplitude at its surface, thus the choice between different shapes will depend upon the purpose and high intensities of a few hundreds of W/cm^2 could be reached (Mason and Lorimer, 2002).

Conclusions

The interest given to the study of ultrasound is especially due to the acoustic cavity effects which can be used for improving the processes in food industry. In recent years, many common methods used in food processing have been replaced with ultrasound technology due to its positive contribution in shortening the processes, in improving the quality of finite products, in improving production efficiency reflected by price. Future researches are required in order to use the ultrasound technology at a larger industrial scale.

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