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REVIEW ARTICLE

Advances in Ultrasonic and Megasonic Processing of Foods

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Abstract This article reviews the latest research in the airborne and aqueous-based applications of ultrasound across the wider range of frequencies in food processing. The effects of ultrasound at low and high frequencies, including stable and unstable sound-induced cavitation, microstreaming, biochemical stress responses and effects achieved through standing waves, are described in terms of food structural changes in dairy, meat and vegetable materials. Advances for recent applications in low- and/or high-frequency ultrasound, including enhanced drying and defoaming with airborne ultrasound, emulsification and homogenisation, green extraction methods and high-frequency ultrasound (megasonics) for non-solvent separation in oil extraction processes and separation or fractionation of milk fat. The manuscript concludes with recommendations for reactor design based on the experience gathered lately in installing transducers in industrial vessels for commercial applications in the oil and dairy industries.

Keywords Ultrasound · Megasonics · Dairy processing · Fruit and vegetable processing · Meat processing · Oil recovery · Extraction · Texture modification · Drying · Defoaming · Emulsification

Introduction

The use of ultrasound for food processing is of growing interest in the food industry, due to its capability to induce a range of beneficial effects and outcomes. The specific effects provided by ultrasound in foods depend greatly on the frequency (20 kHz to 3 MHz), power (1–10,000 W/cm^2) and medium (liquid, solid or gas) through which it is delivered.

In the food industry, ultrasonics has been mainly applied in diagnostics, e.g. non-destructive testing for flaw detection in material science, and processes such as cutting (e.g. frozen or soft foods through ultrasonic vibration on the edge of a cutting tool), homogenisation (e.g. sauces and mayonnaise through turbulent mixing induced by cavitation), extraction (e.g. enhanced yields of oils, flavourings and nutraceuticals from plants caused by the breakdown of cell walls), degassing (e.g. beverages before canning or bottling) and antifouling (of e.g. heat exchangers and membranes) [135]. Ultrasonic applications have broadened as a result of fairly recent developments of systems able to generate ultrasound in air at reasonable power levels as well as systems able to generate ultrasound at higher frequencies (>400 kHz) with various power levels (in the order of >100 W). Latest developments include the use of airborne ultrasound for defoaming and drying applications (e.g. [111, 117]), ultrasound at low frequencies to alter the casein micelles in dairy products (e.g. [80]), ultrasound at low and high frequencies for texture and structure modification of processed foods (by inducing biochemical stress responses; e.g. [30]), megasonics for enhanced palm oil separation (e.g. [63]) and megasonics for enhanced milk fat separation (e.g. [64]).

Ultrasound refers to acoustic pressure waves that have a frequency 20 kHz or higher [106]. Ultrasonics can be broadly categorised into two regions of efficacy for food processing, low-frequency, high-power ultrasound and high-frequency,

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low-power ultrasound. Firstly, the low-frequency, high-power ultrasound region, also known as the power ultrasound region [72], spans the frequency range between 20 and 100 kHz. This frequency range is characterised by the formation of large resonance size cavitation bubbles, which can collapse violently (i.e. ‘transient’ cavitation), generating locally high pressures (in excess of 500 bar) and temperatures (up to 5000 °C) ([135]) (Fig. 1). The strong shear forces created by low-frequency, high-power ultrasound are used in food processing to initiate rupture and breakdown of materials, which is highly useful for applications such as homogenisation [4], emulsification [67] and extraction [19]. The temperature hot spots can be used to modify temperature-sensitive materials such as proteins [120] or increase the rate of chemical transformation in foods [157].

The high-frequency low-power ultrasound region that is suited for food processing spans the frequency range from 100 kHz to 3 MHz. As the frequency of applied ultrasound increases, the resonance size for the bubble collapse events decrease, coinciding with a decline in the intensity of the physical shear forces and temperature hot spots. This type of ultrasound tends to produce cavitation that is more likely to be of the ‘stable’ variety (less violent collapse of smaller bubbles), while inducing other acoustic effects such as microstreaming due to the increased attenuation of high-frequency ultrasound as it travels through a medium [125]. It should be noted that although the intensity of bubble collapse declines, peak sonochemical radical formation occurs in the frequency range between 400 and 800 kHz [68, 87], due to there being an optimal population of sonochemically active bubbles. The food applications studied in this frequency range are fewer, since the chemical radicals formed can greatly accelerate oxidation of lipids in foods [22, 65, 138], which is highly undesirable.

Other effects in high-frequency ultrasound applications include inducing biochemical stress responses in living tissue (e.g. fruits and vegetables), which can promote the synthesis of lignin by reinforcing intercellular adhesion, and the modification of the pectin structure, enabling greater calcium ion bonding and cell wall stiffening [30]. High-frequency plate transducers may be used to create standing waves in purpose fit reactor designs [64]. These standing waves can form between an ultrasound source and a reflector placed at a distance multiple of a half-wavelength in parallel orientation to the source. Particles or droplets suspended in a continuous phase experience an acoustic force, which, depending on the material properties, move either towards the nodes or the antinodes of the standing wave field (Fig. 2). Particle separation is particularly effective when applied at megasonic frequencies (i.e. 1–3 MHz), as the acoustic forces responsible scale with increasing frequency.

This review will highlight some of the recent advances in ultrasonic and megasonic food processing, with a focus on aspects that existing reviews have not yet covered in great detail.

Review of Recent Advances of Ultrasonic and Megasonic Processing of Foods

Low-Frequency Ultrasound

Over the last few decades, researchers around the world have investigated the use of ultrasound at low frequencies and high powers for various food processing aspects, including not only the aforementioned applications, such as emulsification, dispersion and antifouling, but also the application for other purposes such as microbial and enzyme inactivation. Yet, there are a few applications of low-frequency ultrasound that have not been investigated in great detail, such as airborne ultrasound for defoaming and utilisation of the technology for altering properties in vegetables, meats and dairy products. The following section will give an overview on recent studies.

Airborne Ultrasound for Defoaming

Foam generation in tanks (e.g. in biotechnology applications such as fermenters, and tank applications in other processing industries) and on beverage bottling lines through aeration and agitation of liquids, can lead to significant losses of product and decreases the useful volume of processing equipment, culminating in a major decline to the operating efficiency.

Current methods to reduce foaming include cooling the product, using mechanical breakers or by using chemical antifoaming agents. These methods are not convenient because liquid cooling demands high energy input whilst mechanical breakers or antifoaming agents may cause contamination to the product [111], producing a negative effect on the product quality and impact on the environment.

Reducing product loss by defoaming through low-energy airborne ultrasound application shows great potential for improving the environmental sustainability by reducing water and energy consumption during production. Ultrasound can be applied through air to interact with foams, provided short working distances (i.e. few centimetres) are used [110]. Foam bubbles are broken almost instantly when the applied ultrasonic energy is dissipated within the elastic foam layer, causing it to collapse. Research at the CSIRO in Australia has evolved around the development of sonotrodes, capable of transmitting the mechanical energy generated in the transducer at relatively high powers into the air, where the sound waves lead to efficient defoaming capacity on soft drink bottling lines [28].

While the concept of the technology has been proven [111], the underlying mechanism remains unclear. It has been hypothesised that high-frequency vibration of the foam

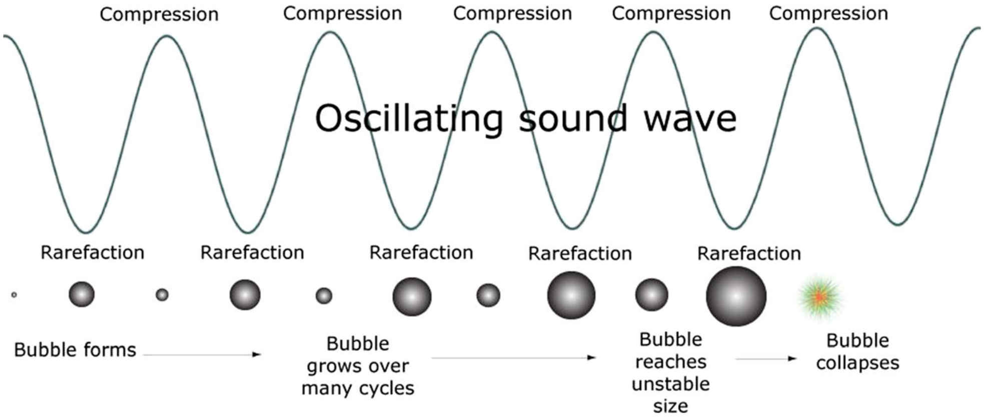

Fig. 1 Visual representation of the growth and collapse of an ultrasound-induced cavitation bubble, sourced with permission from Leong et al. [72]

bubbles and induced cavitation in the bubble matrix enables overcoming of the surface tension of the liquid film, causing bubble collapse. Another likely mechanism that plays a role is the formation of acoustic streaming currents in the liquid phase of the foam that results in a reduction in the apparent viscosity (due to shear thinning behaviour), causing rapid fluid draining, merging of bubbles and collapse [88].

Foams that are stabilised by high levels of protein may not be able to be defoamed effectively by ultrasound due to the proteins denaturing into stable structures at the aqueous liquid-gas interfaces [102]. Carbonated soft drinks and premixed alcoholic drinks, which have negligible protein content, form weak foam structures that can be readily reduced using ultrasound, as shown by Mawson et al. [88]. However, beer foams are less effectively defoamed, due to the presence of residual barley proteins which can provide high foam stability. Foams in dairy streams or those which are heavily laden with fats stabilised by fatty acid surfactants in waste treatment tanks, are also less effectively defoamed by ultrasound. Despite the aforementioned limitations, ultrasound provides a unique, contactless and hence clean method by which foaming can be controlled in selected food processing applications [103].

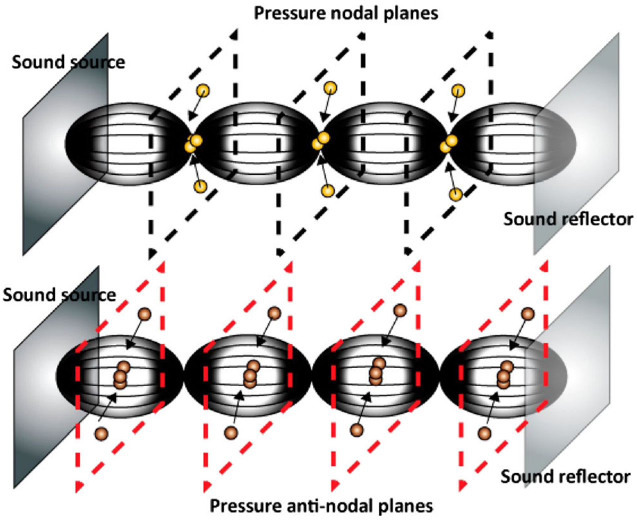

Fig. 2 Visual representation of the formation of standing waves in a reflecting ultrasound system and collection of particles in the pressure nodes, sourced with permission from Leong et al. [75]

Airborne Ultrasound for Enhanced Drying

Drying is a very important process in the food manufacturing industry, commonly used for the preservation of fruits, vegetables and meats as the removal of moisture prolongs shelf life by minimising bacterial growth and enzymatic activity. Convective hot air drying is the most common method employed and involves simultaneous heat and mass transfer to remove moisture from the food product.

Critically, the drying process is very high in energy consumption and probably the largest energy consuming unit operation in the food industry. Drying rates are often slow, and the high temperatures that foods can be exposed to within the hot air stream may result in reduction of the quality of the food such as product shrinkage, undesirable off-flavours and product discoloration [46].

Recent studies on airborne ultrasound in drying applications have shown that product drying time can be reduced by over 50% [117]. This can, therefore, be used not only to shorten the drying time but also to operate the drying process at reduced temperatures, both of which directly impact on the energy consumption. Furthermore, the reduced thermal load on the product leads to better retention of quality attributes, such as colour and flavour, as well as the retention of health promoting heat-sensitive components.

Ultrasound accelerates the drying rate by enhancing the mass transfer rate at which moisture is expelled from solids [31]. There are a number of proposed mechanisms responsible for ultrasonically enhanced drying of solid foods, although the exact mechanism is unclear. Primarily, the drying enhancement is due to the oscillations of ultrasonic waves that cause solid materials to vibrate rapidly. The alternating compressions and expansions that occur as sound is transmitted through the solid promote the formation of microscopic channels inside the material that enhances the diffusion of moisture locked within the material to the outside. This is analogous to the squeezing of a sponge, but at a very rapid rate. An alternative theory is that the diffusion boundary layer forming across the surface of a dried product is disturbed by the pressure waves, which leads to easier water removal from the product and a faster secondary drying stage rate [117]. Cavitation may also occur within the aqueous phase locked inside the solid material as a secondary mechanism, causing localised heating that makes removal of the moisture easier [127].

For drying of solid foods, an ultrasonic transducer can be coupled in direct contact with the food or applied via airborne ultrasound [31] (see Fig. 3). The ultrasound can be applied as a pretreatment step so that the diffusivity of moisture is enhanced during convective drying [38, 99] or the ultrasound can be applied simultaneously during the drying process [31]. Ultrasonic compression and expansion tend to be stronger in highly porous materials or materials that have large acoustic energy adsorption.

Several factors can influence the drying rate. The acoustic power used is one of the key parameters. De la Fuente-Blanco et al. [31] showed how increasing the acoustic power delivered to the system resulted in faster dehydration rates of carrots. An acoustic power of 100 W was applied for 90 min, resulting in sample mass reduction of 70% compared with controls that reduced sample mass by only 10%.

For ultrasound applied through air, the distance of the ultrasound source (i.e. vibrating plate) from the food material determines the intensity of airborne ultrasound arriving to the drying material. This is mainly due to attenuation of ultrasonic energy with propagation distance. Ultrasound intensity follows the inverse square law with increasing distance from the source. That is, the ultrasonic intensity decays rapidly with increasing ultrasonic radiation distance. For example, with 21-kHz ultrasound wave, when the ultrasonic radiation distance is 10 cm, sound intensity and sound power in air experiences a loss of 20%. When the distance is increased to 25 cm, the sound levels are reduced by more than 60% [32]. In the context of ultrasound-assisted drying, the smaller the ultrasonic radiation distance, the smaller the loss of ultrasonic energy, resulting in improved drying effects. For effective propagation of ultrasonic radiation for application of airborne ultrasound, there should be a small distance to enable effective air flow in convective drying.

There are several examples in which ultrasound can be used to successfully accelerate food drying. Fruits and vegetables such as carrots [31], plums [116], apple [117] and papaya [38] can be dried significantly faster, with further advantages including the ability to dry at lower temperatures, thereby improving product quality. Denglin et al. [32] demonstrated that the drying efficiency of carrot slices was improved with a short distance between the sample and the transducer of 15 cm, with the drying time being reduced from 240 to 150 min compared with controls without ultrasound. At longer distances, drying times increased back to 190 and 220 min, respectively, for 25 and 30 cm.

The effect of ultrasound frequency on the drying effectiveness of food materials has been less well studied. Most literature on the application of airborne ultrasound in food drying processes have operated at a constant frequency in the range of 20–26 kHz [10, 32, 70, 100]. Some early works undertaken by Muralidhara and Ensminger [96] on acoustic drying of green rice evaluated the effect of acoustic drying of rice in a fluidised bed at two sound frequencies (12 and 19 kHz). Their results indicated that exposure to either 12 or 19 kHz had little effect on the drying rate.

Low-Frequency Ultrasound for Altering Food Structures

The texture and consistency of food products, which contribute to the palatability and pleasure of eating food [39, 93], is highly dependent on the physical structure of the food. Food structure also affects flavour release and perception, bioavailability and digestibility of nutrients [14, 36, 90], glycemic index [14, 98, 108] and the stability of food products [41].

Ultrasound offers a method that can be used to uniquely modify the structure of food. The primary mechanism for this modification is due to acoustic cavitation, which can be harnessed to modify food structure [121]. Specifically, the cavitation creates enormous shear forces (physical effects) and homolytic cleavage of water molecules to occur, resulting in the formation of hydroxy and hydroxyl free radicals (chemical effects) [86]. The strong shear forces that accompany cavitation can cause changes in the structure of food materials through the breakdown of weak intermolecular interaction forces such as hydrogen bonds and Van der Waals interactions [134], and abrasion and disintegration of particles [87], macromolecules [105, 119] and cellular compartments [37, 143]. Other physical effects such as microstreaming and shear stress may also contribute to the breakdown by altering the permeability of cell membranes and mass transfer [112], thus facilitating biochemical reactions that ultimately result in structural modification.

The free radicals formed during homolytic cleavage may cause chemical modification at the macrolevel of foods [11]. At low intensity, these radicals can elicit stress response reactions that lead to changes in cellular metabolism and structure [153]. In plant tissue, peroxidase-catalysed oxidation of cell wall structural proteins and phenolics can occur, leading to cross-linking between cell wall polysaccharides, toughening of the tissue and ultimately structural modification [13, 16].

Dairy Protein Modification Whey proteins are sensitive to heat treatment and their aggregation when subject to high temperatures will lead to an increase in the viscosity that

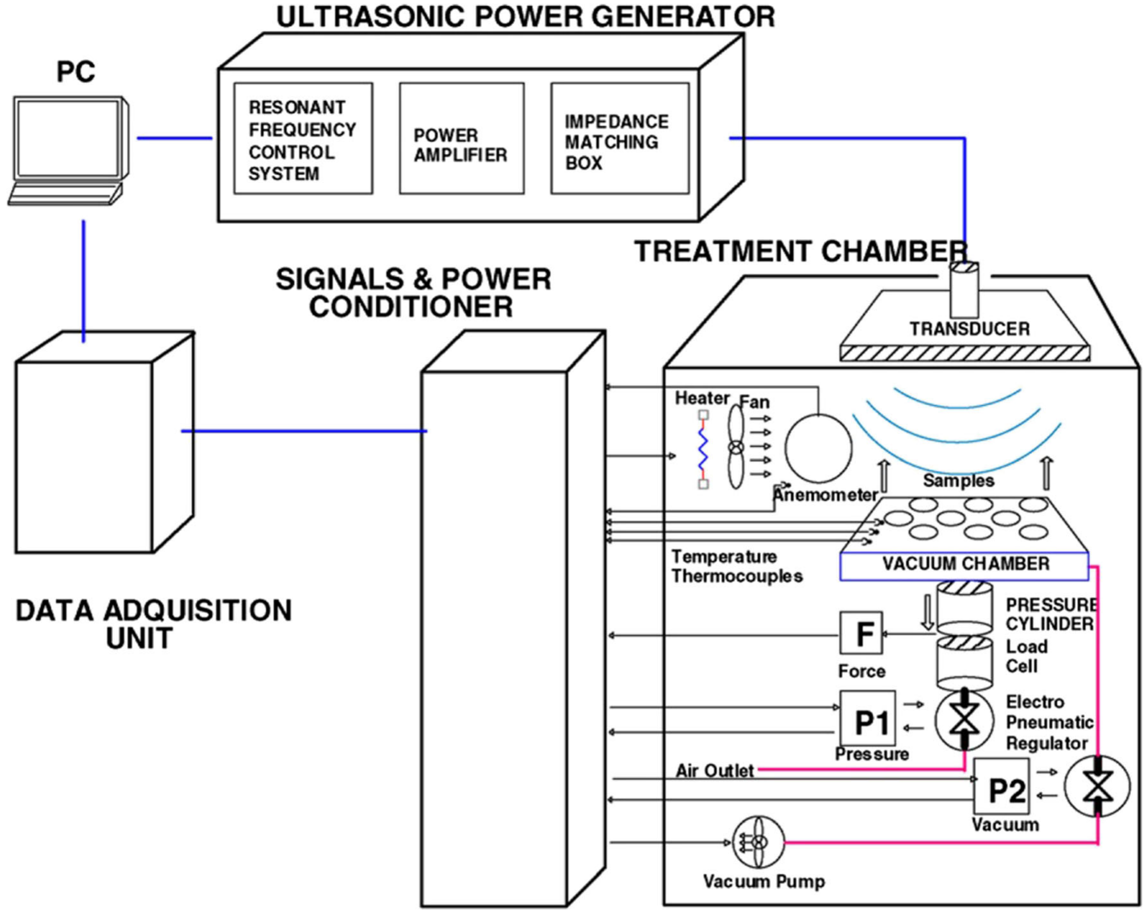

Fig. 3 A schematic of an ultrasonic dehydration system. Reprinted from De la Fuente-Blanco et al. [31], copyright 2006, with permission from Elsevier

makes downstream processing more challenging. It has been shown that in dairy systems, whey protein aggregates, once formed and broken up by ultrasound [6], do not reform again during subsequent thermal treatment. Zisu et al. [158] showed in a pilot-scale reactor that sonication at 20 kHz with an energy density of 260 J/ml could significantly reduce the size of whey protein aggregates formed in a 33 wt% solids solution. This process enables production of a heat-stable dairy powder or fluid which can be of great benefit in downstream food manufacturing.

Ultrasonic cavitation can also be employed beneficially for promoting the formation of dairy gels. The high energy of collapsing bubbles can partially disrupt casein micelles (particularly when applied at elevated pH), which increases the surface area for gelation, enhancing the rennet and acid gelation kinetics [80, 81]. Liu et al. [80] have investigated the effects of ultrasound processing on the physicochemical properties of casein micelles in reconstituted skim milks and have shown that ultrasound at 20 kHz can disrupt casein micelles and reform micelle-like particles with smaller size and comparable ζ-potential to native micelles. In addition, the volume of the casein micelles that are soluble in the serum increased significantly after ultrasound treatment. As a result, the sonicated milk was able to shorten the cheese renneting time compared to untreated milks. Other ultrasound interventions integrated in dairy processing lines can potentially provide modified physicochemical properties of milk proteins and achieve desired functional properties.

Meat Product Modification The quality of meat is dependent upon a complex interaction between biological traits and biochemical processes during the conversion of muscle to meat. For beef, consumer behaviour indicates that tenderness is the most important sensory property in grading its meat quality [51, 118]. Much interest is therefore targeted at understanding how tenderness can be improved in lower grade cuts of meat such that it can be sold at a premium.

Meat is a complex and highly organised tissue, built up of individual cells (fibres) that are held together by connective tissue. Each muscle fibre in meat is composed of myofibrils, about 1–2 μm in diameter, which consist of thick (myosin) and thin (actin) myofilaments that give muscle its striated appearance. The structural characteristics essentially determine the texture and consequently the tenderness of the meat [140]. To improve tenderness, the meat industry typically ages meat by storing at 0–4 °C for up to 2 weeks (wet ageing). Dry ageing in humidity-controlled storage is also performed but creates a product that has reduced yield due to loss of moisture [132]. The storage time tenderises meat due to several factors that occur during postmortem proteolysis, which degrades myofibrillar proteins, causes contraction of the muscle and reduces the amount of connective tissue. The combined effect of these factors in postmortem muscle results in weakening of the myofibrillar matrix through degradation of key structural proteins, which in turn, results in an improvement in tenderness [132].

The effect of ultrasound on the textural quality of meat is dependent on the species and cut of the meat, and the ultrasonic conditions applied for the treatment (i.e. intensity and frequency). In many reports, one or both of these parameters are not stipulated, making meaningful comparison difficult and leading to some contradictory results in the literature on the efficacy of ultrasound for the tenderisation of meat [132].

Beneficial effects on the texture of post-rigour meat with the application of ultrasound have been reported in many studies. It has been hypothesised that the mechanisms of tenderisation are (1) physical disruption of the tissue caused by cavitation [52, 58] and/or (2) release and activation of enzymes [82, 114].

Smith et al. [124] applied high-intensity, low-frequency ultrasound (1000 W, 26 kHz, 2–4 min) to beef cuts (*M. longissimus thoracis et lumborum* and *M. semitendinosis*) and found that the treated muscle was more tender than untreated muscle. Jayasooriya et al. [59] also applied high-power, low-frequency ultrasound (12 W/cm^2, 24 kHz, 0–240 s) to these same cuts of beef and found increased tenderness without needing to age the meat for extended periods of time. These samples had a reduced cook loss, but no effect was observed on colour indicating negligible reduction in visual product quality. Chang et al. [21] showed that meat hardness decreased with 10 min of ultrasound exposure. In addition, ultrasound treatment reduced muscle fibre diameter and the thermal stability of collagen. SEM images showed disordering of collagen fibres resulting in a looser arrangement (Fig. 4). It was suggested that the improvement in texture was due to the effect of ultrasound on the collagen characteristics.

Another study investigated the effect of ultrasound on the microstructure of beef *M. semimembranosus* and showed that ultrasound caused disruption of the muscle proteins within the myofibrillar structure. It was suggested that these changes caused an acceleration of ageing in these samples [126]. For other types of meats, increased tenderness has also been reported in other species with the application of low-frequency ultrasound, including cobia [20], squid [50] and chicken [33, 156].

Some studies using low-intensity ultrasound have reported no beneficial effect on meat texture. Low-intensity ultrasound baths were found to produce no effect on tenderness [82]. Similarly, no tenderisation was reported when low-intensity ultrasound was applied to beef *M. longissimus thoracis et lumborum* and *M. semimembranosus* [84] and lamb *M. longissimus thoracis et lumborum* [83]. Using high-frequency ultrasound (600 kHz, 10 °C, 10 min), Sikes et al. [122] again reported no effect on the texture of postrigor beef *M. longissimus dorsi*. These observations indicate that the beneficial effect of ultrasound depends on the intensity of the process, i.e. there is some threshold of intensity required for structural modification to occur.

There is a promising future in the application of ultrasound for improving the texture of meat. However, for this to be an effective and efficient process in commercial operations for tenderisation of meat, the ultrasound conditions need to be further optimised on a case by case basis.

Vegetable Material Modification The plant cell wall is a highly complex structure composed of the three main polysaccharides cellulose, hemicellulose and pectin. These polysaccharides are associated with structural proteins and phenolic compounds to form the underlying building blocks in plant-based products. The phenolic compounds in the cell wall of fruits and vegetable cells are mainly phenolic esters, such as cinnamic acids, that are found in the cell wall of many vegetables attached to wall polysaccharides [54, 129, 149, 150].

Several structural features contribute to the textural properties of plant-based foods [5]. A force, known as the turgor pressure, exists across the membranes of plant cells due to the presence of sugars and salts in the cell's vacuolar fluid. This osmotic pressure is balanced by flow of water into the cell until the static pressure difference is equalised. Secondly, the cell wall rigidity and cell-cell adhesion are determined by the integrity of the middle lamella and plasmodesmata. The plasmodesmata are small channels that traverse the plant cell wall to provide a cytoplasmic pathway for communication between adjacent cells. The middle lamella is largely composed of a pectin matrix. The cell-to-cell adhesion and rigidity gives firmness and elasticity to the plant tissue [40].

The three main polysaccharides in the primary cell wall respond differently to processing, giving rise to two components of the firmness of plant materials [34]. Generally, the structure derived from the hemicellulose-cellulose network remains largely unaffected by processing, while the pectin component is affected both by enzymatic and non-enzymatic reactions [130], and these lead to effects on the strength of the primary cell wall and cell-to-cell adhesion.

Ultrasound processing may affect the structure and texture of fruits and vegetables in several ways, depending on the type of tissue and the processing conditions. The application of low-frequency, high-power ultrasound may cause significant tissue disruption, resulting in loss of turgor pressure and softening of plant tissue due to the extreme conditions that

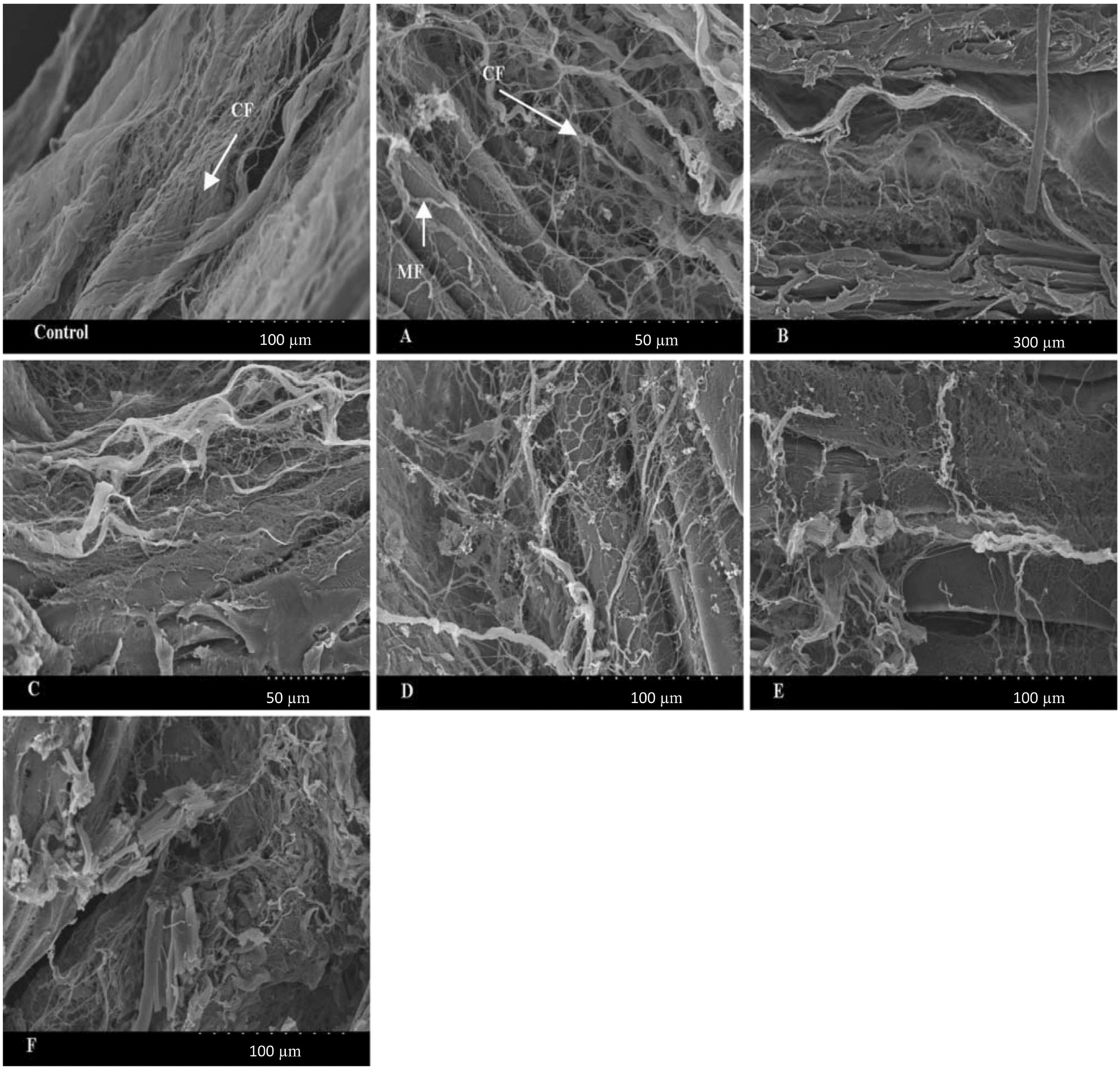

Fig. 4 Scanning electron microscopy of collagen fibres in beef semitendinosus muscle for controls compared with **a** 10, **b** 20, **c**30, **d** 40, **e** 50 and **f** 60-min ultrasonication time. With permissions from Chang et al. [21]

accompany cavitation bubble collapse. Although the process may also result in inactivation of texture-degrading enzymes such as PME (pectin methylesterase) and PG (polygalacturonase) [121, 131], the intense mechanical disruption of plant tissue makes the application of power ultrasound unsuitable for processing whole and sliced fruit and vegetable products.

The use of low-intensity ultrasound as a pretreatment step has been shown to improve the structure and texture of a number of products, including potatoes [29, 129] and carrots [30], although the mechanisms responsible for these modifications are not well understood. It is speculated that the observed effects could be attributed to stress elicitation, activation of the PME catalysed de-esterification of pectin, the activation of the peroxidase-catalysed oxidation of cell wall structure proteins and phenolics by ultrasound-induced production of hydrogen peroxide, or a combination of two or more of these mechanisms. Ultrasound may also activate the peroxidase-catalysed oxidation of cell wall structural proteins and phenolic acids, particularly in the medium frequency range (300–400 kHz), as this results in an optimal generation of sonochemical radicals [7].

Curulli et al. [29] describes an ultrasonic surface texture modification process suited for potatoes and similar starchy vegetables prior to further processing, primarily for French fries production. The ultrasound treatment was performed as a pretreatment to the blanching step in the standard French fries production process, which was followed by partial drying, par frying, blast freezing and final frying steps. Ultrasound treatment at (40 + 380 kHz) and 65 °C for 11 min resulted in a 50% improvement in crunchiness and crispiness as measured by acoustics and a 45% improvement as measured by sensory analysis compared to the untreated control. A 35% improvement in crunchiness and crispiness was observed in samples treated at a single frequency of 40 kHz and 40 °C for 6 min. In all cases, the surface cell wall composition showed moderate to high pectin levels, low residual protein and negligible surface sugar and starch remaining in the ultrasound-treated samples compared to the low pectin and moderate residual protein, starch and sugar on the surface of the cell wall of the conventionally processed French fries. The observed change in texture was related to the change in the surface composition of the product and modification of the cellular structure on the surface [29].

Day et al. [30] studied the effects of low temperature blanching with ultrasound (mixed frequency (40 + 400 kHz), 60 W/L, 34 and 60 °C, 5 and 10 min) on the texture of carrots subjected to retorting (treatment time at 121.1 °C, F_O = 10 min). Firmness of ultrasonically treated samples (10 min, 60 °C) prior to low-temperature blanching displayed threefold increase in firmness compared with untreated controls. Samples subject to ultrasonic treatment at these conditions also resulted in higher tensile strength (twofold increase relative to controls). The microstructure of the samples imaged by confocal microscopy (Fig. 5) showed substantial cell deformation and separation in the untreated samples, whereas limited deformation and cell separation were observed in the ultrasonically treated samples [30]. Possible reasons suggested for the ultrasonic enhancement could be due to (1) gradual decrease of turgor pressure initiated at the early stage of ultrasound application, reducing the sudden impact of turgor loss on cell wall structure; (2) enhanced mass transfer and enzyme-substrate interaction, facilitating the rate of pectin demethylation and diffusion rate of divalent cations such as calcium; and (3) stress response reactions, such as the formation of phenolic cross-linking in response to ultrasonic stress [30].

The consistency and cloud stability of fruit and vegetable purees and juices is also affected by physical and chemical changes in the structure of pectins. Pectinaceous substances in such products serve as a continuous phase in which other particles are suspended [144]. Juice cloud is an important quality attribute in products such as orange juice, contributing to flavour, aroma and colour, and consumers usually associate cloud loss with spoilage and quality degradation [8]. Fruit juices are colloidal systems consisting of a liquid phase, termed the ‘serum’, and a solid phase, termed the ‘cloud’. The cloud is stabilised by the soluble pectin in the juice [8, 18]. In tomato juice, the modification of the pectin structure during processing by chemical conversion (β-elimination) leads to loss of consistency and phase separation [145].

Thermal processing is commonly used to inactivate enzymes during the crushing stage of juice production to minimise pectin degradation. The application of high-intensity ultrasound can indirectly affect the structure and rheological properties of fruit and vegetable juices through inactivation of pectin-degrading enzymes, such as PME and PG at mild temperature [2, 107, 131, 136, 144, 152]. This reduces pectin degradation and improves the consistency and cloud stability of the products during storage. In addition to its effect on PME and PG and pectin biochemistry, ultrasound can also directly modify the physical structure of pectin, leading to a reduction in particle size [136, 152], increase in viscosity [144, 152], juice cloud [24, 136] and juice cloud stability during storage [137]. The application of ultrasound can cause structural modification and breakdown of linear pectin chains as a consequence of damage from microjets generated during asymmetric collapse of cavitation bubble [119, 136]. Furthermore, microstreaming accompanying cavitation and shear stress produced by ultrasound waves can cause degradation of macromolecules even in the absence of bubble collapse [105].

Wu et al. [152] observed a significant reduction in particle size distribution and substantial increase in viscosity following thermosonication (i.e. combined heating and sonication) treatment of tomato juice (24 kHz, 60–70 °C, 480 W/L). An 8-min ultrasonic treatment at 65 °C resulted in the reduction of the mean particle size in the juice from 200 to 15 μm, which was accompanied by a onefold increase in the apparent viscosity of the juice. The observed particle size reduction facilitated by ultrasound was attributed to the physical effects of cavitation and shear stress induced by ultrasonic waves. The higher viscosity in the sonicated samples was attributed to enhanced inactivation of PME and PG as well as the reduced particle size, which yields a larger interfacial area and results in stronger interparticle interactions [152].

Tiwari et al. [136] reported a significant increase in the cloud of orange juice subjected to ultrasonic treatment at 20 kHz and 420–1005 W/L for up to 10 min. The juice cloud of the ultrasound-treated samples resulted in a twofold increase compared with an untreated sample, using a power input of 420 W/L. The observed increase in juice cloud was attributed to the effects of asymmetric cavitation during sonication on the structure and particle size distribution of the pectin matrix, altering its tendency to form a precipitate and settle down. The particle size distribution of the juice prior to and after ultrasound treatment at different conditions showed significant particle size reduction following ultrasonication [136]. Improved juice cloud stability during storage has also been reported for sonicated orange juice [137], which can be attributed to the same effect.

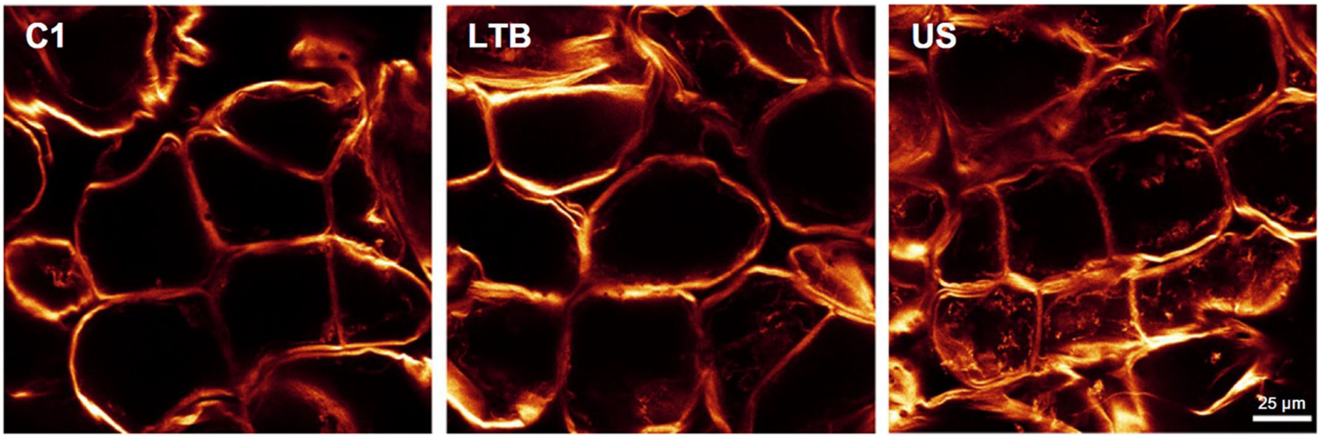

Fig. 5 Micrographs of carrot microstructure for C1 control, LTB low-temperature low-time blanching and US low temperature with US application. Sourced with permission from Day et al. [30]

Polysaccharide-rich fluids such as those containing starch, dextran and pectin, can also undergo gelatinisation due to heating in the presence of water, transforming the aqueous suspension into an amorphous gel phase. Iida et al. [53] extensively studied the effects of ultrasonication on starch solutions after gelatinisation. It was found that sonication for 30 min could reduce the viscosity of starch solutions (5–10 wt%) by 2 orders of magnitude, to ~100 mPa s. High concentration starch gels (20–30 wt%) could even become liquefied after treatment with ultrasonication. This liquefication and viscosity reduction can be useful as a pretreatment of viscous starch solutions to increase the efficiency of spray drying [53].

Homogenisation in Dairy Processing

Dairy products such as milk, yoghurt and ice cream are homogenised to improve the product stability against creaming during storage. In recent years, the use of ultrasound has attracted much interest for the homogenisation of milk [4, 15, 35, 151].

Koh et al. [69] investigated the homogenisation of whey protein concentrate (WPC) systems using ultrasonication and high-pressure homogeniser to investigate the particle size reduction in the presence/absence of cavitation. The size reduction of particles in WPC systems formed using a high-pressure homogeniser showed comparable results to those formed using ultrasound under similar energy conditions. Notably, cavitation-derived radicals were found to be absent in high-pressure homogenisation, indicating that the primary mechanism in high-pressure homogenisation is due to shear forces formed in the absence of cavitation. In the case of ultrasound application, the shear forces are resultant from strong bubble collapse that also form chemical radicals.

The frequency, amplitude and diameter of the ultrasound applied can have a significant influence on the physical properties and variance of milk that is homogenised. Bermúdez-Aguirre et al. [12] showed that ultrasound (400 W, 24 kHz, using a 22 mm probe) applied to milk at 63 °C for 30 min caused disintegration of the milk fat globule membrane. The size of the milk fat globules was reduced to <1 μm compared to native fat globule dimensions of 4.3 μm. Furthermore, a granular surface morphology was formed due to the interaction of fat with casein micelles. Villamiel and de Jong [147] reported similar size reductions using continuous flow, high-intensity (150 W, 20 kHz, using a Branson sonifier with 18.76 mL cavity) ultrasonication of milk. At temperatures of 70 or 75 °C, a monomodal particle size distribution was obtained, whereas a bimodal distribution was observed at lower temperatures. Bosiljkov et al. [15] showed that an increase of the amplitude (20, 60 and 100%) and time (2–15 min) of ultrasound (30 kHz, using 7 and 10 mm probes) significantly influenced the degree of homogenisation of milk.

Wu et al. [151] reported that high-amplitude ultrasound (90, 225 and 450 W, 20 kHz) not only effectively homogenised milk but also improved the viscosity and water holding capacity. Yoghurts produced from ultrasonically homogenised milks were found to have reduced syneresis. These effects can be explained by the yoghurt structure which consists of strings or clusters of casein micelles interacting physically with each other and with denatured serum proteins entrapping serum and fat globules. The homogenisation of the fat by ultrasound causes minor denaturation of the proteins and the reduced size of the fat globules increases their ability to interact with themselves and/or casein micelles.

Emulsification

Emulsification is the process of dispersing two or more immiscible liquids, typically oil and water, together to form a semi-stable mixture. Common food emulsions include mayonnaise, sauces and spreads. Emulsions are thermodynamically unstable mixtures and usually require the addition of surface active materials, known as emulsifiers, to stabilise the formed interface between the oil and water [67]. One of the attributes that govern the stability, appearance and taste of a food emulsion is the emulsion droplet size (EDS) and size distribution. Whereas emulsions with droplet sizes >100 nm display a 'milky' opaque appearance, emulsions with smaller EDS (~100 nm) can appear translucent and almost clear [79]. This can be used to create drinks that have a more 'attractive' appearance. The formation of nanoemulsions is of high interest to the food industry as it is recognised as an efficient delivery method for lipophilic bioactives [89], such as omega 3 fatty acids, carotenoids and phytosterols.

The creation of small droplets is facilitated by the application of high-energy shearing. The techniques available for emulsification include rotor-stator devices, high-pressure homogenisation, microfluidisation and ultrasonication. It is commonly recognised that the Microfluidizer™ is the most effective nanoemulsion preparation technique currently available [56], with relatively high energy efficiency for producing emulsions with very small and narrowly distributed EDS. Microfluidisation is however quite expensive to operate and so is less suited for production of high-volume, low-value products such as food emulsions. A significant advantage that ultrasonication has over microfluidisation is its relative ease of use in regards to cleaning and maintenance [57]. Jafari et al. [56] have compared emulsion preparation using ultrasound and microfluidisation at matched energy input (20 kJ/kg) and found comparable performance. Particle size reduction by Microfluidizer achieved mean volume-weighted particle size of 0.83 μm compared with 1.02 μm for ultrasonication at 20 kHz.

Ultrasonic emulsification is proposed to be due to two mechanisms [67]. Firstly, the sound field produces interfacial waves that become unstable as they propagate through the fluid that results in the eruption of the oil phase into the continuous water phase to form mid- to large-sized droplets. Secondly, the collapse of cavitation bubbles generates high shear and turbulence which act to break up these initially formed droplets of dispersed oil into droplets of sub-micron size [133]. The first mechanism is only relevant when the continuous and dispersed phases are separate prior to the beginning of emulsification. Once a coarse emulsion is formed, further droplet size reduction occurs by the shearing forces from acoustic cavitation.

Competing mechanisms are at play during ultrasonic emulsification that may result in droplet re-coalescence and hence an increase to the EDS. Ostwald ripening, Brownian motion and droplet collisions all contribute to an increased likelihood of droplet coalescence. Coalescence during emulsification can be reduced by ensuring fast stabilisation of the new interfaces by having sufficient and appropriate surfactant molecules present in the mixture or by increasing hydrodynamic effects such as retardation of liquid drainage between two colliding droplets [55].

Ultrasonication has potential to reduce or even eliminate the requirement for using synthetic emulsifiers to stabilise an emulsion. Shanmugam and Ashokkumar [120] have shown that ultrasonic emulsification can be used to create stable food-based emulsions of flax seed oil in skim milk, without requiring any additional synthetic surfactants. Stability was instead inferred by the native milk proteins which are partially denatured (less than 1%) by the ultrasound. This enabled them to effectively coat the formed oil droplets, stabilising the emulsion for at least 7 days. By comparison, stable emulsions could not be formed in the absence of ultrasound even when using matched applied energies in a rotor-stator system, suggesting the importance of acoustic cavitation to the stabilisation process. Recently, Leong et al. [78] demonstrated the ability to form what are known as double emulsions using ultrasonication, again using the native milk proteins to stabilise the external interface. These emulsions (Fig. 6) were able to encapsulate an inner aqueous phase within the oil droplets and could be a viable delivery vehicle for encapsulating aqueous-based bioactives in foods.

Although ultrasonic emulsification is typically achieved using low frequency (i.e. 20–100 kHz) ultrasound, higher-frequency ultrasound (400 kHz–3 MHz) has also been reported to be useful at forming nanosize emulsion droplets when applied following low-frequency ultrasound through a process known as *tandem acoustic emulsification* [66, 97]. The emulsions formed using this process were found to be stable for 1 to 2 years and, notably, were formed without requiring any additional surfactant. Oleic acid/water nanoemulsions were prepared by Kamogawa et al. [66] using this technique, while Nakabayashi et al. [97] also reported the production of transparent emulsions of ethylenedioxythiophene (EDOT) monomer, formed by sequential emulsification at 20-kHz, 1.6-MHz and 2.4-MHz ultrasound.

It was proposed that the small droplets formed using tandem acoustic emulsification with high-frequency ultrasound was due to the enhanced acceleration of solvent and the emulsion droplets caused by acoustic radiation forces and acoustic streaming [66] such that they collide together and break apart into smaller droplets. These acceleration forces become stronger with increased frequency, and the sequence in which the different frequencies of ultrasound are applied is noted to be very important. A reversal of the order in which the ultrasonic frequencies is applied (i.e. high frequency followed by low frequency) resulted in ineffective emulsification.

Recent research in ultrasonic emulsification has highlighted the capability of using ultrasonically produced nanoemulsions to deliver essential oils into food or pharmaceutical-based products. Essential oils have a wide range of beneficial properties such as high nutritional value [123] and antimicrobial properties [94], but can be challenging to incorporate stably into aqueous-based food products due to the hydrophobic nature of the oils. Ultrasonication

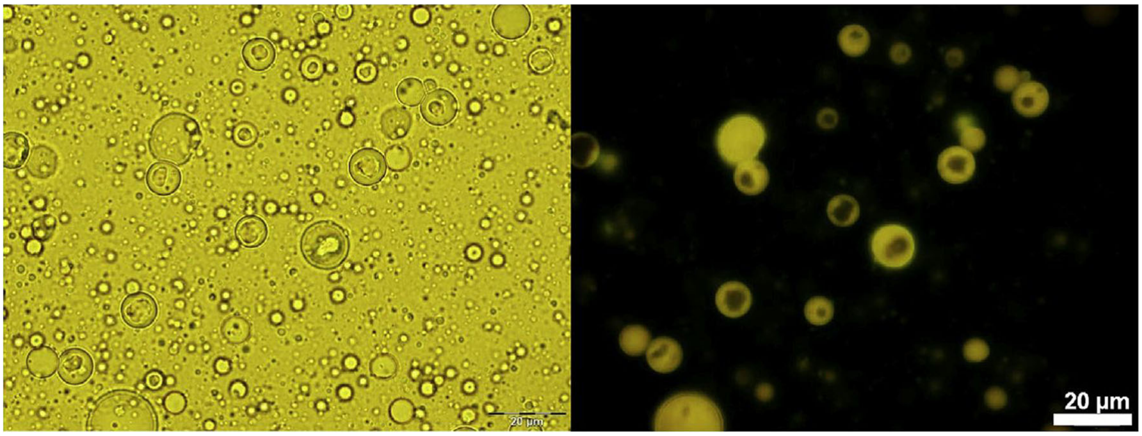

Fig. 6 Double emulsion of skim milk encapsulated within sunflower oil dispersed within skim milk, formed using ultrasonication. Reprinted from Leong et al. [78], copyright 2017, with permission from Elsevier

has been used as an effective method to create stable nanoemulsions using essential oils derived from basil [42], orange peel [48, 49], annatto seed [123] and *Thymus daenensis* [94]. These nanoemulsions were found to be exceptionally shelf stable for many months, and also found to possess good to excellent antimicrobial activity that were superior to the oils in their native state [94]. A possible reason for this increased activity was because the nanoemulsions enabled the essential oils to come nearer to the bacterial cell membrane interface, enabling more effective disruption of the phospholipid bilayer.

Extraction

A key source of essential oils, aromas, pigments, antioxidants and other bioorganic compounds are natural foods such as herbs and vegetables, most of which are often locked within the cell walls of the material. These compounds can be extracted selectively from foods to produce valuable nutraceuticals. Extractions are performed using polar or non-polar solvents, depending on the nature of the material being extracted. Oil-based extractions employ hexane and mixtures of hexane with isopropanol [127] while polar compounds are extracted using solvents such as water, ethanol, methanol or acetone. Solvent extractions can take many hours or even days to complete, and often large amounts of solvents are required. The solvents used must eventually be disposed of or regenerated using high temperatures, creating a large environmental footprint.

Supercritical fluid extraction is an alternative technique that can be used for such extractions [23]. CO_2 is a widely used supercritical extraction solvent, because it is non-toxic, non-flammable, cheap and easily removed after extraction. It also has high solvating capability for non-polar molecules [23]. While supercritical CO_2 extraction is highly effective and can eliminate requirement for toxic solvents, it is an energy demanding and expensive process that requires complex pressurised equipment.

Ultrasound can be used to enhance and accelerate the extraction of food compounds using solvent and supercritical extraction processes. Mechanistically, the enhancement is due to increase of heat and mass transfer between the solvent and the solid boundaries of the food material by physical shearing effects resultant from cavitation, such as microjetting and pressure shockwaves. The release of extractable compounds can be enhanced due to disruption of the cell walls of the food material, accelerating the movement of compounds into the solvent phase.

Ultrasound in the 20–100-kHz region is known to be most effective for extraction. The outcome of ultrasound application is often higher extraction yields, reduced extraction time and lowered solvent consumption [127]. Extractions requiring many hours can be completed reproducibly using ultrasonication in a manner of minutes. Ultrasonic extraction can also enhance the effectiveness of solvents, so that safer less toxic solvents (e.g. water) can be used instead of organic-based solvents. This is a possible strategy to reduce environmental impact and costs [19].

There exists a vast number of food materials with which ultrasound [146] is able to accelerate extraction and improve yields, including herbs, crushed fruit pomace and oil seeds from which products such as polyphenolics, anthocyanins, aromatic compounds, polysaccharides and functional compounds can be extracted. Wu et al. [154] achieved a threefold increase extraction rate of saponins from ginseng. Vinatoru [148] achieved increased yields of up to 34% compared to conventional stirred reactors for extraction of extracts from fennel, hops, marigold and mint. Balachandran et al. [9] used ultrasound as a pretreatment for supercritical CO_2 extraction, improving extraction yield by 30% and reducing required extraction time.

Valuable compounds such as polyphenols can be recovered from waste materials. Grape marc, which is a waste stream from the wine industry, was used by Vilkhu et al. [146] to extract phenolics. A reported increase in yield of 17–35%

was achieved when ultrasound was used. Mechanistically, in all of these examples, the application of ultrasound interacts with the plant materials to alter its physical and, in some cases, chemical properties. Further examples of applications and methods of application are reported in Table 1. Recent work has also demonstrated increased antioxidant activity in compounds from bitter gourd [47]. A review in ultrasound-assisted carotenoid extraction from algal and seaweed products has recently been published [104].

Operating temperatures required for effective extraction is application dependent. Generally, an increased temperature increases the softening and swelling of the raw materials and also increases the solubility of the desired compounds to be extracted in the solvent. In the context of ultrasonication, temperatures cannot be raised too high, as the vapour pressure of the cavitation bubbles may also increase, resulting in a significant decline in the cavitation intensity. Ultrasound is therefore more effective when used for enhancing extractions at lower temperatures where the energy generated upon cavitation bubble collapse is greater [19].

High-Frequency Ultrasound Standing Waves (Megasonics)

Megasonics, operating at frequencies at or above 400 kHz, has been commercially applied mainly for cleaning of fine structures, e.g. in the semiconductor industry. This is where the development of these systems at larger scale and relatively high powers was pushed over the last decade. Only with the availability of larger scale systems has it become feasible to conduct studies related to food processing. The researchers at CSIRO have recently spent considerable effort into developing a basic understanding of the mechanisms involved in utilising high-frequency ultrasound for various applications, such as enhanced separation and texture modification of processed foods.

Megasonic Milk Fat Separation

Milk fat is a valuable commodity, so a common practice in the dairy industry is to remove all of the fat from the milk, and then recombining the separated cream as required back into skimmed milk to make products with standardised fat concentrations. This separation is typically achieved by using a centrifuge, a high-energy demanding process, which removes the fat from milk by application of high *g*-forces by rapid spinning. Traditionally, separations were performed simply by letting the cream in milk rise to the top where it could be skimmed off. This is however a slow process, requiring many hours to achieve sufficient separation. Cheese makers in Northern Italy famed for Parmesan and Romano cheeses [17] continue to apply natural gravity separation methods, since it creates milk with a fat size distribution that contributes to the unique flavours in these cheeses and creates a point of differentiation that cannot be achieved using centrifugation [85]. Ultrasonic separation has been recognised as a complementary technology that can significantly enhance the separation rate of fat from milk by 'natural' methods [74].

When an acoustic standing wave is applied to whole milk, the fat globules collect at the pressure antinodes, which can be observed as 'bands' of fat globules as observed by Miles et al. [92] in a small cuvette container. The milk fat globules collecting and concentrating at the pressure antinodes have an increased probability to aggregate into larger floccules. As fat globules collect into larger flocs, they begin to rise rapidly to the surface due to the increased hydrodynamic radius as described by Stokes' law [71]. This enhances the 'natural' tendency for fat globules to separate due to buoyancy.

Recent research has shown that high-frequency ultrasound (400 kHz to 3 MHz), can enhance milk fat separation in small-scale systems able to treat a few millilitres of sample (e.g. [45]). Juliano et al. [64] have investigated the effect of ultrasonic standing waves on milk fat creaming of a recombined coarse milk emulsion in a 6-L reactor and the influence of different frequencies and transducer configurations in direct contact with the fluid. Runs were performed with one or two transducers placed in vertical (parallel or perpendicular) and horizontal positions (at the reactor base) at 400 kHz, 1 MHz and/or 2 MHz. They found most efficient creaming after treatment at 400 kHz in single and double vertical transducer configurations.

The parameters found to achieve rapid separation of a recombined milk emulsion, however, were not successful in separating fat globules in natural whole milk [128]. Several reasons can be attributed to this. The particle size distribution and surface composition of whole milk are different to a recombined milk emulsion. The smaller-sized droplets in whole milk require high-frequency ultrasound or strong acoustic energy density to manipulate and separate effectively. Effective application of high-ultrasound frequency >1 MHz requires a geometry such that the distance between transducer and reflector is small to minimise attenuation.

Leong et al. [74, 76] used these concepts to achieve effective separation of cream from natural whole milk without significant impact on milk quality when using 2 MHz or when using very high specific energies >230 kJ/kg [60, 65]. Leong et al. [74] showed that higher-frequency ultrasound, in this case >1 MHz, could effectively manipulate and separate the fat globules present in natural whole milk provided that a short transducer-reflector separation distance (between 30 and 85 mm) was used. Furthermore, the application of two transducers arranged in a parallel setup could influence more rapid skimming of fat due to the ability to achieve a higher energy density per unit volume. The same concepts can be applied to promote the separation of fat from whey, as demonstrated by Torkamani et al. [139].

Table 1 Ultrasonic extraction applications

Product	Ultrasound delivery	Solvent	Performance outcomes	Reference
Almond oils	Batch, 20 kHz	Supercritical CO_2	30% increased yield or extraction time reduction	Riera et al. [109]
Herbal extracts (fennel, hops, marigold, mint)	Stirred batch, 20 to 2400 kHz	Water and ethanol	Up to 34% increased yield over stirred	Vinatoru [148]
Ginseng saponins	Batch, 38.5 kHz	Water, methanol and *n*-butanol	Threefold increase of extraction rate	Wu et al. [154]
Ginger	Batch, 20 kHz	Supercritical CO_2	30% increased yield or extraction time reduction	Balachandran et al. [9]
Soy protein	Continuous, 20 kHz, 3 W per gramme	Water and sodium hydroxide	53 and 23% yield increase over equivalent ultrasonic batch conditions	Moulton and Wang [95]
Soy isoflavones	Batch, 24 kHz	Water and solvent	Up to 15% increase in extraction efficiency	Rostagno et al. [115]
Rutin from Chinese Scholar Trees	Batch, 20 kHz	Water and methanol	Up to 20% increase in 30 min	Paniwnyk et al. [101]
Carnosic acid from rosemary	Batch, 20 and 40 kHz	Butanone and ethyl acetate	Reduction in extraction time	Albu et al. [3]
Polyphenols, amino acid and caffeine from green tea	Batch, 40 kHz	Water	Increased yield at 65 °C, compared with 85 °C	Xia et al. [155]
Pyrethrines from flowers	Batch, 20 and 40 kHz	Hexane	Increased yield at 40 °C, compared with 66 °C	Romdhane and Gourdon [113]

Adapted from Vilkhu et al. [146], copyright 2008, with permission from Elsevier

Leong et al. [76] also showed that there is a temperature range over which the rate of milk fat separation is highest. A temperature range between 20 and 60 °C was found to offer fast separation of milk fat in the experimental trials. Leong et al. [76] recently highlighted the ability of ultrasonic separation to initiate fractionation of milk fat globules into streams with enhanced proportions of small or large fat globules. It was established that the milk fat became distributed in the separation vessel after sonication, such that the smallest fat globules were retained towards the bottom of the vessel, and large-sized fat globules were enriched within the cream collected near the top of the vessel. These fractions positioned at the top and bottom of the vessel can be collected specifically by overflow/underflow. These collected streams may have potential for creating new types of dairy products with modified microstructure that are creamier and tastier.

Enhanced Vegetable oil Recovery

To produce olive oil, whole olive fruits are first crushed to form a paste. The paste is pumped into a vessel controlled at 23–30 °C where it is slowly macerated by a set of kneading blades rotating at 20–30 rpm, to facilitate further disruption of cell tissues through the action of natural enzymes. The residence time and temperature of the paste during malaxation will affect the amount of oil released. Generally, a longer time and higher temperature exposure will release more oil [25].

A critical factor to consider in the malaxation process is oil quality, which decreases with an increase in malaxation time and temperature, promoting oxidative reactions and increasing the solubilisation of antioxidant polyphenols into the water phase and away from the oil phase. Co-adjuvants may be added during malaxation to enable more rapid or increased oil yields; these may include talc to reduce the moisture content in the paste or enzymes to promote cell wall breakage and release of oil bodies during the malaxation process. The final step of olive oil separation is to pump the malaxed paste into a decanter centrifuge where the oil is recovered from the paste.

Megasonics can be applied to the olive paste at three stages of the olive oil process: (a) before malaxation, (b) during malaxation and/or (c) after malaxation [62] (Fig. 7). Juliano

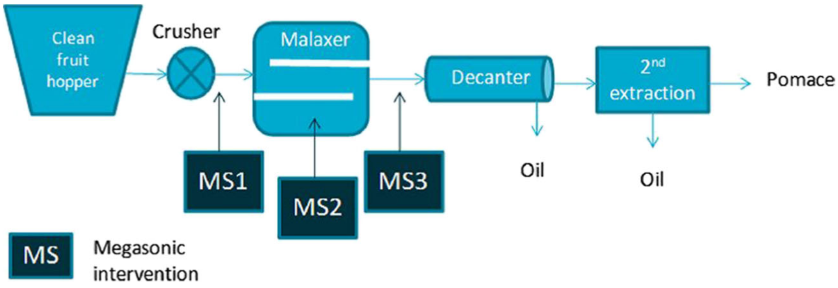

Fig. 7 Megasonic interventions during the olive oil extraction process. Sourced with permission from Juliano et al. [62]

et al. [62] demonstrated the benefits of applying megasonics before malaxation and after malaxation at laboratory scale with extractability improvements between 2 and 3%. They showed the benefits of megasonic post-malaxation with 300 kg batches and water addition with extractability improvements near 1.2–1.5%.

Much effort has been made to investigate the effects of low-frequency ultrasound on olive oil quality and extractability of other components such as chlorophyll, carotenoids, tocopherol compounds and phenolic compounds [1, 26, 27]. Trials conducted in an ultrasonic bath (2.5 kg paste) at 35 kHz, 150 W up to 10 min and 30 °C showed improvements in the antioxidant content of virgin olive oils from two varieties as seen by increased tocopherols and carotenoids. However, ultrasound promoted additional chlorophyll release and decreased polyphenol concentration, with positive effect on sensory properties. Further research is required to understand extractability of components after megasonic treatment and its impact on sensory properties and nutrition.

Palm oil In the palm oil milling industry, depending on the efficiency of the mill, a substantial part of the contained oil can be lost in the effluent stream. Therefore, an intervention at any processing stage that increases recoverable oil and reduces oil in the discharged palm oil mill effluent is expected to improve palm milling performance. Juliano et al. [63] have examined the effects of applying ultrasound on the oil recovery from the ex-screw press feed and the underflow sludge from a palm oil vertical clarification tank to determine the usefulness of an intervention based on ultrasound. Megasonics was applied at frequencies of 400 kHz to 1.6 MHz in a standing wave field and the effects on two process streams (containing oil, non-oil solids and water) in palm oil milling were examined. Ultrasonication of the ex-screw press feed obtained upon crushing of the sterilised palm fruit and of the underflow sludge from the vertical clarification tank enhanced oil separation on gravity settling. Megasonics also enhanced the total oil recoverable, which consisted of the sum of the oil separated under gravity and the decantable oil separated upon centrifugation of the remaining fraction. It was concluded that ultrasound-assisted separation of oil from process streams was attributed to the acoustic forces exerted on the suspended particles in the feed, similar to the effects in milk fat separation, which causes the oil to migrate to the antinodes and non-oil solids (comprising vegetal matter and residual oil) to the nodes. This work demonstrated the potential of applying high frequency ultrasound to improve the separation of oil in the clarification tanks and reduce oil that is lost in the non-oil fraction from the separators (i.e. sludge underflow). This application represents a step-change innovation in palm oil milling operations to reduce oil loss during milling.

Coconut oil Coconut oil is mostly produced by a solvent extraction process. Coconut endosperm is firstly removed and dried to produce the dried coconut meat, and then subsequently extracted using solvents. The use of organic solvents is expensive and carries safety risks and environmental concerns; however, it achieves higher oil yield than aqueous-based extraction methods. There is a shift in interest towards development of aqueous based extraction as it provides the opportunity to more optimally utilise food and non-food materials in the coconut towards the production of value added bioproducts. Full utilisation and conversion of this material into high value products could provide greater returns and also minimise contamination due to biological and solvent residues.

Preliminary work at the CSIRO has evaluated potential megasonic intervention in the coconut oil extraction process in aqueous media. High-frequency standing waves can be applied either to the coconut meat during digestion or applied to the coconut milk. After 10-min sonication of a coconut meat/water mixture, it was found that high-frequency 2-MHz standing waves enhanced coconut oil yield more ($P < 0.05$) than those at 600 kHz and the non-ultrasound control [61]. The sample treated with megasonics had a dense cream layer that was clearly separated from the sediment when compared to the control (without megasonics) where separation was less evident. Further work on coconut milk extracted from coconut meat (following a thermal and filtering process) showed the ability of 2-MHz standing waves to increase oil yield. The next steps will include the testing of this system in combination with small-scale centrifugal separators to understand its direct scalability.

Design Aspects of Ultrasonic Reactors

The effectiveness of ultrasonic reactors is underpinned by the ability of the sound waves to provide uniform treatment of material within its confines. The reactor geometries and placement of transducers are application dependent and requires consideration of the spatial and temporal contact arrangement between the ultrasonic transducers used and the food material, as well as how the acoustic wave penetrates the medium. A critical factor to be considered is that ultrasonic waves becomes attenuated and reduced in intensity with distance from the transducer. The design of sonoprocessing and sonochemical reactors have been reviewed in detail by Gogate et al. [43, 44] and so will not be covered in detail here. Recent publications have reviewed the design considerations for large-scale megasonic separation systems [63, 74, 77, 141, 142]. Some of the key parameters for successful designs for megasonic separation, such as the positioning, arrangement and alignment of transducers within a reactor, are summarised in Table 2 and will be touched upon briefly.

The maximum effective distance in which a standing wave field can be formed is limited by attenuation of the sound wave in the fluid medium. The higher the ultrasonic frequency, the more strongly attenuated it becomes. Ultrasound in the MHz region is generally limited to shorter effective working distances to achieve a strong standing wave. Strong attenuation may also give rise to undesirable acoustic streaming in the form of Eckhart streaming currents, which can cause disruption to the separation of particles aligned within the nodal/antinodal planes of the standing wave.

The transducer-reflector distance is therefore a critical design consideration, as separation with high frequencies will not operate effectively if the incident sound wave becomes too attenuated before it can be reflected by the opposing wall. For example, in the work by Leong et al. [74], transducer reflector distances greater than 85 mm became less effective in achieving skimming of cream from milk when using dual 1- and 2-MHz frequency ultrasound. Leong et al. [73] showed that using high-frequency 2-MHz ultrasound resulted in significant decline of sound pressures with distance beyond 55 mm in a large experimental chamber. By contrast, mid-frequency 400-kHz ultrasound was found to penetrate a considerably longer distance (up to 2 m) without decline in pressure level.

The distance between the sound source and reflector also influences the number of pressure nodes and antinodes that are created within the standing wave. The higher the frequency and the greater the distance to the reflector, the larger the number of available sites (nodes/antinodes) available for food material to accumulate and separate.

The alignment and positioning of transducers and the reflector within the reactor to generate standing waves should be considered with respect to (a) the direction of material flow inside the reactor, either horizontally or vertically, and (b) the alignment of multiple transducers in parallel or perpendicular reactor walls. Note that a perfectly parallel alignment between transducer and reflector/transducer, while offering optimal reflection, is in practice not generally required in large systems to produce effective separation. A vertical or horizontal alignment refers to whether the transducer or reflector is positioned in a way such that the nodal/antinodal bands formed are aligned in the vertical or horizontal plane. Where enhanced separation due to gravity is a necessary mechanism for (i.e. product collects at the top or bottom of the container), a vertical alignment is more amenable to separation as once product is aggregated to a sufficient size, the product can sediment or rise rapidly. A horizontal alignment may hinder this natural rise/fall since product must pass through aligned bands that can ‘trap’ the aggregated material prior to eventual rising/falling beyond the active processing region.

When using multiple transducers to increase the density of the energy input, an increased energy density can be beneficial to accelerating particle accumulation and hence separation. In this case, the transducer plates can be arranged facing each other (parallel) or orthogonal (perpendicular) and either position has been shown to be effective [64]. Parallel alignment can be envisioned to be more likely to achieve effective superposition when using identical frequency (or wavelength multiple) transducers. Perpendicular alignment may create more regions where particle accumulation can occur. Parallel alignment has the advantage of allowing close positioning of the transducers when facing each other. Very short transducer-reflector distances, or in this case transducer-transducer distances, can be realised even with large surface area plates when using a parallel alignment. The use of parallel transducers has been shown to work at batch scale, although additional adjustments may be needed to avoid direct sound radiation into the transducer, which may cause damage to the piezoceramics due to excessive heating. If necessary, transmission plates designed with optimal thickness for sound transmission can be used to position the transducers externally [91], which may allow for a more durable system for continuous processing. A perpendicular alignment on the other hand is limited by the size of the transducer. Large transducers will increase the width of the separation chamber. In some cases, this width may be too large to allow for sufficient sound penetration due to sound wave attenuation (e.g. if high frequencies are used). This reduces the effectiveness and efficiency of the separation.

Conclusion

Ultrasound technology has proven to be a versatile technology for a number of different applications, with the potential to

Table 2 Design features of a megasonic reactor

Design feature	Description
Transducer positioning and arrangement	- In front of reflector - Transducers can be either (a) submerged inside the reactor, (b) directly flanged to an opening in the reactor wall or (c) mounted externally by using a transmission plate with a specific thickness (multiple of a half wavelength) in a section of the wall—a coupling medium between the transducer surface and the transmission plate (e.g. water) is needed - Single, parallel, perpendicular arrangements to be considered • Single frequency or combined frequencies
Reactor cross section	- Non-circular with an even number of side edges - Depends on depth of penetration of the transducer being used - Determined by the allowable transducer size (e.g. cross sections with >4 faces possible for small transducer plates)
Reactor volumes	- 1–100 L depending on number of transducers required and cross section - Dictated by power delivery available through the transducer and plate capacity on the walls

Adapted with permission from Leong et al. [77]

increase process speed with minimum energy requirements or allow for effects that are not accessible by any other means. Applications can have benefits from a quality perspective (e.g. better flavour, texture and nutrient retention of the processed food material) but also from an environmental and economical sustainability perspective (e.g. increased throughput, higher extraction yields, lower energy consumption).

While low-frequency ultrasound has been investigated extensively in food processing applications, there are still new opportunities that are in the early stages of research and development (e.g. airborne ultrasound). In the high-frequency spectrum, the use of ultrasound for enhancing food processes is still in its infancy and many of the theories that would explain the observed effects have not been proven yet. However, the industrial application of megasonics can now be found in the palm oil industry and is finding opportunities in the olive oil industry. Furthermore, with the development of higher performance systems and extension of the frequency range to frequencies in excess of 3 MHz, will give rise to new applications not investigated to date.

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