Mechanism of ultrasound assisted nucleation during freezing and its application in food freezing process

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ABSTRACT
Ultrasound assisted freezing (UAF) is a common and remarkable technology of food preservation to conserve the sensory characteristics and nutritional value. The propagation of ultrasound in a medium generates various physical and chemical effects and these effects have been harnessed to improve the efficiency of food freezing. This review provides an overview of recent developments related to the mechanism, influencing factors, equipment of UAF. The applications of high intensity ultrasound to improve the efficiency of freezing process, to control the size and size distribution of ice crystals and to improve the quality of frozen foods are discussed in considerable detail. Then the future development trends and challenges of UAF have also been highlighted.

Introduction
Freezing is an excellent preservation method to retain valuable sensory attributes and nutritional value of fresh food by inhibiting the activity of enzyme and the growth of microorganism.\textsuperscript{1,2} It involves a phase transition process, in which ice crystal size and distribution are closely related to food quality. Therefore, one of the vital steps in determining the effectiveness of the freezing process and the quality of frozen food is the crystallization of ice.\textsuperscript{3} Rapid freezing produces fine crystals distributing inside and outside the cells and they can better maintain the original quality of the food.\textsuperscript{4} Conversely, slow freezing will form large and unevenly distributed ice crystals in the intercellular space, which causes significant damage to the tissues and thereby causes the quality degradation.\textsuperscript{5} New freezing technologies are developed to improve the crystallization process of frozen food.\textsuperscript{6,7}

UAF as a new food processing technology has developed rapidly in the food industry.\textsuperscript{8–11} As shown in Figure 1, depending on the frequencies used, ultrasound applied in the food industry can be divided into two categories: (i) diagnostic ultrasound (5–10 MHz range); (ii) power ultrasound (20–100 kHz range).\textsuperscript{12,13} The former is an analytical tool for nondestructive inspection, process control and quality assessment including determining food properties such as hardness, oil content and total soluble solids, measuring ice content and examining food packaging materials.\textsuperscript{14} The latter is used in food freezing, drying, extraction, and filtration as an auxiliary measure. What’s more, it can be used in food sterilization, emulsification, deaeration and meat tenderization processes.\textsuperscript{15} From the perspective of utilization, ultrasound can also be separated into two categories: (i) low intensity (<1 W/cm\textsuperscript{2}) and (ii) high intensity (10–1000 W/cm\textsuperscript{2}).\textsuperscript{16} Additionally, ultrasound waves with acoustic intensity below 1 W/m\textsuperscript{2} and frequency above 1 MHz is nondestructive, which is generally utilized...
in the field of analysis and can provide the information of the physicochemical characteristics of food materials. UAF could initiate instantaneous nucleation processes and promote the subsequent crystallization.

**Mechanisms and devices of UAF**

The freezing process is categorized into three stages: (i) precooling stage (or chilling stage): cooling the product from the initial temperature to the freezing point; (ii) phase transition stage: eliminating latent heat of crystallization; (iii) subcooling stage: cooling the product to the final temperature. The phase transition stage is the most significant of these three stages since it involves the crystallization, which determines the efficiency of the freezing process and the quality of the frozen products. Crystallization consists of two steps: the formation of nuclei (nucleation) and the subsequent growth of nuclei. Some studies show that crystallization is a spontaneous and random process, which is hard to control and forecast. During the freezing process, ultrasound is applied only in the transition phase to induce nucleation and promote subsequent ice crystal growth. This increases the efficiency of freezing and affects the spontaneous and random nature of the crystallization process.

**Figure 1.** The frequency range of sound waves.

**Figure 2.** The schematic of cavitation induced by ultrasound: (a) Stable cavitation, (b) transient cavitation.
**Acoustic mechanisms of UAF**

UAF enhances the freezing process by initiating nucleation and accelerating the rate of heat and mass transfer.\(^{15}\) UAF-induced nucleation mainly includes two aspects: primary nucleation and secondary nucleation. In the process of primary nucleation, quite a few bubbles are produced thanks to the compression and extension effect of ultrasound, which can do duty for nuclei for small ice crystals formation.\(^{25,26}\) In addition, the steady-state movement of bubbles accelerates the heat and mass transfer process. In the process of secondary nucleation, the collapse of transient cavitation bubbles can release local high temperature and high pressure, micro-jet and shock wave instantly, which can destroy the dendritic structures of ice crystals, limit the size of original ice crystals.\(^{27,28}\) In the meantime, the newly formed small ice crystals can serve again as crystal nuclei to promote the formation of ice crystals, thereby shortening the freezing time.\(^{29}\)

**Primary nucleation**

The primary nucleation includes homogeneous and heterogeneous nucleation. The former occurs in pure systems with a large subcooling; the latter occurs in a system that is not absolutely pure, leading to the growth of ice crystals in uneven locations.\(^{30}\) The acoustic mechanism of UAF has been studied for many years; however, the researchers have not generated uniform explanation. The commonly accepted mechanisms are the cavitation effect and microstreaming.\(^{31}\) As shown in Figure 2, ultrasound is propagated via a series of compression and rarefaction cycles induced by sound waves to the molecules of medium.\(^{12}\) When the ultrasonic amplitude increases and exceeds a certain level, the dimensions of the negative pressure in the areas of rarefaction will ultimately become competent to lead to the liquid to fracture, resulting in the formation of cavitation bubbles.\(^{33,34}\) Depending on whether the cavitation bubbles burst, it can be categorized into stable and transient cavitation.\(^{32}\) Stable cavitation typically occurs at low acoustic pressure. The movement of bubbles in the process of stable cavitation will produce a variety of acoustic effects in the medium. Microstreaming is one of them, which occurs when the oscillating bubble generates a violent circular motion, forming a strong eddy in the fluid around the bubble.\(^{35,36}\) The diffusion of dissolved gas inside and outside the bubble will also produce microcurrent around the bubble. Microstreaming technology could be applied in the degassing and disruption of cell membrane.\(^{37}\) In contrast, transient cavitation typically occurs at high acoustic pressure, where the cavitation bubbles expand more quickly. Before the cavitation bubbles collapse, a large amount of energy is stored in the bubbles. Thus, when the bubbles burst, high pressure (up to 100 MPa) and high temperature (up to 5000 K) are instantaneously generated.\(^{38}\)

A huge number of cavitation bubbles are formed, grown, oscillated, and burst during the propagation of ultrasound and high local pressure (5 GPa) generates in a very short time, which results in an increase in supercooling degree providing a large driving force for nucleation.\(^{39}\) The stable cavitation generates a huge number of stable cavitation bubbles without the implosion phenomenon. The explosion and propagation of cavitation bubbles under the action of ultrasonic field can generate microstreaming. The vigorous agitation resulting from ultrasonic microstreaming is the dominant factor in enhancing heat and mass transfer during food freezing process, so as to increase the heat and mass transfer, promote the nucleation rate and improve the freezing speed.\(^{40,41}\) Besides, the cavitation bubbles can reach the critical nucleus size acting as nuclei for ice nucleation.\(^{15}\)

Some researchers have indicated that UAF could effectively reduce the degree of subcooling required for nucleation of ice crystals, thereby accelerating the formation of ice crystals. Chow et al.\(^{25}\) found that the supercooling required for the initial nucleation of sucrose solution decreased with the increase of ultrasonic output power and load cycle. The number of bubbles in sucrose solution also increased with the increasing output power. Therefore, the author believed that the formation of crystal nuclei in sucrose solution was related to the intensity of ultrasonic cavitation effect. Patrick et al.\(^{42}\) came to a similar conclusion that ultrasound intensity needed to be adjusted to reduce the supercooling for ice crystal nucleation. Only the cavitation effect can affect the nucleation temperature of the sample. The liquid flow caused by cavitation bubbles also affected the nucleation of ice crystals.\(^{43}\) Chow et al.\(^{44}\)
reported that the primary nucleation of ice with different sucrose concentrations occurred at continuously increasing temperatures (lower subcooling), rather than in repeated experiments without ultrasound. Besides, the nucleation temperature can be reproduced with a lower standard deviation and a higher accuracy than the controlled conditions (no ultrasound). Figure 3 distinctly reveals the ice crystals generated in the ultrasound field recorded via video.\textsuperscript{[25]}

**Figure 3.** Photographs of ice crystals nucleated in a 15 w.t. % sucrose solution at $-3.4^\circ\text{C}$ by a commercial ultrasonic device (output 4, 10% duty cycle): (a) ice crystals following an ultrasonic pulse, (b) crystals 5 s later.\textsuperscript{[25]}

**Figure 4.** Ultrasonic secondary nucleation of ice crystals in pure water and the images are displayed at 1s intervals.
Secondary nucleation

Secondary nucleation is induced by the preexisting crystal, which distinctly belongs to the heterogeneous nucleation. There are two types of secondary nucleation, depending on the initial template. One is the secondary nucleation and occurs when existing crystals act as templates for ice nuclei to form new ice crystals. Another possibility is that more nucleation sites are produced due to crystal fragmentation. [44] Chow et al. [45] reported the influence of ultrasound on the secondary nucleation of pure water. Figures 4 and 5 show the influence of ultrasound on dendritic ice crystals in pure water and sucrose solutions. The dendritic ice crystals were observed without ultrasonic treatment in pure water (Figure 4a). The application of ultrasound contributed to a large number of cavitation bubbles appearing (Figure 4b). After an interval of 1 s, the dendrite tip fell off, cracks appeared, and many melting points were resulted from cavitation bubbles (Figure 4c). Eventually, the cracks started to grow; in addition, new cracks were generated and the ice crystals further melted (Figure 4d). Chow et al. [44] reported that the effect of low frequency ultrasound on the secondary nucleation of ice crystals in a 15 wt % sucrose solution. There were a large number of ice dendrites generated in sucrose solution without ultrasonic treatment (Figure 5a). Some ice dendrites were broken into smaller ice crystals after 1.36 s of ultrasonic treatment (Figure 5b). Ice crystals were broken under the effect of cavitation bubbles after 2.38 s of ultrasonic treatment (Figure 5c). The cavitation bubbles were extremely fluid and moved randomly as shown in the video recording, destroying the ice crystals in the path. At the same time, the original dendritic structures scarcely retained. Figure 5d displays the small crystal fragments left when the ultrasound turned off after a total ultrasonic time of 17.38 s. Besides, these crystal nuclei could grow and produce large quantities of new ice crystals.

UAF could induce nucleation in aqueous solutions and solid food and extremely improve the nucleation certainty or repeatability. [46] Inada et al. [47] reported that the application of UAF highly

Figure 5. The microscopic effect of ultrasound on the secondary nucleation of ice in a 15 wt. % sucrose solution. [44]
enhanced the possibility of nucleation in supercooled water. However, the selection of ultrasonic intensity is considerable to reproducible results. Zhang et al.\textsuperscript{[48]} found that the possibility of phase transition was closely related to the number of bubble nuclei caused by ultrasonic vibration. Although various aspects of UAF-induced nucleation have been widely reported, the mechanism of ultrasound-induced nucleation has not been fully understood, and its mechanism of action remains unclear.\textsuperscript{[49]} With the deepening of research, different researchers have proposed different theories to explain the phenomenon of UAF-induced nucleation. Among them, the theory proposed by Hickling\textsuperscript{[50]} is the most concerned and commonly used. The severe collapse of cavitation bubbles caused by UAF produces high local pressure in a short time. This collapse produces a high degree of subcooling, which is conducive to the formation of transient ice cores. The nucleation occurs immediately after the cavitation bubbles collapse. However, the nucleation does not occur immediately even in the presence of cavitation bubbles, and there is a delay between the collapse of cavitation bubbles and the start of nucleation.\textsuperscript{[49]} Zheng et al.\textsuperscript{[51]} considered that ultrasonic propagates in liquid medium generated a pressure field with alternating positive and negative pressures, making the bubbles dissolved in the liquid bigger and bigger. When the bubbles reach the critical size required for nucleation, they could act as heterogeneous crystal nuclei to promote nucleation. However, this theory has not been verified by experiments so far. Zhang et al.\textsuperscript{[51]} proposed that the flow stream generated by the stable cavitation bubble motion could also be the main driving force for nucleation during freezing. Chow et al.\textsuperscript{[25]} confirmed these results by observing a flow pattern around the cavitation bubbles through a microscope observation. Molecular segregation is another theory that explains the power ultrasonic-induced nucleation during the crystallization of different solutions.\textsuperscript{[52,53]} Some researchers believed that the pressure gradient around the ultrasonic cavitation bubble could be used as the driving force for nucleation to induce nucleation. This pressure gradient accelerated particle diffusion and helped nucleation. The particle diffusion was not an instantaneous process, and particle transmission took some time. Therefore, the observed delay between bubble collapse and nucleation could be explained by the time required for the mass diffusion around the bubble.

**Crystal growth**

Crystal growth is also a highly momentous procedure in the crystallization process and considered as a process of adding molecules to the surface of liquid-solid.\textsuperscript{[54]} UAF is appositive effect on the crystallization process, containing the initiation of the nuclei and the following crystal growth. The effect of UAF on crystal growth did not appear to be as significant as the effect on nucleation, and was largely due to the enhanced bulk-phase mass transfer.\textsuperscript{[55]} The collapse or movement of cavitation bubbles could result in mechanical disturbances, which changed the hydrodynamic characteristics and distinctly enhanced the heat and mass transfer coefficients. Increase in the heat transfer coefficient between frozen food and freezing medium was conducive to the rapid removal of sensible heat and latent heat of fusion.\textsuperscript{[56,57]} During the process of crystallization, a large amount of latent heat needs to release. Therefore, heat and mass transfer are the driving force for the growth of ice crystals. Ohsaka et al.\textsuperscript{[58]} found that low-frequency UAF (21 kHz) was applied to the crystallization process of liquid water or solution containing more air, the ultrasonic cavitation effect could strengthen the formation of crystal nuclei, thus reducing the growth rate of ice crystals.

**Experimental devices of UAF**

Full-immersion Type: The full-immersion type device is composed of ultrasonic system, coolant-circulating system, refrigeration cycle system and temperature detecting system (Figure 6a). The full immersion device has been diffusely utilized in several kinds of freezing performance samples, including liquid samples, solid samples and semi-solid samples (Table 1). Generally, liquid and semi-solid samples are placed in test tubes before UAF, while solid samples are placed directly into the coolant (Figure 6a). The temperature of the coolant was extremely low and even reached −30 °C.
during the freezing process. The most normally used coolants are ethylene glycol, calcium chloride, silicone oil and so on.

Half-immersion Type: The half-immersed UAF devices are displayed in Figure 6b and the ultrasonic transducer horn are firmly combined with the aluminum plate and connected to the ultrasonic generator as well in order to acquire a sufficient resonance frequency on the plate. During the freezing process, the choice of the position of the vial on the plate is very important as there are nodes on the surface of the board caused by the ultrasonic resonance phenomenon. A thin layer of powder is placed on the plate and then ultrasound is exerted. If the powder does not move on the surface of the plate, then these are the nodes where the ultrasonic waves cannot propagate inside these vials at all. Some researchers have shown that semi-immersion devices are commonly used to freeze liquid samples, such as bovine serum albumin\textsuperscript{[24]} or mannitol aqueous solution.\textsuperscript{[66]}

Non-immersion Type: The non-immersion type of UAF device is also known as direct contact UAF by Islam et al.\textsuperscript{[67]} This device has many similarities with full-immersion device (Figure 6c). Multiple piezoelectric ultrasonic transducers usually linked to the bottom of the stainless-steel freezing tank and connected to the generator. This device is used in freezing solid samples and the samples can be directly frozen without the need for a cooling liquid. Therefore, the frozen products are not contaminated by the freezing medium. However, the disadvantage of the device is that the transmission efficiency of UAF in the air is considerably low, leading to a serious loss of ultrasound energy during freezing.

**Application of UAF in food freezing**

Applications of UAF in food freezing process have been reported by many studies and they were summarized in Table 2.
Table 1. Comparison of ultrasound-assisted freezing devices.

| Type                | Main components                                                                 | Features                                                                                           | Frozen substrate | Freezing medium                         | Process parameters                                                                 | References |
|---------------------|----------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------|------------------|------------------------------------------|------------------------------------------------------------------------------------|------------|
| Full-immersion type | Ultrasonic system, coolant-circulating system, refrigeration cycle system, and temperature-detecting system. | The propagation efficiency of ultrasound is high. However, the disadvantage is that the frozen product is completely immersed in the cooling liquid to freeze. Thus, the frozen products may be easily contaminated by the coolant. | Potato           | Solution of glycerol and water (50/50, w/w) | 35 kHz; Ultrasound processor: 300 W; Vibration amplitude: 0%-100%; Treatment time: 8 s; Pulses duration: 1 s; Immersion temperature: −6 °C | [59]       |
| Lotus root          | Solution of calcium chloride and water (29/71, w/w)                              | 30 kHz; UIF-1 (90 W, 30 s on/30 s off), UIF-2 (150 W, 30 s on/30 s off), UIF-3 (210 W, 30 s on/30 s off), UIF-4 (150 W, 15 s on/45 s off) and UIF-5 (150 W, 45 s on/15 s off); Immersion temperature: −25 °C. | Lotus root       | 95% ethanol                              | 30 kHz; Ultrasonic power levels: 0, 125, 150, 175, 200, and 225 W; Duty cycle: 30 s on/30 s off. | [60]       |
| Common carp         | 30% (w/v) CaCl<sub>2</sub> solution                                              | 30 kHz; Ultrasound intensity: 0.09, 0.17, 0.28, 0.42, and 0.51 W/cm<sup>2</sup>; Exposure time: 30 s; | Strawberry       | 30% (w/v) CaCl<sub>2</sub> water solution | 30 kHz; Power intensities: 0.09, 0.17, 0.26, or 0.37 W/cm<sup>2</sup>; Duty cycle: 30 s on/30 s off. | [61]       |
| Red radish          | 30% (w/v) CaCl<sub>2</sub> water solution                                        | 20 kHz; Power intensities: 0.09, 0.17, 0.26, or 0.37 W/cm<sup>2</sup>; Duty cycle: 30 s on/30 s off. | Red radish       | ethylene glycol-water mixture            | Irradiation temperature: −2, −3, −4 and −5°C; Irradiation duration: 0, 1, 3 s, 5, 10 or 15 s; Ultrasound intensity: 0.07, 0.14, 0.25, 0.35 and 0.42 W/cm<sup>2</sup>. | [62]       |
| Agar gel samples    | Ethylene glycol-water mixture                                                   | 20 kHz; Ultrasound intensities: 0.26, 0.38, 0.54 and 0.69 W/cm<sup>2</sup>; Duty cycle: 10 s on/10 s off. | Gelatin gel samples | 30% (w/v) CaCl<sub>2</sub> solution | 25 kHz; Ultrasound intensity: 0.25 W/cm<sup>2</sup>; Irradiation duration: 0, 1, 3, 5, 10 and 15 s; Immersion temperature: −20 °C; Immersion temperature: −20 °C; | [63]       |
| Water               | A mixture of ethylene glycol and water (50%:50% in volume)                      | 30 kHz; Ultrasonic power levels: 0, 125, 165, 205, and 245 W; Duty cycle: 30 s on/30 s off; Duration: 8 min. | Water            | Solution of 95% (v/v) ethanol and 5% fluoride |                                                                                     |            |

(Continued)
| Type                  | Main components                                                                 | Features                                                                                                                                                                                                 | Frozen substrate | Freezing medium       | Process parameters                | References |
|----------------------|----------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------|-----------------------|-----------------------------------|------------|
| Half-immersion type  | Ultrasonic system, coolant-circulating system, refrigeration system, and       | The metal plate is tightly bound to ultrasonic transducers, while samples inside the container (basically liquid and semisolid samples) are placed on the plate with the medium to improve the propagation of ultrasound between the plate and the container. The nodal points exist on the plate surface caused by the resonance phenomenon of ultrasound. The finding of nodal points needs to be carried out in advance before starting the freezing process. These nodal points should be avoided for the location of vials during freezing. | Mannitol         | Liquid nitrogen       | 35 kHz; Ultrasound intensity: 0.2 W/cm². | [27,66]   |
| Non-immersion type   | Ultrasonic system, refrigeration system, and temperature-detecting system.     | The samples are frozen directly without any coolant immersion. Thus, the frozen products are not contaminated by the coolant. However, the propagation efficiency of ultrasound in the air is relatively low, resulting in serious loss of ultrasonic energy during the freezing process. | Mushroom –¹      | –¹                   | 20 kHz; Ultrasound intensity: 0.11 W/cm²; Duty cycle: 10 s on/20 s off. | [67]      |

¹ None.
| Applications                  | Food substrate                          | Operating conditions                                                                 | Significant results                                                                                                                                                                                                 | References |
|------------------------------|-----------------------------------------|--------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|
| Inducing ice nucleation      | L-glutamic acid                         | 20 kHz; Tip diameter: 6 mm; Continuous irradiation: 9.0–22.5 W, 100% duty cycle; Pulsed irradiation: 13.5 W, duty cycle of 16.7%, 50%, and 83.3%. | The induction time was shortened with application of ultrasound, and the induction time was further shortened with higher ultrasonic power.                                                                                              | [68]       |
| Agar gel sample              | 25 kHz                                  | Irradiation duration: 0, 1, 3, 5, 10 and 15 s; Ultrasound intensity: 0.07, 0.14, 0.25, 0.35 and 0.42 W/cm²; Onset temperature: −2, −3, −4 and −SoC | Ultrasound irradiation can initiate nucleation in agar gel at different supercooling temperatures as long as the suitable ultrasonic intensity and time are selected.                                                          | [49]       |
| Radish cylinder              | 20 kHz                                  | Duration: 0, 3, 7, 10 and 15 s; Ultrasound intensity: 0.09, 0.17, 0.26 and 0.37 W/cm²; Onset temperature: −0.5, −1, −1.5 and −2 °C | Ultrasound irradiation temperature at −0.5°C for 7 s duration with intensity of 0.26 W/cm², was an optimal ultrasound application conditions for the nucleation inducement of radish cylinder samples. | [69]       |
| Supercooled water            | 28 kHz, 0–100 W                         | Power: 40 W; Duration: 1 s                                                          | Ultrasound vibration forcefully facilitates the phase transition from supercooled water to ice.                                                                                                                                                             | [47]       |
| Supercooled water            | 39 kHz, 4.4 kW/m²                        |                                                                                     | The probability of phase transition was inseparable bound up with the number of nuclei induced by ultrasonic vibration.                                                                                                                                  | [48]       |
| Controlling ice crystals' size and shape | Mannitol solution                      | 35 kHz; Acoustic power: 0.25, 70, 115, 140 a.u.; Supercooling degree: 2.9, 4, 6, 8, 9.1 K; Duration: 1 s | Increasing supercooling and acoustic power resulted in decreasing ice crystals’ mean size and increasing their mean circularity.                                                                                                                      | [70]       |
| Mannitol solution; BSA; Sucrose solution | 35.89 kHz                              | Electric power: 40 W; Duration: 1 s                                                  |                                                                                                                                                                                                                                                              | [24]       |
| Agar gel sample              | 25 kHz                                  | Irradiation duration: 90, 180 and 270 s; Ultrasound intensity: 0.07, 0.25 and 0.42 W/cm²; Duty cycle: 30%, 50%, and 70% | Higher nucleation temperatures caused by UAF contributed to initiation of comparatively larger and directional ice crystals.                                                                                                                                | [71]       |
|                              |                                         |                                                                                     | The size distribution of ice crystals was affected by ultrasound induced nucleation temperature. However, no regular relationship between the evaluated parameters (intensity, duration and duty cycle) and the crystal sizes was detected.                                        |            |

(Continued)
| Applications                           | Food substrate   | Operating conditions                                                                                       | Significant results                                                                                                                                                                                                 | References |
|---------------------------------------|------------------|----------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|
| Accelerating freezing rate            | Frozen dough     | 5 kHz; Electric power levels: 0, 175, 224, 288, 360 and 418 W; Duty cycle: 30 s on/30 s off                | At 288 or 360 W power levels, the total time for dough freezing shortened more than 11% significantly (P < .05), and the required time for each stage was reduced.                                                         | [73]       |
|                                       | Grass carp       | 40 kHz; Ultrasound intensity: 0, 0.30, 0.38, 0.48 and 0.60 W/cm²; Duty cycle: 8 s on/2 s off             | The application of power ultrasound at 0.38 W/cm² and above during immersion freezing can significantly shorten the length of precooling stage, phase transition and, total freezing time (P < .05), protect muscle fiber structure, resulting in decreased thawing loss. | [73]       |
|                                       | Broccoli         | 20 or 30 kHz; Ultrasound power: 0, 125, 150, 175 and 190 W; Duty cycle: 60 s on/60 s off                  | The total freezing time and times required for pre-cooling, phase change and sub-cooling stages of broccoli were significantly reduced by the application of UAF (UAF) at 150 (30 kHz) or 175 W (20 kHz) power level. | [74]       |
|                                       | Potato            | 25 kHz; Actual ultrasound power: 0, 7.34, 15.85 and 28.89 W; Exposure time: 0, 1, 1.5, 2 and 2.5 min    | The freezing rate was improved greatly when 15.85 W ultrasound power was applied for 2 min (P < .05).                                                                                                                     | [75]       |
| Improving the Microstructure of frozen food | Porcine longissimus muscle | Ultrasound power: 180 W; the geometric center temperature: −18°C; storage time: 0, 30, 60, 90, 120, 150, and 180 d | Ultrasound at particular powers is an efficient method in reducing quality deterioration of muscles during long-term frozen storage.                                                                                                    | [75]       |
|                                       | Potato            | Ultrasound power: 270 W; Dual-frequency: 20 kHz, 28 kHz; Duty cycle: 50%; Total exposure time: 120 s     | Orthogonal ultrasound-assisted freezing gives small ice crystals and effectively preserves tissue structure. Microstructure analysis proves effective upgrade in frozen product quality by orthogonal ultrasound-assisted freezing. | [76]       |
|                                       | Thunnus tonggol  | 40 kHz; Ultrasound power: 0, 160, 280,400 W; Ultrasound intensity: 11.63 W/cm²; Onset temperature: 15 ± 1°C | Tuna thawed at 280 W suffered fewer negative effects on its microstructure.                                                                                                                                              | [76]       |
|                                       | Mushrooms        | 20 kHz; Acoustic intensity: 0, 0.13, 0.27 and 0.39 W/cm²; Duty cycle: 10 s on/10 s off                   | Ultrasound at 0.39 W cm² (20 kHz) resulted in the highest textural hardness values in all the three mushroom varieties.                                                                                                   | [77]       |
|                                       | Potato            | 25 kHz; Actual ultrasound power: 0, 7.34, 15.85 and 28.89 W; Exposure time: 2 min                       | Under the ultrasonic power of 15.85 W, the tissue structure of potato was significantly improved.                                                                                                                       | [78]       |
|                                       | Broccoli         | 20 or 30 kHz; Ultrasound power: 0, 125, 150, 175 and 190 W; Duty cycle: 60 s on/60 s off                 | The application of UAF at selected acoustic intensity with a range of 0.250–0.412 W/cm² decreased the freezing time and the loss of cell-wall bound calcium content. Compared to normal freezing, the values of textural properties, color, l-ascorbic acid content were better preserved and the drip loss was significantly minimized by the application of UAF. | [79]       |
**Inducing ice nucleation**

During the process of UAF, characteristics of frozen food such as the organizational status, gas content, water content and distribution, porosity, bubble size and viscosity affect the UAF in varying degrees. The state of food organization is extremely critical. There are some differences in the mode of ultrasonic action and the internal mechanism of inducing nucleation in liquid, semi-solid and solid foods. When the UAF is used for frozen crystallization of liquid food, the ultrasonic intensity should be greater than 2 W/L, the frequency range is 20–40 kHz, and the ultrasonic action time should be as short as possible. Kiani et al. found that UAF induced nucleation in both liquid and simulated solid foods, and the nucleation behavior was highly repetitive under different ultrasonic radiation temperatures. There was a linear relationship between ultrasonic radiation temperature and nucleation temperature. Studies show that ultrasound-induced nucleation of liquid foods is more reproducible than that of solid foods, and its mechanism of action remains to be further investigated. The content of gas in food also affects the effect of UAF. When ultrasound is applied to liquid food, bubbles in the solution can be released to form cavitation bubbles under negative pressure due to the periodic change of pressure. The cavitation bubbles induce nucleation as crystal nucleus and the vibration of the bubbles enhance the mass transfer and heat transfer, improving the freezing rate. Yu et al. showed that UAF improved the nucleation temperature of both pure water and degassed water. The degree of supercooling required for degassed water nucleation is larger than that of pure water due to the cavitation effect and other acoustic effects. Hu et al. found that the nucleation took place within 1 s after UAF for liquid food with bubble injection in advance, which indicated that the bubbles injected in advance of liquid food formed cavitation bubbles under the action of ultrasound inducing nucleation.

**Controlling ice crystals’ size and shape**

High-quality frozen foods should be similar to the original unfrozen foods in appearance, aroma, texture, color, and nutritional value. Water in tissues can be divided into extracellular water (intercellular) and intracellular water (intracellular). During freezing, water outside the cell is more easily converted into ice crystals than water inside the cell. With the gradual formation of ice crystals, the remaining concentration of unfrozen extracellular water will become higher and higher, resulting in a decrease in vapor pressure. Furthermore, the water inside the cell will remain in a supercooled state, and its steam exceeds the water outside the cell, causing water to migrate from inside the cell to outside. The ice crystals outside the cell become larger and larger, causing the cell wall to be squeezed, resulting in cell deformation and even damage. The intra – and extra-cellular water need to be transformed into ice crystals as much as possible in order to obtain high-quality frozen foods, and the formation of ice crystals needs to be as small as possible. At present, it has been widely confirmed that high freezing speed can achieve this purpose, thereby obtaining high-quality frozen products.

In the process of freezing, the quality of the final frozen products depends largely on the size and shape of ice crystals. The formation of super-large ice crystals and the uneven distribution in food tissues could irreversibly damage the cell structure, leading to the poor sensory characteristics and loss of nutrients. In some cases, ice crystals with large size and vertical direction are expected to be obtained during freeze-drying and freeze concentration. The larger ice crystals help to improve the drying rate in the subsequent sublimation stage and promote the separation of ice crystals from them, which could achieve the purpose of reducing energy consumption and cost. UAF could control the formation of ice nucleation in frozen foods. A large number of ice cores formed because the nucleation rate is much higher than the crystal growth rate.

Saclier et al. successfully controlled the ice crystal size in freeze-drying process with UAF. Only a few large crystals formed and they were initially disc-shaped and then grew into star like dendrites when the sucrose solution was irradiated with ultrasound for 5 s under supercooling of 1 °C. However,
there were multiple nucleation points of rapid solidification, and the crystal was smaller when ultrasound acted on the supercooling of 5 °C. Some large holes observed in the former sample and much smaller holes observed in the latter after drying. Ultrasound was used for a short time to produce large ice crystals as the continuous application of ultrasound led to the breaking of ice crystals. Kiani et al.\[71\] showed that the size and distribution of ice crystals were closely connected with the initial temperature of ultrasonic action and other ultrasonic parameters, such as action time, mode and ultrasonic intensity (Figure 7). Saclier et al.\[70\] reported that the increase of subcooling and ultrasonic power were helpful to reduce the average size and increased the perimeter of ice crystal. Xin et al.\[79\] indicated that UAF significantly shortened the freezing time, reduce the loss of water drop and maintain the calcium binding of cell wall in the power range of 0.250–0.412 W/cm² under the selected sound intensity. Tu et al.\[60\] also found that the freezing time of UAF was shortened by about 17% at the interval of 150 W and 30 s, and the hardness of frozen lotus samples was improved, the drop loss was reduced, and the quality of frozen food was improved.
**Improving freezing rate**

Fast freezing can better maintain the quality of frozen food than slow freezing and UAF accelerates the freezing process of food materials. The intense agitation of ultrasonic microbeaming increases the effect of heat and mass transfer and the freezing rate is closely related to the heat and mass transfer in the freezing process.\(^{[56]}\) UAF can accelerate the freezing speed. Kiani et al.\(^{[97]}\) revealed that higher ultrasonic intensity led to higher cooling rate, which increased the Nusselt number (Nu) from about 23–27 to 25–108. However, UAF also has a thermal effect and generates heat on the surface of the sphere offsetting some of the positive effects of UAF and even bringing negative effects to the freezing process. Acoustic flow and cavitation are the two main factors affecting heat transfer. As shown in Figure 8, the clouds of cavitation bubbles on the surface of the sphere are the main factor generating the heating effect.\(^{[97]}\) Kiani et al.\(^{[98]}\) evaluated that the rate of heat transfer between a submerged object and cooling medium was accelerated by the application of ultrasound irradiation. In addition, the impact of ultrasound irradiation on heat transfer was unconsidered with the ball diameter. What’s more, relationship between the ultrasound intensity and the Nu values was a linear. Extensive researches have been reported that the UAF could accelerate the freezing speed. Sun et al.\(^{[41]}\) found that ultrasound-assisted immersion freezing (UIF) accelerated the freezing rate of common carp at 175 W. Comandini et al.\(^{[59]}\) confirmed that the freezing time was significantly shorter when ultrasound was applied at −2.0 °C.

UAF time is prolonged and the ultrasonic thermal effect is enhanced as the intensity or pulse value of the ultrasound increased. The heat can hinder the formation of ice crystals and slow down the freezing rate. The appropriate ultrasonic parameters should be studied to improve the effect of power ultrasonic enhanced freezing. Li et al.\(^{[56]}\) found that UAF with power of 15.89 W for 2 min significantly improved the potato freezing rate. Xu et al.\(^{[69]}\) showed that UAF with 0.26 W/cm\(^2\) had the best effect on ice nucleation at −0.5 °C for 7 s. Kiani et al.\(^{[49]}\) pointed out the optimum processing parameters on water nucleation in agar gel samples were 0.25 W/cm\(^2\) at −2 °C for 3 s. Wang et al.\(^{[92]}\) reported that the ultrasonic power, frequency and processing time could improve the dehydration performance of vacuum frozen strawberry slices. After orthogonal experiment optimization, power is the most important factor, followed by frequency and time.

**Improving the microstructure of frozen food**

The size and distribution of ice crystals in frozen food are strongly associated with the freezing speed and UAF can prominently accelerate the freezing speed.\(^{[99–101]}\) The traditional freezing methods, such as direct-contact freezing, immersion freezing and air blast freezing, produce large ice crystals. UAF can effectively promote the formation of small and evenly distributed ice crystals and improve the microstructure. Zhu et al.\(^{[102]}\) evaluated the effect of triple-ultrasound assisted freezing on potato quality. Figure 9 show scanning electron microscopy (SEM) images of potato samples with different freezing conditions. The structure of the TUF (triple-ultrasound assisted freezing) treated sample is the densest with the fine pores, which indicates that the ice crystals formed by multi-frequency ultrasound are smaller in size and more uniformly distributed. The SUF (single-ultrasound assisted freezing) (Figure 9b) and DUF (dual-ultrasound assisted freezing) (Figure 9c) treated samples have larger ice crystal pores compared with TUF, whereas the non-treated samples (Figure 9a) have the largest ice crystal pores. These results showed that the size of ice crystals in potato samples could be reduced by UAF at different frequencies, which leads to the change of microstructure. With the increase of ultrasonic frequency, the effect becomes more significant. TUF sample had better quality than that of SUF and DUF as the multi frequency ultrasound could produce more cavitation nuclei and enhance the cavitation effect. Sun et al.\(^{[41]}\) reported appropriate ultrasonic power (175 W) promoted the formation of smaller and more uniform ice crystals in the muscle tissue, and maintain the integrity of sarcomere in common carp. Zhang et al.\(^{[75]}\) showed that the crystal size of UIF (ultrasound-assisted immersion freezing) treated longissimus dorsi muscle samples was smaller and
the distribution of ice crystal was uniform. In addition, ultrasonic treatment can help to maintain the microstructure of thawed food. According to Qiu et al.,[16] application of ultrasound can better keep the microstructure, decrease drip loss, reduce color and texture changes and maintain certain natural nutrients of food items during freezing process. In the meantime, quality enhancement is likewise observed in food thawed by ultrasound-assisted thawing technique. The effects of single/dual-frequency orthogonal ultrasound-assisted freezing as compared to single direction ultrasound-assisted freezing on the quality of potatoes were studied by Tian et al.[26] Results indicated that orthogonal ultrasound could significantly reduce drip loss and increase hardness by enhancing the freezing rate and reducing the total freezing time. It is showed that dual-frequency orthogonal ultrasound-assisted freezing endowed potatoes with the finest pores and the plumpest tissue in terms of microstructure observation. This investigation reveals the enormous potential of the dual-frequency orthogonal ultrasound-assisted freezing technique in promoting the preservation of sensory characteristics and nutritional value of foods. Li et al.[76] showed ultrasonic thawing had a positive effect on the microstructure of tuna myofibrillar protein and Peruvian squid.
Future trends and developments for UAF

Exact mechanism of UAF

As early as the 1960s, some researchers summarized the mechanism of UAF, which mainly involved the cavitation effect, mechanical effect, thermal effect, heterogeneous nucleation and other theories. UAF causes a series of complex and intense physical changes in the process of ultrasonic propagation in the medium. However, the mechanism of power ultrasonic enhanced freezing has not formed a unified understanding, and the specific mechanism is still uncertain. The related theories need further research and discuss.

Optimization of UAF process parameters

The heat produced by the thermal effect of ultrasound are absorbed by the liquid medium, which counteracts some of the positive effects of ultrasound and even has a negative impact on the freezing process. The increase of ultrasonic power, pulse value or treatment time can enhance the effect of micro-jet, increase the coefficient of heat and mass transfer. Besides, the continuous ultrasonic action can also produce a lot of heat, which hinders the formation of ice crystal. It is necessary to further optimize the process parameters according to the product properties in the process of UAF.

Inhomogeneous distribution of ultrasonic power intensity

For the UAF equipment, the transducer is usually located on the bottom of the freezing tank. UAF has a certain degree of energy loss as it propagates through the medium and the ultrasonic power intensity gradually decreases with the distance from the bottom increasing. This leads to non-uniformity of the ultrasonic power intensity distribution in the freezing tank. The intensity of the ultrasonic power received by different parts of the food sample is different in the UAF process. It is essential to establish an UAF device with uniform ultrasonic power distribution.

Coupling of ultrasonic and other technologies to assist the freezing process

With the in-depth study of UAF mechanism and the development of food machinery industry, as well as the formulation of the best process parameters and the rational use of thermal mechanism, UAF coupled with other technologies to assist the freezing process could broaden the application of UAF technology.

Conclusion

Although ultrasonic assisted freezing technology still has some shortcomings, it can promote nucleation, strengthen heat and mass transfer, accelerate freezing rate, regulate the size and distribution of ice crystals, and improve the quality of frozen food. It is a green and safe novel processing technology. It is believed that with the unremitting exploration, optimization and improvement of ultrasonic assisted freezing technology, the technology will play a more important role in food freezing processing to promote the development of the frozen food industry.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.
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