FREEZING UNDER ELECTRICAL AND MAGNETIC DISTURBANCES; A REVIEW

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ABSTRACT

Phase change in biological tissues may be affected by electrical and magnetic disturbances. This presentation proposes a review based on existing literature as well as some very recent results on freezing under static electric field and microwave conditions.
Freezing under static electric field of water, aqueous solution and pork meat has been investigated by the authors, showing the ability of this process to refine ice crystals in frozen matrices. SEF permits to lower the supercooling and to trigger the phase change of water to ice. The use of radiofrequencies and microwaves has been used recently by researchers to promote refined ice crystallization in food systems. Even though the energy optimization of the process is still in debate, the benefit in terms of size of ice crystals has been evidenced.

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1. INTRODUCTION

The freezing process of food matrices and the impact on the final food quality is mainly governed by water to ice phase transition. Emerging technologies have been developed to control the crystallization process and the size of the ice crystals. Electrical disturbances have been considered in the past and recent works tend to confirm the interest of this physical disturbance to refine the ice crystal in order to reduce the freeze damage. The mechanisms which arise from the existing literature are analyzed in this paper. This paper presents a summary of the review chapter n° 30 “Freezing Combined with Electrical and Magnetic Disturbances” by E. Xanthakis, A. Le-Bail, M. Havet published in Emerging Technologies for Food Processing, Da Wen Sun 2nd Edition, 2014 (Xanthakis et al., 2014-b)

2. WATER PROPERTIES

The effect of electric and magnetic fields on water is linked to the molecule characteristics of water but also to the interactions between them. Martin Chaplin describes 63 abnormal water properties related to hydrogen bond network (http://www.lsbu.ac.uk/water/). The internuclear distance between the oxygen and each hydrogen atom is 0.96 Å while the van der Waals radii for oxygen and hydrogen are 1.40 Å and 1.20 Å respectively (Fennema, 1996); (Chaplin, 2013); (Marechal, 2007). The atom of oxygen attracts more strongly the hydrogens than they are attracted between them due to its electro negativity. Each water molecule can form up to four hydrogen bonds. It is crucial to mention that the hydrogen bonds could be assessed as weak bonds comparatively with the other kinds of bonds. For this reason, in liquid state water the structures are perpetually changing as hydrogen bonds break and reform. Due to the weak hydrogen bonding and the polarity, if the water molecule is placed in an electric field, it will not only respond through the interaction of its dipole moment with that field but as the molecule has significant dipole polarizability, its dipole moment will be enhanced in the presence of this field too (Vaklavik and Christian 2008); (Finney, 2004).

What can simply describe freezing is that upon cooling water metamorphoses into ice. But this naive description of phase transition of water into ice is hiding numerous phenomena which to this day haven't been answered completely and still remain bleary for the scientists.

Freezing can be presented as 3 parts. At first, there is the chilling stage where the removal of sensible heat is taking place and the temperature of the water falls to its freezing point. The following step is the phase transition stage which involves the nucleation and the crystal growth parts. Both of these two successive steps can be defined as the crystallization process. At the beginning of this process, water is cooled below its freezing point but it still remains in a liquid state. At a critical point (nucleation temperature) the decrease of temperature ceases and rises back to the freezing point temperature. This phenomenon is called supercooling and the term which is used to quantify its level is called degree of supercooling; it is defined as the difference between the freezing point and the nucleation temperature. Once the major amount of water has been transformed into ice the temperature of the system tends to equilibrate with the temperature of the cooling medium during the tempering stage.

The interactions between nucleation and crystal growth are of great importance as they are responsible for the size, distribution and morphology of ice crystals. During freezing the majority of a food matrix will transform into ice. Therefore, the size of the ice crystals is crucial for the final quality of the frozen food as they can cause mechanical stress and extended irreversible damage to the cellular structure which in turn degrades the texture, colour, taste, and nutritional value.

Taking advantage of the fact that water molecule can be influenced by the presence of magnetic and/or electrical disturbances, in recent years several efforts have been made and many innovative techniques and patents have been introduced for the control of ice nucleation and subsequent crystal growth during freezing. In particular, new technologies applying electro-magnetic disturbances during freezing have been proposed (Orlowska et al., 2009); (Petersen et al., 2006a); (Petersen et al., 2006b); (Hozumi et al., 2005).

3. PHASE CHANGE UNDER ELECTRICAL DISTURBANCES

The intrinsic electric dipole moment and dipole polarizability of water molecules make water a dielectric material which in turn can be influenced by applied electric fields. Louis Dufour (Dufour, 1861) is maybe the first scientist who tried to apply electric field on supercooled water in an attempt to explain the freezing of water droplets in the atmosphere. Various molecular dynamics studies indicated that the dipoles of water are polarized in the direction of an applied external static electric field (SEF). Depending on the temperature and the strength of the applied SEF, SEF is able to break or weaken the pre-existing hydrogen bonding and as a result to realign the water molecules (Chaplin, 2013); (Woo and Mujumdar 2010); (Wei et al., 2008); (Yan and Patey 2012); (Aragones et al., 2011).

Two kind of studies on the impact of electrical disturbances on water-ice properties can be found: i) impact of an external applied electric field and ii) influence of electric field when it is applied internally to the water by means of immersed charged surfaces (Orlowska et al., 2009); (Svishchev and Kusalik 1994); (Vegiri and Schevkunov 2001); (Zangi and Mark 2004); (Hozumi et al., 2003); (Hozumi et al., 2005) (Petersen et al., 2006b).

Recently, Orlowska *et al*., (2009), based on Guggenheim (1967), Marand *et al*., (1988) and Mullin (2001) reported information, published a thermodynamic approach concerning the influence of an applied SEF on Gibbs free energy of a liquid's phase change. The hypothesis was regarding the Gibbs free energy of a spherical nucleus formation. When a formulation is not under the presence of electric field, the Gibbs free energy ΔG_0 could be described by the following general equation:

$$\Delta G_0 = \Delta G_s + \Delta G_v \quad (1)$$

The terms ΔG_s and ΔG_v in the Eq. (1) represent the free energy of surface and volume respectively. Taking into account the spherical characteristics of the employed formulation the previous equation was transformed into Eq. (2).

$$\Delta G_0 = 4\pi r^2 \gamma - \frac{4}{3}\pi r^3 \Delta G_v \quad (2)$$

where: r is the radius of the sphere, γ is the surface free energy of the crystal fluid interface and ΔG_v is the free energy change of the transformation per unit volume. When this formation is subjected to a static electric field E, a linear system shows a permanent polarization P. Under these conditions the free energy of the system G_E can be generally described by the following equation:

$$G_E = U - TS + pV - \left[V_C E \cdot \left(\varepsilon_0 \varepsilon_r E + P\right)\right] \quad (3)$$

where: V_C is the volume of the spherical system that is subjected to the field, ε_r is the relative permittivity of the system and ε_0 is the vacuum permittivity. The final proposed equation by Orlowska *et al*., (2009) in order to express the Gibbs free energy of a spherical nucleus under the presence of an electric field was Eq. (4).

$$\Delta G_E = 4\pi r^2\gamma - \frac{4}{3}\pi r^3\left(\Delta G_v + PE\right) \tag{4}$$

where: r is the radius of the sphere, γ is the surface free energy of the crystal fluid interface and ΔG_v is the free energy change of the transformation per unit volume. The conclusion of this approach was that there is a clear difference in terms of Gibbs free energy if the Eqs. (2) and (4) are compared. The general thermodynamic rule which correlates ice nucleation with Gibbs free energy is that ice nucleation occurs when the Gibbs free energy of a system is minimized. In Eq. (4) it could be marked that the term related to the polarization of the system as well as to the strength of the applied field causes an additional decrease to the free energy of the system further than the decrease caused by the temperature and entropy diminution. Therefore, this thermodynamic hypothesis could lead to the conjecture that the application of an electric field to water during its supercooling may enhance the nucleation process and bring a premature ice crystal formation in the system. In a previous study Yan *et al*., (1997) had predicted that an applied electric field could induce the crystallization process by using basic thermodynamic equations. In their approach they proposed a theoretical model for correlating the nucleation temperature when electric field is applied (T^e) (Eq. 5).

$$T^e = T^0 + \frac{\varepsilon_0}{2(S_2 - S_1)}\left(\varepsilon_2 V_2 - \varepsilon_1 V_1 + \Delta V_{1-2}\right)E^2 \tag{5}$$

T^0 is the temperature of crystallization in the absence of electric field, ΔV_{1-2} is the volume of crystallization contraction, ε_0 is the permittivity of free space, ε is the relative permittivity, E the intensity of the applied electric field, S_1 and S_2 are the entropies for the liquid and crystal phase respectively. The basic thermodynamic relation regarding the change of entropy in terms of temperature is described by Eq. (6), where ΔQ is the change in the heat of crystallization.

$$\Delta S = -\left(\frac{\Delta Q}{T}\right) \tag{6}$$

They also proposed that in case of an applied electric field the above relation can be transformed into Eq. (7)

$$T^e = \left(\frac{\Delta Q^e}{\Delta Q^0}\right)T^0 \tag{7}$$

where: the superscripts e and 0 represent the presence and the absence of electric field respectively. In this thermodynamic approach by combining the previous equations Yan indicated that the crystallization temperature will increase under the presence of electric field and the crystals will appear earlier than in the case of zero electric field. These thermodynamic aspects could be correlated, offering theoretical evidence, with the results of various experimental and simulation studies which have been carried out over the previous years concerning the influence of an applied electric field on water's crystallization process which will be subsequently presented.

4. MAGNETIC FIELD and PHASE CHANGE

Water can be assumed as a diamagnetic material which means that the magnetic dipole moment is developed under the influence of an external magnetic field. Weak magnetic fields seem to have a minor effect on water, while strong magnetic fields (>10 T) can exert enough force to levitate water against gravity (Ikezoe et al., 1998); (Tagami et al., 1999); (Wowk, 2012). Zhou (Zhou et al., 2000) showed that a significant change in the internal energy and heat capacity occurred at a magnetic flux density of about 0.2 T. The magnetized effect of water increases in tandem with the increasing strengths of the applied magnetic field or the application time. However, once the externally applied magnetic field is eliminated it has been proved that the magnetized effect does not disappear immediately (Pang et al., 2008). This phenomenon is called memory effect of magnetized water.

There is a lack of research regarding the effects of magnetic fields on the crystallization of water. Tagami et al., (1999) studied the solidification of magnetically levitating water in hybrid magnet consisting of an inner water-cooled magnet and an outer superconducting magnet capable of generating up to 23 T magnetic fields.

During their experiments the authors observed that the nucleation of crystals was suppressed while the supercooled liquid state of water was stabilized. The solidification began abruptly forming the ice shell instantaneously, but the adiabatic condition decelerated the following solidification due to the fact that a transferring pass of the freezing latent heat was nonexistent (Tagami et al., 1999).
In 2000, the effect of a static magnetic field on the solidification behavior of water drops under the atmospheric conditions was studied by Aleksandrov et al. They applied MF strength ranged from 0 T to 0.505 T while the degree of supercooling decreased from 7.6 °C to almost 0 °C respectively. These results indicated that as the degree of supercooling reached the threshold value of 0 °C under the application of 0.505 T. Inaba et al., (2004) carried out a study regarding the effects of a magnetic field on the melting transition of purified water and heavy water using high resolution and supersensitive differential scanning calorimeter. Their results indicated that for both tested forms of water the melting temperature was higher under a magnetic field of 6 T. The temperature shifts of the phase transition due to the magnetic field were proportional to the square of the magnetic field. Suzuki et al., (2011) investigated the effect of weak magnetic field (about 0.0005 T) on the freezing process of several kinds of foods by means of a specially designed freezer facilitated with a magnetic field generator. The investigation included the comparison of freezing curves, drip loss, physicochemical evaluations on colour and texture, observation of microstructure, and sensory evaluation. The authors mentioned that compared to the control experiments a weak magnetic field of around 0.0005 T made no significant difference on temperature history during freezing and on the qualities of frozen foods within their experimental conditions. Their results are in accordance with the aforementioned studies concerning the influence of a MF strength on liquid water.
The application of magnetic fields on water's crystallization could have positive results on the quality of the frozen material. Despite the tentative steps of the scientists to investigate this area, promising results have arisen. Therefore, the application of magnetic disturbances during freezing could be a challenging area for an in depth investigation.

5. FREEZING UNDER ELECTRIC FIELD

The electrofreezing phenomenon applied to water were described by Pruppacher in 1973; an externally applied strong electric fields can promote the ice nucleation of water droplets. All these very early experimental studies were mostly related to the behavior of water drops in the thunderclouds (Pruppacher, 1973); (Wang and Pruppacher 1980); (Martin et al., 1980). Electrofreezing studies deal either with the application of direct current between charged electrodes or with the exposition of supercooled samples to an external electrostatic field (alternative (AC) or direct (DC)). In 1981 Shichiri and Nagata investigated the influence of the DC electrical current applied with various electrode materials on the nucleation temperature of ice crystals in supercooled distilled water. The authors observed a spontaneous nucleation on the vessel wall and a rise of the nucleation temperature when DC electric current was applied. The tested direct current ranged from 0.1 mA to 1.0 mA. They observed that ice nucleation occurred at electrodes at lower degree of supercooling and more specifically, when the current was growing higher the degree of supercooling was decreased. Many other studies are available on this field showing the importance of the material of the electrodes on freezing.
More recent experimental studies, concerning the influence of SEF on crystallization of water during freezing, showed that SEF application could cause differences in the nucleation process (Wei et al., 2008); (Orlowska et al., 2009); (Xanthakis et al., 2013). In all three cases, original experimental set ups were developed in which the samples were not in contact with the plate electrodes which were charged by a high voltage generator in order to create SEF. This structure of the experimental set ups was designed in order to ensure that current flow wouldn't occur and in result any possible electrolytic processes would be eliminated. At this point, an explanatory parenthesis is deemed crucial to be placed concerning the electric field estimation by the authors (Orlowska et al., 2009); (Wei et al., 2008). The authors used the basic equation (8) for calculating the electric field (E) based on the applied electric potential (V) and the distance (d) between the electrodes.

$$E = \frac{V}{d} \tag{8}$$

This equation would be correct under the premise that the sample it is homogenous, it is in contact with the electrodes and there are not any interactions with the other materials which could warp the streamlines of the electric field. But in these cases the different dielectric permittivities of the materials which constitute the systems influence the electric field distribution. Havet et al., (2009) performed numerical simulations in order to analyze the electric field strength and distribution with respect to Orlowska's et al., (2009) experimental set up. Their results indicated that the electric field is warped and its strength can be even twenty times lower than the value deduced from the basic approach (Figure 4). Therefore, the values for the strength of electric fields, can be used to indicate the difference of the field strength when the same methodology is followed, but not to compare the impact of the field when different experimental set up and working materials are being used.

Orlowska's et al., (2009) showed that the supercooling of water and aqueous solutions can be significantly reduced with increasing SEF. In 2013, Xanthakis et al., studied the impact of SEF application during freezing of a real food matrix, sample of pork tenderloin (1 g). The experimental set up was similar to the one used by Orlowska et al., (2009) apart from modifications made on the sample holder. The maximal effect was recorded at a voltage of 12kV. Under this level of electric field, the nucleation temperature was -2.66 ± 1.55 °C while in the case of electric field absence it was detected at -4.82 ± 1.31 °C.

The experimental results obtained by Wei et al., (2008), Orlowska et al., (2009) and Xanthakis et al., (2013) are in good agreement with the theoretical studies on the influence of electrostatic field on water properties. The cacophony between the experimental results and the theoretical approaches lies in the fact that water crystallization can be induced at electrostatic fields of much lower magnitudes than it was predicted on the basis of results from computer simulation analyses (Svishchev and Kusalik 1994); (Jung et al., 1999); (Sun et al., 2008). Although the controversial results between simulations and experimental studies are limited to the estimated effectiveness of an applied electric field, they are in symphony concerning the general theory. Certainly, the application of electric fields on phase changes of water offers great opportunities for research as several challenging issues still remain to be solved. Last but not least, the recent studies concerning the improvement of the microstructure of a frozen food matrix highlight the potentials for applications in the food industry.

6. LIPIDS UNDER ELECTRIC FIELD

Yan et al., (1997) carried out tests on dielectric material (butter composed of six fatty acids with typical properties of polymorphism) under EF freezing. Lipids are another important food component and are contained in several frozen food matrices. During the experiments voltage of 15 kV was applied to the rod electrode which was placed inside the butter while the temperature of the cooling source was set to be constant at 15 °C. The time for appearance of the crystals was reduced and the crystallization process was accelerated with high voltage. Although they didn't conduct an investigation on the structure of the crystals, their DSC results concerning the heat flow melting curve indicated that the crystals formed under EF freezing were more stable compared with the untreated (0 kV) sample.

7. OSCILLATING ELECTROMAGNETIC FIELDS

As it was assumed by the aforementioned studies, water molecule reacts when electric or magnetic fields try to disturb its tranquility. Electrical and magnetic disturbances are factors that could rearrange the hydrogen-bonded network of water. Herein, studies regarding the application of electric and magnetic or oscillating fields will be reviewed. Concerning the electromagnetic effects on water Chaplin (2013) mentioned that the presence of electromagnetic fields (EMF) which attempt to reorient the water molecules means that some hydrogen bonds of the network will be broken. He referred that the influence of electromagnetic radiation on water lies in the electrical rather than the magnetic impact. Chaplin (2013) refers that further than microwaves (central frequency 2.45 GHz) (MW) or radiofrequency irradiation (RF) even extremely low frequency fields (ELF) (3 Hz – 300 Hz) have significant and lasting effects on liquid water.

In 2012, a study concerning the application of radiofrequency EMF during freezing of meat was published by Anese et al. A pilot scale RF equipment was used as a RF source of energy while liquid nitrogen was flowing in a chamber in order to provide cryogenic freezing conditions to their samples. Frozen samples were prepared by RF assisted cryogenic freezing but also as control samples by cryogenic and air freezing (Figure 6). The working food matrix selected for this study consisted of pork tenderloin pieces (16 cm x 11 cm x 6 cm). Although RF pulses with various voltages were applied, a meat sample was completely frozen only at 2 kV. The various frozen samples were subsequently analyzed with respect to color, drip loss and firmness after thawing. The authors indicated that no significant effect was observed concerning the color between the differently frozen samples. Their results regarding the drip loss after 25 h of thermal equilibration at 10 °C showed that the RF assisted the frozen sample to lose only 1.5 % of its weight while under the same conditions the drip loss for air and cryogenically frozen meat was around 4 %. A similar work has been carried out by Xanthakis (2014-a) considering microwave disturbances instead of RF. Pork tenderloin was considered; microwaves was applied from time to time during cooling and resulted in temperature oscillation in the sample. The cooling rate during phase change (freezing) was slowed down when microwave was applied. The degree of supercooling was decreased up to 92% when MW was applied. A decrease of the size of the ice crystals of up to 62% was observed in the meat sample in comparison the conventional freezing process. The explanation proposed by the authors were that i) the temperature oscillation during freezing yield in a higher secondary nucleation and/or ii) that the MW disrupt the ice crystals. The two latest study indicated new potential concerning the application of electrical and magnetic disturbances on the phase change of water in food during freezing.

8. CONCLUSION

In this chapter the highlights of a literature survey concerning the area of freezing under the influence of electric and magnetic fields were outlined. Most of the aforementioned research studies focused mainly on the water properties when electric, magnetic or oscillating fields were applied during freezing. Freezing under these conditions has arisen as an innovative freezing process which can induce controlled ice nucleation and influence the morphology of the formed ice crystals. Water constitutes one of the major food components and greatly affects food quality. Therefore, these techniques are promising tools for improving food preservation through freezing. Recent applications of static electric fields and radiofrequency oscillating fields on food matrices showed that the damage of microstructure due to ice crystals was significantly minimized. Although there are still several blurry issues to be clarified concerning the impact of electrical and magnetic disturbances on water's behavior, the application of such conditions on food processing and preservation is an emerging and auspicious scientific area. Consequently, further research in order to reveal the effects of electric fields alone or in combination with magnetic fields, either static or oscillated, on food freezing is still necessary to be undertaken.

Abbreviations

AC – alternating current
CPAs – cryoprotective agents
DC – direct current
DSC – differential scanning calorimetry
EF – electrofreezing
HES – hydroxyethylostarch
MW – micro waves
NMR – nuclear magnetic resonance
PTFE – polytetrafluoroethylene (Teflon®)
RDF – radial distribution function
RF – radio frequency
SEF – static electric field
STM – scanning tunneling microscope

Nomenclature

E – electrostatic field (V/m)
ΔG – Gibbs free energy (J)
ΔG_0 – free energy of the system without applied electrostatic field (J)
ΔG_E – free energy of the system with applied electrostatic field (J)
ΔG_v – free energy of melting per unit volume (J/m^3)
ΔGs – free energy of surface (J/m^3)
p – pressure (Pa)
P – permanent polarization (C/m^2)
r – radius of the nuclei (m)
S – entropy (J/K)
T – temperature (K)
U – inner energy (J)
V – volume (m3)
V_c – volume of the system subjected to electrostatic field (m^3)
ε_o – vacuum permittivity, $\varepsilon_0 = 8.85410^{-12}$ ($A^2s^4kg^{-1}m^{-3}$)
ε_r – relative permittivity of the system
γ – surface free energy of the crystal fluid interface (J/m^2)
π – circumference of any circle, divided by its diameter, π= 3.14159
ΔQ – heat of crystallization
d – interelectrode distance
EqD – equivalent circular diameter (µm)

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