STUDY OF INDIVIDUAL QUICK FREEZING USING LIQUID NITROGEN: AN ECOLOGICAL FOODS FREEZING TECHNIQUE

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Abstract

Freezing represent one of the most utilized food preservation methods. Classical freezing techniques have harmful environmental effects especially because of the refrigerants, which contribute to the ozone layer depletion and to the increase of the greenhouse effect. Individual quick freezing (IQF) by direct contact with cryogenic agents such as liquid nitrogen (LN) has a significantly reduced ecological footprint, since nitrogen is a component of atmospheric air, and therefore it does not pollute. Liquid nitrogen is a by-product of air liquefaction which otherwise should be disposed. The effectiveness of liquid nitrogen IQF is vastly superior to that obtained by means of classical methods. The paper aims at highlighting this superiority, reflected mainly by the greatly reduced freezing time. The Lacroix and Castaigne method is used to determine the total time necessary to reach the prescribed freezing temperature in the thermal center of the product (in this case, green peas). This way, we prove that individual quick freezing using liquid nitrogen is characterized by much higher process rates and in addition it benefits from using an environmentally friendly refrigerant, thus contributing to sustainable development.

Key words: individual quick freezing, liquid nitrogen, freezing time, environmentally friendly refrigerants, sustainable development

Individual quick freezing (IQF) with direct contact with cryogenic agents represents a modern freezing technique for small size foods, such as fish fillets, shrimp, fruit – blackberries, raspberries, strawberries, and vegetables – green peas, diced carrots, etc. (Archer L.A., 2004). This way dehydration is avoided (Chourot J.M., Lauwers J., Massoji N., Lucas T., 2001), the method is very well suited for continuous feeding processes, and the cold losses are much lesser (7%) than those of the air-blast technique, characterized by 20 ... 30% losses (Horbaniuc B., 2007). Classical freezing methods using air or brines involve the employment of freons or ammonia, which either put the ozone layer in jeopardy and act as greenhouse gases (freons), or are toxic (ammonia).

The issue of designing environmentally friendly refrigeration processes is critical, since these represent a sustainable development component in agriculture and in related industries (Robu B., Ioan C.C, Robu E., Macoveanu M., 2009).

Sustainable development concepts, which are permanently renewed and enriched, direct towards seeking for new paths to attain specific desiderates based on the creation of an ecological attitude in setting the foundations and in defining innovative refrigeration techniques and methods (Ioan C.C., Horbaniuc B., Dumitraşcu Gh., 2005), (Ioan C.C., 2008). In the area of freezing, one of such methods is IQF using liquid nitrogen (LN).

Liquid nitrogen has the advantage of being a by-product of air liquefaction, which otherwise would be disposed in the atmosphere, having no other significant applications. Being an atmospheric air component, nitrogen is totally harmless and this is why it represents the ideal cryogenic agent from the standpoint of its environmental impact. Other reasons that lead to the adoption of this cryogenic agent are related to its properties (odorless, colorless, chemically inert, non flammable, non combustible, does not explode) and to the fact that it reduces product dehydration to a minimum – under 1% (Horbaniuc B., 2006), (Horbaniuc B., 2007).

When using LN as a freezing agent, 48% of the heat removed from the product is turned into latent heat of vaporization and 52% into sensible heat.

The advantages of liquid nitrogen individual quick freezing (LNIQF) are:

- low freezing time;
- good preservation of the initial product properties, which results in a very high food

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quality;
- small size of the freezing equipment;
- almost complete automation possible;
- high productivity.

LNIQF can be performed in two ways:

1. by immersion − nowadays abandoned, because it only uses the latent heat of vaporization and produces cracks on the product's surface (Pham Q.T., Le Bail A., Hayert M., Tremeac B., 2005);
2. by spraying − products pre-cooled in the gaseous nitrogen stream resulted from the vaporization of the cryogenic agent are sprayed with LN and thus the entire cooling capacity is used. One ton of LN has the equivalent of 100 kW of mechanical refrigeration power.

Nitrogen has a drawback: when in high concentration in air, it causes suffocation. Therefore, work safety prescriptions in work places where LN freezing facilities are in operation are very severe, a very effective ventilation being required.

Liquid nitrogen is transported in special tanks and is subsequently transferred at the consumer in 20 … 50 t thermally insulated tanks where it is stored at atmospheric pressure. The temperature of the liquid agent is maintained by controlled release of the vapor resulting from the isobaric boiling of the nitrogen. Losses are of the order of 0.5% per day.

Due to its extremely low temperature, use of LN requires special precautions. Atmospheric oxygen, which boils at −183°C, may condense on un-insulated surfaces of the freezing equipment that have the temperature of LN and liquid oxygen drops may fall on on organic materials such as lubricants, rubber, etc., which instantaneously burst into flames, the combustion being explosive. Usual materials such as carbon-steel, rubber, plastics, which parts of the equipment are made of become brittle at LN temperature and therefore one must use other materials that are less affected by low temperatures (stainless steel, copper, aluminum, etc.).

MATERIAL AND METHOD

The main advantage of LNIQF is represented by the high rate of the process, which results in reduced residence times of the product in the freezing equipment. In order to design it, one needs to determine the total time necessary for the temperature in the thermal center of the product to reach the prescribed value.

It is difficult to calculate this time, because the process is composed of three stages (pre-cooling, freezing, and subcooling), during which the thermophysical properties of the material have different values, and during freezing, the phase transition of the contained water takes place. Moreover, the shape of the products is complex, which results in further complications of the numerical modeling (Perusello C. A., Mariani V. C. Amarante A.C., 2011).

The methods used to determine the total freezing time are very diversified: analytical (Zorilla E., Rubiolo A., 2005a), (Zorilla E., Rubiolo A., 2005b), (Frolov S.V., 1997), numerical (Santos M.V., Lespinard A.R., 2011), semi-empirical (Lacroix C., Castaigne F., 1988), (Becker B.R., Fricke B., 1997), (Becker B.R, 1999), etc.

In order to get a quick and at the same time sufficiently accurate estimation of the freezing time, Plank's equation is usually applied for simple geometrical shapes (Plank, R., 1941):

$$\tau = \frac{L_v}{T_s - T_m}\left(\frac{PD}{\alpha} + \frac{RD^2}{\lambda_f}\right) \qquad (1)$$

where τ is the freezing time, L_v is the volumetric latent heat of freezing, T_s is the freezing temperature, T_m is the temperature of the cooling medium, D is the characteristic length (in the case of spheres, this length is the diameter), α is the convective heat transfer coefficient, λ_f is the thermal conductivity of the product at the final temperature, and P and R are factors that take into account the geometry of the product.

Since one needs to take into account the pre-cooling and cooling times, we have considered a more elaborated model, derived from that of Plank, which has been proposed by Lacroix and Castaigne and that allows to also determine these times (Lacroix C., Castaigne F., 1988). According to this model, the total time τ_{total} necessary to reach the prescribed temperature in the thermal center of the product is the sum of the three times: pre-cooling (τ_1), freezing (τ_2), and subcooling (τ_3):

$$\tau_{total} = \tau_1 + \tau_2 + \tau_3 \qquad (2)$$

The expressions for the three intervals are (Lacroix C., Castaigne F., 1988):

$$\tau_1 = f_1 \lg\left(j_1 \frac{T_m - T_0}{T_m - T_s}\right) \qquad (3)$$

where f_1 and j_1 are coefficients that depend on the shape of the product and of the Biot number, and T_0 is the initial temperature of the product.

$$\tau_2 = \frac{L_v D^2}{(T_s - T_m)\lambda_f}\left(\frac{P}{2Bi_f} + R\right) \qquad (4)$$

where P and R are functions of the geometrical shape of the product, and Bi_f is the Biot number for freezing, defined by:

$$Bi_f = \frac{\alpha D}{\lambda_f} \qquad (5)$$

$$\tau_3 = f_3 \lg\left(j_3 \frac{T_m - T_s}{T_m - T_c}\right) \qquad (6)$$

where f_3 and j_3 are coefficients that depend on the the shape of the product and of the Biot number, and T_c is the final temperature of the process, measured in the thermal center of the product.

Table 1

Calculus of coefficients *f* and *j* for spheres (Lacroix C., Castaigne F., 1987)

Range for the Biot number	Equations for *f* and *j*
$Bi \le 0.1$	$\frac{fa}{L^2} = \frac{\ln 10}{3Bi}$ $j = 1.0$
$0.1 < Bi \le 100$	$\frac{fa}{L^2} = \frac{\ln 10}{w^2}$ $j = \frac{2(\sin w - w\cos w)}{w - \sin w \cos w}$ $w = 1.573729 + 0.642906 \ln Bi + 0.047859(\ln Bi)^2 - 0.03553(\ln Bi)^3 -$ $- 0.004907(\ln Bi)^4 + 0.001563(\ln Bi)^5$
$Bi > 100$	$\frac{fa}{L^2} = 0.2333$ $j = 2.0$

In the equations from *table 1*, *a* is the thermal diffusivity of the product, and *L* is a characteristic length equal to the radius in this case.

RESULTS AND DISCUSSION

We chose a product of spherical shape, namely green peas. For this geometrical shape, the coefficients P and R have the values P = 0.19665 and R = 0.03939 respectively (Lacroix C., Castaigne F., 1987). From the same paper published by Lacroix and Castaigne, *table 1* presents the equations for the factors *f* and *j* for spherical products. The thermophysical properties of the product have been compiled from (Barbos-Cànovas G.V., Juliano P., Peleg M., 1999) and (Iliescu Gh., Vasile C., 1982).

The diameter of the green pea beans has been selected in the range 6 … 12 mm and the intensity of the cooling process has been considered by means of the convection heat transfer coefficient α, the values of which have been chosen in the range 30 ... 300 W/m^2 K.

The considered temperatures were:

- the initial temperature of the product: t_0 = 20°C;
- the freezing temperature: $t_s = -1.3$°C;
- liquid nitrogen temperature: $t_m = -198$°C;
- the final temperature of the product in the thermal center: $t_c = -28$°C.

With these entry data, we have calculated the times of the three stages and the total freezing time and the results have been plotted in *figs. 1…3*.

The plot in *fig. 1* presents the variation (versus the diameter) of the time intervals corresponding to pre-cooling (τ_1), freezing (τ_2), subcooling (τ_3) and of the total time (τ_{total}) respectively for a value of 150 W/m^2 K of the convective heat transfer coefficient, which can be easily achieved by the LNIQF method. As expected, as the pea diameter increases, the process stage times increase from τ_1 = 16.4 s, τ_2 = 5.2 s, τ_1 = 4.9 s for D = 6 mm, to τ_1 = 57.9 s, τ_2 = 13.6 s, τ_1 = 16.9 s for D = 12 mm, the total time varying in the range 26.5 ... 88.4 s.

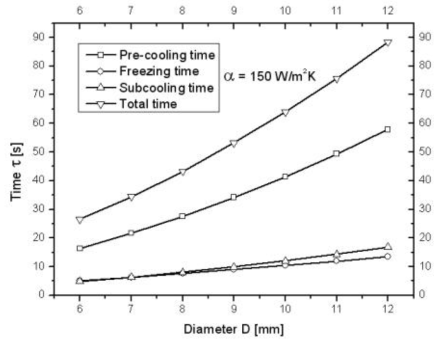

Figure 1 **Time intervals for the three stages and the total freezing time versus the diameter, for α = 150W/m^2 K**

The plot of the partial and total times versus the intensity of the convection heat transfer for 9 mm diameter peas is presented in *fig. 2*.

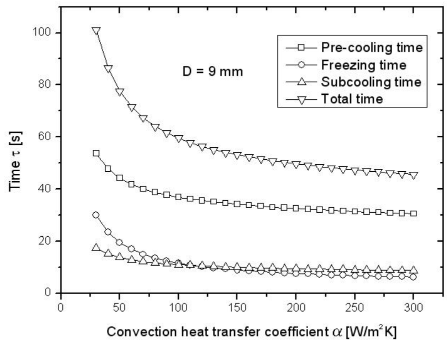

Figure 2 **Time intervals for the three stages and the total freezing time versus α, for D = 9 mm**

One notices that the partial times decrease as α increases, but this decrease is less important (about 43% for τ_1, respectively 50% for τ_3). The freezing time τ_2 represents an exception, exhibiting a more pronounced variation of about 71%.

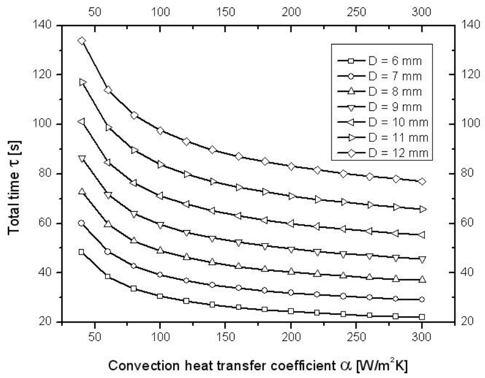

Figure 3 **The total freezing time versus α, for diameter values within the considered range**

The total time varies by approx. 45%. Overall, one notices a relatively little variation of partial and total times, although the convective heat transfer coefficient varies within very large limits (30 … 300 W/m^2 K). Its less significant influence can be explained by the important temperature difference between the product and the cryogenic agent. Consequently, measures to increase α are not absolutely necessary, a fact that does not happen in the case of air-blast IQF equipment, where it is crucially important to obtain the highest possible values of this coefficient. This way, important savings are achieved in terms of IQF equipment design and its energy consumption during operation. Noting that the time plot versus α is steeper for small values of the latter and then flattens, it results that it is not economically justified to achieve high values of α because the gain ceases to be significant. Therefore, in the case of the example considered in *fig. 2*, a value of 120 W/m^2 K is optimal from the economical standpoint. This is in fact the value usually achieved in LNIQF plants (Horbaniuc B., 2007), (Kennedy C.J., 2000). The total freezing time versus the convective heat transfer coefficient is plotted in *fig. 3* for the chosen values of the diameter. One has to notice the high process rate, even in the case of large size products, which leads to a high productivity of LNIQF installations compared to that of classical ones, either air or brine operated. This can be explained by the large temperature difference between the product's surface and the freezing medium, which determines a very high intensity of the process. Also, all of the plots have the same shape, exhibiting a quasi-flatness for values of α higher than 120 W/m^2K.

CONCLUSIONS

Individual quick freezing using liquid nitrogen is a high productivity food freezing technique characterized by low environmental impact due to the use of LN as a by-product of air liquefaction units.

The verification of the Lacroix-Castaigne model for the calculus of the partial and total times of the freezing process in the case of green peas has led to the following conclusions:

1. The greater the diameter of the product, the longer the process, as a result of the increase of the pre-cooling, freezing and subcooling time intervals.

2. The magnitude of the variation of pre-cooling and subcooling times for a fixed diameter as a function of the convection heat transfer α is low, whereas that of the freezing time τ_2 is a little more pronounced. These effects are transferred on the total freezing time τ_{total} so as to determine a steep decrease in the domain of low values of α followed by an almost insignificant diminution at high values of the convection heat transfer. This behavior makes heat transfer coefficient enhancement measures unnecessary above 120 W/m^2 K, which can be considered as the optimal value.

3. Regardless of the product diameter, the process rate is much higher than in the case of air-blast IQF, and the total freezing time plots have all the same shape, exhibiting an insignificant decrease as α increases in the domain of high values. Liquid nitrogen equipment is similar to that using classical agents and is used for IQF or pre-freezing (frozen crust building on the surface of sensitive products). The typical residence time is of the order of seconds, and the maximum productivity can be up to 5 t/h. From the configuration standpoint, the cryogenic spraying equipment is practically identical to air-blast one: tunnels, spirals, and with discontinuous (batch) feeding.

Tunnels are the cryogenic version of air-blast tunnels. They are provided with nozzles that spray liquid nitrogen on the products that are placed on a moving belt. In cryogenic tunnels freezing rates are much higher than those of air-blast ones and the versatility is highly superior since while in air-blast tunnels the cooling rate is fixed from design by the maximum speed of the fans that blow the air over the heat transfer surface of the evaporator, in LN tunnels it can be adjusted by modifying the flow rate of the cryogenic agent that is sprayed by the nozzles. Some of the cryogenic tunnels (the simple ones) consist of two zones: a spraying region, where the product is sprayed with LN and a circulation one, where the gaseous nitrogen resulted in the first zone, being still very cold, finishes the freezing. At the more sophisticated ones the spray nozzles are distributed along the product path, allowing a very accurate temperature tuning by adjusting the flow rate of each nozzle. Thus, the cooling load can be put into correspondence with the cryogenic agent consumption and therefore, the outlet temperature of the latter results close to the final temperature of the product, leading to important energy savings. A special issue arises in equipment having a single spraying region because of the imprecise temperature control, which results in a poor cooling potential recovery of LN, which leaves the installation at a temperature much different from that of the product.

A correct set up of the gaseous agent circulation results in an effective and energy-saving operation of the equipment. An example is represented by the tunnel Cryo-Quick VT manufactured by the American company Air Products, where a specially-shaped ceiling induces two eddies which facilitate a very effective mixing and recirculation of the gaseous agent (Kennedy C. J., 2000). Thus, the heat transfer coefficient reaches values in excess of 120 W/m^2K, which is the double of that achieved by an air-blast tunnel. The fans are placed underneath the belt in order to obtain a better circulation of the agent.

The comparison criterion of LNIQF equipment is the specific consumption that is the amount of cryogenic agent consumed to freeze one kilogram of product. Typically, the specific consumption of cryogenic tunnels ranges between 0.3 for low humidity products to 2 for "difficult" ones (sea food for example).

Spirals are used when long residence times impose an unacceptable linear length of the belt and therefore, this length is "wrapped" in a spiral, thus resulting an acceptable size of the freezing equipment. Batch-fed equipment is less employed because of its lower productivity caused by "dead" times necessary to load/unload the products.

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