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STUDY ON DETERMINING THE FREEZING MODE OF FROZEN FILLET BIGEYE TUNA (*THUNNUS OBESUS*)

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ABSTRACT

Bigeye tuna (*Thunnus obesus*), a kind of delicious seafood, can be processed to be several valuable products. To maintain the product quality, harvested tunas had been strictly persevered, transported and frozen at low temperature. This study was carried out to determine the technological mode of the freezing process of the fillet tuna to find the optimum temperature and freezing time to reduce mass loss and keep its quality. The combining two-level orthogonal arrays was used to build the relationship between objective functions and income variables. The results found the optimized freezing mode of the fillet tuna: the freezing environment temperature was -42.5°C and the freezing time was 2.12h. Carrying out the experiment with optimized freezing mode, showed that the temperature at the end of the fillet tuna freezing process was reached at –22.5°C and the yield of weight loss was ~~is~~ 3.1%. That meaned all internal water of the product was completely crystallized and the loss of quality was negligible. The freezing mode can be applied in industrial scale for the frozen fillet tuna manufacturing process.

1. Introduction

Bigeye tuna (Thunnus obesus), commonly known as Bò Gù (Vietnamese local name), is a large sea fish belonging to the Scombridae (mackerel) family (Graham J. B. and Dickson K. A., 2004), living in the warm sea, 185 km far from the coastline. In Vietnam, the tuna lives mainly in Binh Dinh, Phu Yen and Khanh Hoa provinces (Lewis A. D.., 2005). The tuna, a kind of delicious seafood, having big and nutritious eyes, can be processed into various delicious dishes and made to be several valuable products for domestic consumption or for exporting to oversea markets. The harvested tunas had been strictly persevered and transported to factories (Lewis A. D.., 2005), in which the tunas were filleted to be plate pieces in size of 300×150×20 mm (length×width×thickness), the tuna chemical ingredients were shown in Table 1 and Table 2.

Table 1. Chemical ingredients and energy in 100g fillet bigeye tuna

Water (wet) (g)	*Protein (g)*	*Lipid (g)*	*Ash~~es~~ (g)*	*Energy (kcal)*
72.6	23.4	1.8	2.2	105

Table 1 and Table 2 showed that the bigeye tunas are rich of nutrients, and are the suitable environment for the growing and developing of bacteria. Thus, during post-harvesting, if it is not having suitable methods for preservation, the quality of the tuna will be rapidly reduced by the effect of internal enzymes, domestic or contaminated bacteria. These effects lead to the debasement of using and economical values of tunas. To preserve fillet bigeye tunas for domestic consumption and exporting purpose, the freezing method is commonly used to preserve the tunas in seafood processing factories, because only this method can lower the rate of quality decreasing of the products (Dzung N.T, Dzung T.V and Ba T.D, 2012; Dzung N.T., 2015).

Table 2. Chemical ingredients in 100g fillet bigeye~~s~~ tuna

Mineral				
Calcium (mg)	*Phosphor (mg)*	*Ferrous (mg)*	*Sodium (mg)*	*Potassium (mg)*
68	482	2.4	7.4	6.8
Vitamin				
A (μg)	*B1 (mg)*	*B2 (mg)*	*PP (mg)*	*C (mg)*
162	0.32	0.53	20.4	0.12

However, when using the freezing method for tuna preservation, some of the following problems should be noted:

Firstly, the products at the end of the freezing process have to reach an optimum temperature (Dzung N.T, Dzung T.V and Ba T.D, 2012; Dzung N.T., 2015). At that time, all liquid water inside the products has been completely crystallized then the shelf-life has been extended. At this temperature, the operating freezing systems can be stopped to save energy and product costs.

Secondly, if the freezing rate is low, the size of crystalized water inside the tissue of products will be large lead to destroying the texture of cells, when the frozen products are defrosted, the fluid in cells will be leaked, and it causes the large mass loss of products. Then, their quality will be decreased because nutrition in the cell fluid will be loosed. If the freezing rate is quick or super quick, the water inside the products is crystallized at its position, and the size of the crystallized water is small in micro or nano size, it does not destroy the texture of cells. When the products are defrosted, their quality are kept as that of initial. But quick frozen or super quick frozen have to be carried out in the deep low temperature, lower than –40°C, since the freezers will spend a large amount of energy. These are the unexpected problems occurring in manufacturing (Dzung N.T, Dzung T.V and Ba T.D, 2012; Dzung N.T., 2015).

Thus, the problem that needs to be solved is how to build the optimum freezing temperature and how to minimize The freezing time making products – the frozen fillet bigeye tunas in size of 300 mm × 150 mm × 20 mm – reach the optimum temperature, making all water inside the products is crystallized, and making the mass loss of post-defrosted frozen products is lowest.

According to some authors *(*Cleland A. C. and Earle R. L.., 1976; Holman J.., 2009; Dzung N.T., 2015), crystallizing temperature of fillet tuna internal water is –1.24°C, and the average temperature of crystallized water inside the final frozen fillet tunas are –22.5°C – the Eutectic point of internal water of products. There are the fundamentals to build the optimized mathematical problem to find the freezing technology of the frozen fillet bigeye tuna.

2. Materials and methods

2.1. Materials

Whole harvested bigeye tunas caught from Binh Dinh to Khanh Hoa, were stored at –45°C to –40°C, then they were transported to factories. At the factories, they were filleted and cut to be plat pieces in size of 300mm length, 150mm width, and 20mm thick (see 01), each piece was put into a PE bag to avoid loss of water and cold burning on the surface, put into trays and frozen (Charm S. E. and Slavin J.., 1962; Heldman D. R.., 1982; Dzung N.T., 2014).

Figure 1. Frozen fillet bigeye tuna pieces

2.2. Apparatus

Scale Sartorius, Basic Type BA310S ((Sartorius, Germany): accuracy class: F1 (Guided by International Organization Of Legal Metrology), Dual Digital Thermometer – range (−50 ÷ 70)°C, readability ± 0.05°C – (Omron, Japan), the freezer DL-3 – build by the Faculty of Chemical and Food Technology, Ho Chi Minh city University of Technology and Education, Vietnam (02) – controllable freezing temperature: lowest −50°C, the cooling system controlled by computer (Dzung N.T., 2015).

Figure 2. Freezing system DL-3

2.3. Methods

In this study, some methods had been used:

2.3.1. Temperature measuring

The temperature of the freezing environment was measured by temperature sensors, and calculated automatically by a computer (Charm S. E. and Slavin J.., 1962; Dzung N.T., 2014). The temperature of the surface and center of the frozen fillet tuna fish were measured by the Dual Digital Thermometer. To measure the center temperature of the fillet tuna products at the end of freezing, the lenght of the PT100 sensor was placed paralelled to the length of the frozen piece so that the sensor was inside of the center position of the piece

Average final temperature of the frozen fillet tuna pieces was determined by the following formula (Dzung N.T, Dzung T.V and Ba T.D, 2012; Dzung N.T., 2015):

$$Y_1 = T_a = \frac{T_s + T_c}{2} \quad (1)$$

Where: T_a, T_s, T_c (°C) were average, surface and center temperatures of the fillet tuna products at the end of freezing, respectively.

2.3.2. Defrosting of frozen fillet tuna

Frozen fillet tuna pieces were defrosted by 3.5 MHz ultrasonic at 28°C cooling chamber for 45 minutes. The ultrasonic was regularly turned on and off every minute.

2.3.3. Mass loss determining

Mass of the pre-frozen fillet tuna products, and that of post-defrosted frozen fillet tunas were determined by the Scale (Sartorius), and the yield of mass loss of post-defrosted frozen fillet tuna products was calculated by formula (Dzung N.T, Dzung T.V and Ba T.D, 2012; Dzung N.T., 2015):

$$Y_2 = \frac{m_1 - m_2}{m_1}.100\% = \frac{\Delta m}{m_1}.100\% \quad (2)$$

Where: m_1, m_2 (g) were mass of the pre-frozen and post-defrosted frozen fillet tuna products, respectively.

Y_2 (%) Yield of mass loss of post-defrosted frozen fillet tuna products.

2.3.4. Building the freezing mode

Using the combining two-level orthogonal arrays to build the relationship between objective functions (Y_1, Y_2) including average final temperature of frozen fillet tuna products (Y_1, °C), and the yield of mass loss of post-defrosted frozen fillet tuna products (Y_2, %); and technological factors (X_1, X_2) directly affecting to the process such as: temperature of freezing environment (X_1, °C), time of freezing process (X_2, h), and the thickness of the fillet tuna plat (X_3, mm). Because the thickness of the bigeye tuna is standardized as X_3 = 20mm, X_3 had been omitted to the variable sheet. Thus, the relationship between objective functions (Y_1, Y_2) and technological factors (X_1, X_2) is described by the following formula (Dzung N.T, et al., 2012):

$$Y = b_0 + \sum_{j=1}^{k} b_j x_j + \sum_{j \neq i; j=1}^{k} b_{ji} x_j x_i + \sum_{j=1}^{k} b_{jj} \left(x_j^2 - \lambda\right) \quad (3)$$

Where x_1, x_2 are variables coded from the real variables X_1, X_2 as following formula:

$$x_i = \frac{X_i - X_i^0}{\Delta X_i};$$

$$X_i = x_i.\Delta X_i + X_i^0 \quad (4)$$

Where:

$X_i^0 = (X_i^{max} + X_i^{min})/2;$

$\Delta X_i = (X_i^{max} - X_i^{min})/2;$

$X_i^{min} \leq X_i \leq X_i^{max}; i = 1 \div 2$

Number of experiment was identified by (Dzung N.T., 2012):

$$N = n_k + n_* + n_0 = 2^k + 2k + n_0 = 9 \quad (5)$$

Where:

$k = 2;$

$n_k = 2^k = 2^2 = 4;$

$n_* = 2k = 2\times2 = 4; n_0 = 1$

Swing arm of orthogonal matrix was identified (Dzung N.T., 2012):

$$\alpha = \sqrt{\sqrt{N.2^{(k-2)}} - 2^{(k-1)}} = \sqrt{\sqrt{9.2^{(2-2)}} - 2^{(2-1)}} = 1 \quad (6)$$

Conditions of empirical orthogonal matrix (Dzung N.T., 2012):

$$\lambda = \frac{1}{N}\left(2^k + 2\alpha^2\right) = \frac{1}{9}\left(2^2 + 2.1^2\right) = 2/3 \quad (7)$$

2.3.5. Other measurements

In addition, mathematical and information technical tools had been used to solve the optimized problems describing the freezing process, and to identify the technical models of the freezing process of fillet tuna pieces.

3. Results and discussions

3.1. Building the mathematical models of the freezing process of fillet tuna

The average final temperature of fillet tuna products at the end of the freezing process (Y_1, ºC), and the yield of mass loss of post-defrosted frozen fillet tuna products are dependent on the freezing process, and directly affected by technological factors such as: the temperature of freezing environment (X_1, ºC), time of the freezing process (X_2, h).

Technological factors X_1 and X_2 were individually surveyed to find the extreme domain of Y_1, Y_2. The vital surveying domain was found and shown in Table 3, (Dzung N.T., 2012).

The experiments, with levels of technological factors X_1, X_2 in Table 3, were carried out with the combining two-level orthogonal arrays shown in Table 4 to build the mathematical model $Y_1 = f_1(X_1, X_2) = f_1(x_1, x_2)$ và $Y_2 = f_2(X_1, X_2) = f_2(x_1, x_2)$ describing the freezing process. Results identified Y_1, Y_2 depended on x_1, x_2 shown in the column "Objective function values" of Table 4, (Dzung N.T., 2012).

MS Excel 2019 (Microsoft, US) was used to calculate the experimental data in Table 4, to calculate coefficients, b_j and b_{ji}. The significant coefficients of regression equations (3) were tested by Student's test, and the compatibility of the regression equation was tested by Fisher test (Fisher R.., 1929; Dzung N.T., 2012). Results built mathematical modeling of experiments describing the freezing process of the fillet tuna pieces.

Table 3. Levels of technological factors affect to freezing process of fillet tuna pieces

Factors		X_1 (ºC)	X_2 (h)
Levels of experiment	*- α (-1)*	-45	0.4
	Lower level, (-1)	-45	0.4
	Medium level, (0)	-40	2.2
	Upper level, (+1)	-35	4.0
	+α (+1)	-35	4.0
Variance range ΔZ_i		5	1.8

Table 4. The combining two-level orthogonal arrays and the experiment results finding objective function (Y_1, Y_2) of the freezing process of fillet tuna pieces

Number of experiments		Real variable		Coded variable		Objective function values	
		X_1, °C	X_2, h	x_1	x_2	Y_1	Y_2
2^k	1	–35	4.5	+1	+1	–27.77	5.15
	2	–45	4.5	–1	+1	–36.38	4.74
	3	–35	0.5	+1	–1	–2.19	1.52
	4	–45	0.5	–1	–1	–2.83	0.99
2k	5	–35	2.5	+1	0	–20.45	3.61
	6	–45	2.5	–1	0	–24.34	3.15
	7	–40	4.5	0	+1	–31.24	4.65
	8	–40	0.5	0	–1	–2.77	1.03
n_0	9	–40	2.5	0	0	–21.93	3.19

After testing the compatibility by Fisher test (Dzung N.T., 2012), the results confirmed that the mathematical models (8), (9) are totally compatible with the experimental data shown in Table 4. Thus, they can describe the freezing process of the fillet tuna.

3.1.1. *Temperature of fillet tuna products at the end of freezing process*

$$Y_1 = f_1(x_1, x_2) = -18.88 + 2.2x_1 - 14.59x_2 + 2.01x_1x_2 - 0.34\left(x_1^2 - \frac{2}{3}\right) + 5.04(x_2^2 - \frac{2}{3}) \quad (8)$$

3.1.2. *Yield of mass loss of post-defrosted frozen fillet tuna products*

$$Y_2 = f_2(x_1, x_2) = 3.11 + 0.231x_1 - 1.829x_2 - 0.03x_1x_2 + 0.24\left(x_1^2 - \frac{2}{3}\right) - 0.31(x_2^2 - \frac{2}{3}) \quad (9)$$

Simulating the mathematical models (8) and (9), the results received responsive or curved surfaces $Y_1 = f_1(x_1, x_2)$ and $Y_2 = f_2(x_1, x_2)$ shown in 03 and 04.

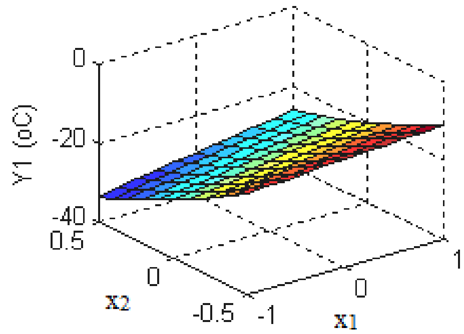


Figure 3. Relationship between frozen fillet tuna pieces and technological factors: temperature of freezing environment, and freezing time

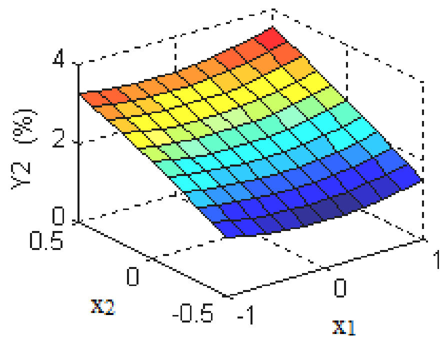


Figure 4. Relationship between yield of mass loss of post-defrosted of the frozen fillet tuna products and technological factors: temperature of freezing environment, and freezing time

The results showed that the relationship between objective functions (Y_1 (°C) and Y_2 (%)) and the technological factors (x_1 and x_2) directly affecting the freezing process, is the quadratic nonlinear. It is more complicated than that of first-order linear. Normally, the quadratic nonlinear function consists of two value ranges on two branches, one branch is covariant and the other is inverse depending on a quadratic coefficient. If the coefficient is negative, the parabola curve is convex. In contrast, the parabola curve is concave. The boundary point between these two branches is the extreme pitch of the parabola.

The formula (8) showed the coefficient 5.04 linking to x_2^2, thus it is misunderstood that if freezing time (x_2) is prolonging - time increase, the temperature of fillet tuna products at the end of freezing process will be increasing; It is the wrong situation. In fact, the freezing process, assessed in the negative temperatures (x_1), is belonging to the inverse brand. It means that when the freezing time is increasing, the product temperature during the freezing process will be decreasing, see 03. Similarly, the model (9) showed the quadratic coefficient –0.31 linking to x_2^2, thus the freezing process is assessed in the covariant brand of parabola, which means that when the time is increasing, the Yield of mass loss of post-defrosted frozen fillet tuna products will be increasing, see 04 (Dzung N.T., 2012).

3.2. Building and solving the optimized problems describing freezing process of fillet bigeye tuna

3.2.1. Building the optimized problems describing freezing process of fillet bigeye tuna

Based on technical conditions answering the requirements of economy and engineering, the freezing process of the fillet tuna (in size of 300mm×150mm×20mm) had been carried out as quick as possible, and not only whether all internal water of the tuna is completely crystallized or temperature of the product reaches the optimum temperature T_{Fopt} = –22.5°C but also the quality of post-frozen products have to be paid attention (Banin A. and Duwayne A. M.., 1974; Dzung N.T, et al., 2012). To solve this problem, the optimized problem had been established as follow: Let's determine the optimized problem $x^{opt} = (x_1^{opt}, x_2^{opt}) \in \Omega_x = \{-1 \le x_1, x_2 \le 1\}$ to:

$$\begin{cases} Y_1 = f_1(x_1^{opt}, x_2^{opt}) = T_{Fopt} = -22.5 \\ Y_{2\min} = f_2(x_1^{opt}, x_2^{opt}) = \mathrm{Min}\{f_2(x_1, x_2)\} \\ \forall x = (x_1, x_2) \in \Omega_x = \{-1 \le x_1, x_2 \le 1\} \end{cases} \quad (10)$$

3.2.2. Solving the optimized problems describing freezing process of fillet bigeye tuna

Solving the optimized problem (10) by Lagrange method as follow:

$$L(x_1, x_2, h) = f_2(x_1, x_2) + h.(f_1(x_1, x_2) + 22.5) \quad (11)$$

Find extremes by Lagrange method (11) as follow:

$$\begin{cases} \dfrac{\partial L(x_1, x_2, h)}{\partial x_1} = 0 \\ \dfrac{\partial L(x_1, x_2, h)}{\partial x_2} = 0 \\ \dfrac{\partial L(x_1, x_2, h)}{\partial h} = 0 \\ -1 \le x_1, x_2 \le 1 \end{cases} \quad (12)$$

Equation (12) can be written as follow

$$\begin{cases} 0.231 - 0.03x_2 + 0.048x_1 \\ \qquad + h \times (2.2 + 2.01x_2 - 0.68x_1) = 0 \\ -1.829 - 0.03x_1 - 0.62x_2 \\ \qquad + h \times (-14.59 + 2.01x_1 + 10.08x_2) = 0 \\ -18.88 + 2.2x_1 - 14.59x_2 + 2.01x_1x_2 - \\ \quad 0.34 \times \left(x_1^2 - \dfrac{2}{3}\right) + 5.04 \times \left(x_2^2 - \dfrac{2}{3}\right) = 0 \\ -1 \le x_1, x_2 \le 1 \end{cases} \quad (13)$$

With meshing algorithm under the supporting of Visual Basic 6.0 to solve equations (13), the results found that roots of the equations (10) together with Lagrange multiplier coefficient, h = 0.109, are:

$$x_1^{opt} = -0.502; \; x_2^{opt} = -0.044; \quad (14)$$

Converting to real variable by equation (4) received:

$$X_1 = -42.5^{o}C; \; X_2 = 2.12h \quad (15)$$

Replacing the optimal roots x_1^{opt} and x_2^{opt} into equations (8) and (9), found:

$$Y_1 = -22.5^{o}C; \qquad Y_2^{min} = 3.1\% \quad (16)$$

3.2.3. Verification of the found optimized problems described the freezing process of fillet tuna

Carrying out the Freezing of the fillet tuna (in size of 300mm×150mm×20mm) on the freezing system DL-3 at the optimized freezing modes: the environment temperature is –42.5°C, and kept constantly during freezing process, and the freezing time is 2.12h. Temperature of products and yield of mass loss were measured as described in ***Error! Reference source not f ound.***. At the end of the freezing process, the results are followed: the average temperature of the center products is –22.57°C, Yield of mass loss of post-defrosted frozen fillet tuna products is 3.19%.

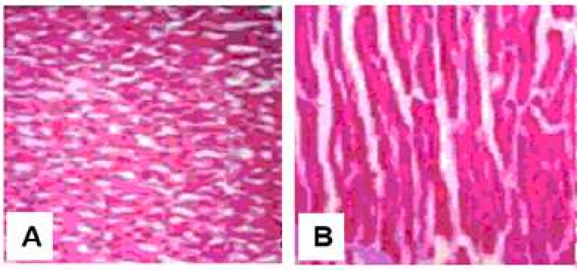


Figure 5. The SEM image of ice crystal structure in frozen fillet tuna in size of 300mm×150mm×20mm. A. The freezing mode: temperature of freezing environment, -42.5°C, and freezing time, 2.12h; B. The freezing mode: temperature of freezing environment, -38.55°C, and freezing time, 3.95h. The images A and B were scanned and captured at the same magnification (1000×)

It can be seen that the optimized freezing process model of fillet tuna is completely consistent with reality. This technical condition, thus, can be applied to real manufacturing to preserve the fillet tuna pieces to serve for domestic consumption and export. Besides, in this study, the fillet tuna had been frozen in another mode that differs from the optimized mode: the temperature of freezing environment –38.5°C to make the temperature of the products at the end of the freezing process reach –22.5°C then all internal water of products is crystallized, and the freezing time is 3.95h. At that time, yield of mass loss of post-defrosted frozen fillet tuna products is 6.79%.

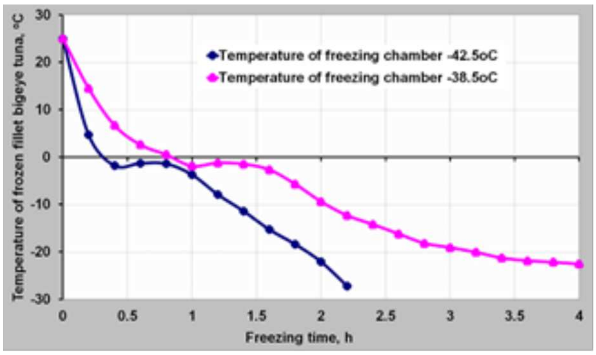
What is the reason why when the freezing environment temperature is –38.5°C, and the freezing time is 3.95h, the yield of mass loss of post-defrosted frozen fillet tuna products increases to 6.79%. The reason is that in the low rate freezing process, ice crystals in the products are formed in a large size (see Figure 5B), then they do destroy the texture of cell membranes and tore the tissue membranes (Banin A. and Duwayne A. M.., 1974; Haugvalstad G. H., Skipnes D. and Sivertsvik M.., 2005). When the products are defrosted to consume, the melting process of ices inside the products will attract cell fluid, lead to mass and nutrition loss will increase. When freezing at the optimized mode, the mass loss is very low and considered negligible. The reason is that ices inside the products are formed with a small size (see Figure 5A), they cause tiny injury to cell membranes. Then, when the products are defrosted, nutrients are maintained as initial, and the frozen products are high quality.

Table 5. Experimental data of the process of freezing of fillet tuna pieces: $T_1 = f_1(t)$ when temperature of freezing chamber -42.5°C; $T_2 = f_2(t)$ when temperature of freezing chamber -38.5°C

Time of freezing process, t (h)	Temperature of fillet tuna pieces $T_1 = f_1(t)$ when temperature of freezing chamber -42.5°C	Temperature of fillet tuna pieces $T_2 = f_2(t)$ when temperature of freezing chamber -38.5°C
0.0	25.0	25.0
0.2	4.7	14.5
0.4	-1.8	6.7
0.6	-1.3	2.6
0.8	-1.4	0.5
1.0	-3.7	-1.9
1.2	-7.9	-1.24
1.4	-11.4	-1.48
1.6	-15.3	-2.68
1.8	-18.4	-5.7
2.0	-22.1	-9.4
2.2	-27.2	-12.3

2.4		-14.2
2.6		-16.2
2.8		-18.2
3.0		-19.1
3.2		-20.1
3.4		-21.3
3.6		-21.9
3.8		-22.2
4.0		-22.6

Table 5 and Figure 6. showed that the crystallizing temperature of water in the fillet tuna products is –1.24°C, not 0°C, the reason is that product internal water is not pure water, it is solution form.



06. Dynamics graph of the freezing process fillet tuna pieces built by experiment

The crystallizing temperature of the pure solvent is not been changing during the freezing process, but that of solution is being decreased continuously during the process (Banin A. and Duwayne A. M.., 1974; Haugvalstad G. H., Skipnes D.and Sivertsvik M.., 2005) to reach the Eutectic point where all amount of product internal water is crystallized. When the temperature of the product reduces to –1.24°C, some amount of water is crystallized, and the crystallized water will separate from the solution. It increases the solution concentration, and leads to a decrease in the crystallized temperature. Thus, when the temperature of products decreases to –22.5°C, all internal water in the products is crystallized (This point is called optimal freezing temperature or the Eutectic point). However, crystallized water is only free and physio-chemically bonded water, chemically bonded water is nearly not crystallized.

4. Conclusions

This research found the optimized freezing mode of the fillet bigeye tuna (in size of 300mm length, 150mm width, and 20mm thick): environment temperature was –42.5°C and the freezing time was 2.12h. When this mode was applied for freezing, the temperature of fillet tuna at the end of the freezing process reached –22.5°C and the yield of weight loss was 3.1%. It meaned that all internal water of the product was crystallized and the quality loss is negligible. This technology regime, thus, can be can be applied in industrial scale for the frozen fillet tuna manufacturing process and served for domestic trade and exporting purposes.

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