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Thermal properties of commercial hydrobionts' tissues in the freezing process

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Abstract: The paper describes changes in thermal properties in the process of freezing of marine raw materials. The study objects were the skin of giant octopus (*Octopus dofleini* L.), pallium of Pacific squid (*Todarodes pacificus* L.), milt of Pacific herring (*Clupea pallasii* L.), and muscle tissue of Japanese cucumaria (*Cucumaria japonica* L.). The mathematical relations of the studied thermal parameters allowing the calculation of specific heat capacity, thermal conductivity coefficient and tissue density of the studied objects in the process of freezing were obtained. It was found that the change in the total specific heat capacity during the freezing of all the objects under study was of the same type: first, this figure increases due to the intensive ice formation in the tissues of hydrobionts, and then decreases due to a significant decrease in the content of the liquid aqueous phase. The values of the total specific heat capacity before the freezing of seafood were determined (kJ/kg·K): 4.26 for squid, 3.58 for milt of Pacific herring, 3.66 for octopus skin, and 3.95 for the shell of cucumaria. It was revealed that an increase in the amount of frozen out water decreased the density of samples of frozen raw materials. This was due to the high (77.4–88.9%) content of water, turning into ice, which has a lower density index. The values of hydrobionts' tissue density before freezing were obtained (ρ_0, kg/m^3): 1226.74 for squid, 1209.6 for milt of Pacific herring, 1128.55 for octopus skin, and 1031.26 for shell of cucumaria. It was established that the thermal conductivity of the hydrobiont tissue samples in the process of freezing increased with the growth of the proportion of frozen out water contained, approaching the thermal conductivity of ice. The calculated values of thermal conductivity coefficient of seafood tissue prior to freezing equal (W/m·K): 0.52 for squid, 0.47 for milt of Pacific herring, 0.63 for octopus skin, and 0.53 for cucumaria. The obtained thermal characteristics values of the objects studied are recommended for use in technical and technological calculations of aquatic biological resources cooling treatment processes.

Keywords: Hydrobionts, waste, water content, freezing, ice formation, heat capacity, thermal conductivity, density, approximation

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INTRODUCTION

Although containing a number of nutrients in their composition, some parts of commercial hydrobionts are not widely used in food production, thus being wasted while processing. These include octopus skin, which makes up to 37% by weight of raw material and is rich in caratinoids, collagen, taurine, selenium, high-limit fatty acids [1–4]. Processing of Pacific herring produces rarely used now milt (up to 12.4% by weight of raw materials), which contains nucleoproteins, including biologically active substances (deoxyribonucleic acid and ribonucleic acid), and polyunsaturated fatty acids, including ω-3 and ω-6 families [5]. Among other insufficiently used raw materials, sources are the Pacific squid and Japanese cucumaria [6, 7]. However, these commercial objects provide sources of such biologically active substances as complete protein, hexosamines, chondroitin sulfate, triterpene glycosides, and polyunsaturated fatty acids [3, 8–10]. Getting with food in the human body, they slow down the aging process and have a corrective effect on metabolic processes, thus improving the quality of life and promoting longevity.

Cryotechnology is a promising trend in the industrial processing of biologically highly valuable raw materials. The method allows obtaining concentrates with highly preserved natural properties and biological activity [11–13]. Since the resulting cryopowders, as a rule, have the properties of biologically active additives, they are often used as biological correctors in the production of various food products and cosmetic materials, also being included in formulation compositions [14–18].

There are three main processes in cryogenic processing of raw materials of animal and plant origin: cryopreservation, cryogenic grindingб and freeze drying. Cryopreservation consists in rapid freezing of raw materials to a much lower than cryoscopic temperature, when most of the water turns into ice. It not only suppresses the activity of enzymes and the vital activity of microorganisms, but also creates favorable conditions for easier destruction of tissues during subsequent cryogenic grinding [11, 19]. By now, the process of freezing fish as a method of preservation has been widely studied, but there is lack of data on low-temperature processing of non-fish commercial objects. Also lacking are data on seafood thermal properties in the course of low-temperature processing. However, this knowledge is necessary when performing engineering calculations of processes and equipment related to cryogenic processing.

In this regard, the aim of the paper was to study the changes in thermal properties in the process of freezing raw materials of marine origin. Total specific heat capacity, thermal conductivity coefficient and density were calculated for the selected objects of study.

STUDY OBJECTS AND METHODS

The study objects were the skin of giant octopus (*Octopus L. dofleini L.*), pallium of Pacific squid (*Todarodes pacificus*), milt of Pacific herring (*Clupea pallasii L.*), and muscle tissue of Japanese cucumaria (*Cucumaria japonica L.*).

The amount of water in the samples, being the main factor of the freezing process, was determined by the standard method according to State Standard 7636-85 [20].

The standard software package of Microsoft Office 2007 and CurveExpert 1.4 were used for statistical data processing and graphs plotting with formula derivation.

Total specific heat capacity determination. The specific heat capacity of food products as multicomponent substances is calculated according to the law of additivity [21]:

$$c = g_1 c_1 + g_2 c_2 + g_3 c_3 + \ldots + g_n c_n$$

where $c_1, c_2, c_3, \ldots, c_n$ are specific heat capacities of components, kJ/kg·K;
$g_1, g_2, g_3, \ldots g_n$ are mass fractions of the components.

Consider the body of the study object as a two-component mixture containing W parts of water and (1–W) parts of dry substances with corresponding specific heat capacities for each component c_w and $c_{d.s}$. Heat capacity of the product in the temperature range before ice formation is determined by the expression:

$$c = c_w W + c_{d.s}(1 - W) \quad (1)$$

where c_w = 4.19 kJ/kg·K is water heat capacity (4.19 kJ/kg·K);
$c_{d.s}$ is specific heat capacity of dry substances in raw materials [22].

Since at negative temperatures part of the water ω in the object under study transforms into ice, whose heat capacity is c_i , the heat capacity of the frozen raw material c_{frm} is calculated by the formula:

$$c_{frm} = c_w W(1 - \omega) + c_i W \omega + c_{d.s}(1 - W) \quad (2)$$

where c_i is the heat capacity of ice (2.1 kJ/kg·K).

When freezing, the heat of ice formation will be removed from the mass unit at a lower temperature, which is defined a

$$dq_\omega = L_f W \frac{d\omega}{dt} \quad (3)$$

where L_f is the specific heat capacity of ice formation ($334.2 + 2.12t + 0.0042t^2$ kJ/kg);
W – total water content of the sample, kg/kg.
t – temperature of frozen raw materials, °C.

If temperature change of one degree is adopted in the expression (3), the amount of heat will receive the dimension and meaning of the component of the specific total heat capacity and be recorded as:

$$q_\omega = L_f W(\omega_2 - \omega_1) \quad (4)$$

where ω_1 is the amount of frozen out water at the initial temperature;
and ω_2 is the amount of frozen out water at the final temperature.

The sum of calculated heat capacity of the frozen raw material c_{frm} and the heat of ice formation q_ω will give the total specific heat capacity:

$$c_{tot} = c_{frm} + q_\omega \quad (5)$$

Thermal conductivity coefficient determination. When the temperature drops below the cryoscopic value and the product is in the process of ice formation, its thermal conductivity increases significantly, since thermal conductivity of ice is four times greater than that of water.

The increase in thermal conductivity of the product with decrease in temperature almost ceases with the end of water freezing out, granted that further insignificant change in the thermal conductivity of ice and other components of the product is neglected. The thermal conductivity coefficient of products in the range of negative temperatures λ_{fr} depends on the amount of frozen out water and approximates to the equation [23]:

$$\lambda_{fr} = \lambda_0 + \omega \Delta\lambda \quad (6)$$

where λ_0 is the coefficient of thermal conductivity of the product before freezing, W/m·°C;

$\Delta\lambda$ is the change in thermal conductivity of the product in the temperature range from the start of freezing t_s to t_c corresponding to completion of ice formation.

Considering raw materials as a two-component mixture containing parts of water W and (1–W) parts of dry substances with respective thermal conductivity coefficients of λ_w and $\lambda_{d.s}$, the heat capacity of the product in the temperature range before ice formation is determined by the expression:

$$\lambda_m = \lambda_w W + \lambda_{d.s}(1 - W)$$

where λ_w = 0,597 W/m²·K is the coefficient of water thermal conductivity;

$\lambda_{d.s}$ = thermal conductivity coefficient of dry substances [6].

The coefficient of thermal conductivity can be calculated by the formula based on the models of Krisher [5]:

$$\lambda_f = \frac{1}{\lambda_i - \varepsilon_p(\lambda_i - \lambda_{fr})}\left[\lambda_{fr}\lambda_i + \frac{(\lambda_i - \lambda_{fr})^2}{2}\left(\varepsilon_p - \varepsilon_p^2\right)\right] \quad (7)$$

where λ_i is thermal conductivity of ice coefficient within the temperature range 273–208 K (2,22 W/m·K);

ε_p – porosity coefficient which depends on the amount of frozen out water and chemical composition.

The structure of the frozen product can be considered as a dispersed system consisting of ice pores with coefficient of thermal conductivity λ_i and a matter containing unfrozen water and dry substances with a coefficient of thermal conductivity approximately equal to λ_0 before freezing.

Porosity coefficient of the assumed structure will be determined by the expression:

$$\varepsilon_p = \frac{W\omega}{m\rho_i + W\left[\frac{\rho_i}{\rho_w} + \omega\left(1 - \frac{\rho_i}{\rho_w}\right)\right]} \quad (8)$$

where ρ_i is ice density, kg/m³;

ρ_w is product density before freezing, kg/m³;

m is mass fraction of dry substances in raw materials.

Taking into consideration stable weight fraction of dry substances in the process of freezing, and practically unvarying density ρ_m

$$m = \frac{1}{\rho_m} - \frac{W}{\rho_w} \quad (9)$$

Frozen raw material density determination. Consider the body of the object under study as a three-component mixture consisting of unfrozen water, ice, and dry matter. Density of the samples can thus be presented as the equation [6]:

$$\rho_{frm} = \frac{1}{\frac{g_1(1-\omega)}{\rho_1} + \frac{g_2}{\rho_2} + \frac{g_1\omega}{\rho_3}} \quad (10)$$

where g_1 is the mass fraction of water contained in the sample body;

g_2 is the mass fraction of solids contained in the sample body;

ρ_1 is water density (1000 kg/m³);

ρ_2 is dry matter density of raw materials, kg/m³ [21];

ρ_3 is ice density (917 kg/m³);

ω is the amount of frozen out water.

Table 1 Water content in the tissues of hydrobionts

Sample	Water content,%
Milt of Pacific herring	77.4
Pallium of Pacific squid	78.6
Skin of octopus	84.8
Japanese cucumaria	88.9

RESULTS AND DISCUSSION

Data on water content determination in the tissues of the studied hydrobionts are given in Table 1.

The objects under study have a high water content ranging from 77.4% (in the milt of Pacific herring) to 88.9% (in the muscle tissue of the Japanese cucumaria), which corresponds to the known data [2, 3, 7, 24].

Using formula (5), we calculate the total specific heat capacity of the samples. To do this, it is necessary to determine the amount of frozen out water at different temperatures using Ryutov's formula [25]. Then we apply formulae (2) and (4) to determine the heat capacity for the selected raw material and the heat of ice formation. The resulting values of the total specific heat capacity of the raw material are depicted as graphs in Fig. 1.

Presented in Fig. 1 graphs show the relation between total specific heat capacity and the amount of frozen out water for the four studied objects. As can be seen, they are of the same type and have two distinct areas. The first one demonstrates an increase in the total specific heat capacity of seafood samples, which is associated with intensive ice formation in their tissues with a decrease in temperature and accompanying heat release. The second area is characterised by a gradual decrease in the total specific heat capacity of seafood samples. This is associated with a significant decrease in the amount of liquid aqueous phase and, accordingly, a decrease in the intensity of its transition to the crystalline form with the release of heat caused by ice formation. At the final stage, when most water is frozen out, the total specific heat capacity of the samples under study tends to the heat capacity of ice becoming one of the main factors of the further freezing process. The transition point of the total specific heat capacity from increase to decrease is reached when the amount of frozen out water gets close to 50%. The obtained values of total specific heat capacity of commercial hydrobionts' tissues are consistent with the data available in the academic literature on aquatic raw materials [25].

Approximating the curves shown in Fig. 1 with Curve Expert Professional 2.3, we get the formulae:

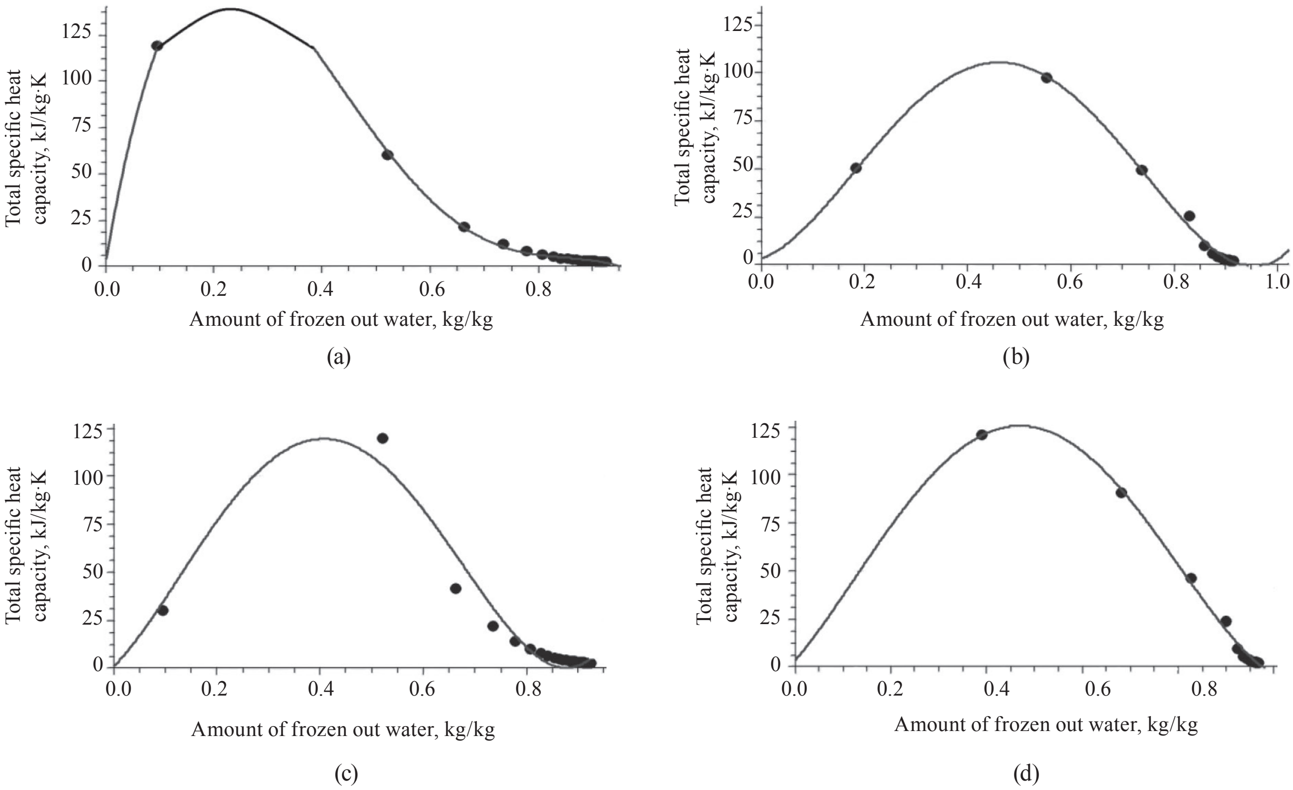

Figure 1 Relation between total specific heat capacity and the amount of frozen out water: (a) pallium of Pacific squid; (b) milt of Pacific herring; (c) octopus skin; (d) Japanese cucumaria

$$c_s = 2751.19\omega^4 - 4888.57\omega^3 + 2159.33\omega^2 + 9.05\omega + 4.26 \quad (11)$$

$$c_m = 1786.77\omega^4 - 3293.67\omega^3 + 1410.95\omega^2 + 95.48\omega + 3.58 \quad (12)$$

$$c_{os} = 2511.06\omega^4 - 4238.40\omega^3 + 1611.53\omega^2 + 149.47\omega + 3.66 \quad (13)$$

$$c_{cu} = 1140.6\omega^4 - 2110.32\omega^3 + 669.94\omega^2 + 291.58\omega + 3.95 \quad (14)$$

These formulae can be used to calculate the relation between total specific heat capacity and the amount of frozen out water for the studied raw materials with a correlation coefficient of 0.99. The free term in the obtained formulae determines the value of total heat capacity of the raw material with the amount of frozen water equal to 0. Therefore, total specific heat capacity of non-frozen seafood equals (kJ/kg·K): 4.26 for squid, 3.58 for milt of Pacific herring, 3.66 for octopus skin, and 3.95 for cucumaria shell. The values of heat capacity of non-frozen raw materials calculated, based on the standard formula (1) were as follows (kJ/kg·K): 4.06 for squid; 3.52 for milt; 4.05 for octopus skin; and 3.93 for cucumaria. The difference between the data obtained according to formulae (11–14) and (1) is 4.9, 1.7, 9.6, and 0.5% for squid, milt, octopus skin, and cucumaria, respectively. This indicates the adequacy of the derived mathematical relationships.

Using formula (7), we calculated the coefficient of thermal conductivity of the selected raw material and plotted the relation to the amount of frozen out water (Fig. 2).

Analysing the graphs in Fig. 2, we see that the dependence of the change in the thermal conductivity of the studied samples is close to linear. The thermal conductivity of the studied seafood in the process of freezing increases with the proportion of frozen out water, tending to the thermal conductivity of ice, which is almost four times greater than the thermal conductivity of water. Approximating the chart data using Curve Expert Professional 2.3, we obtain the formulae:

$$\text{for squid: } \lambda_s = 0.52 + 1.02\omega \quad (15)$$

$$\text{for milt of herring: } \lambda_m = 0.47 + 1.01\omega \quad (16)$$

$$\text{for octopus skin: } \lambda_{os} = 0.63 + 1.07\omega \quad (17)$$

$$\text{for cucumaria: } \lambda_{cu} = 0.53 + 1.54\omega \quad (18)$$

Formulae (15–18) can be used to calculate the thermal conductivity of the studied objects with a correlation coefficient of 0.99. They also allow us to determine the thermal conductivity of the test

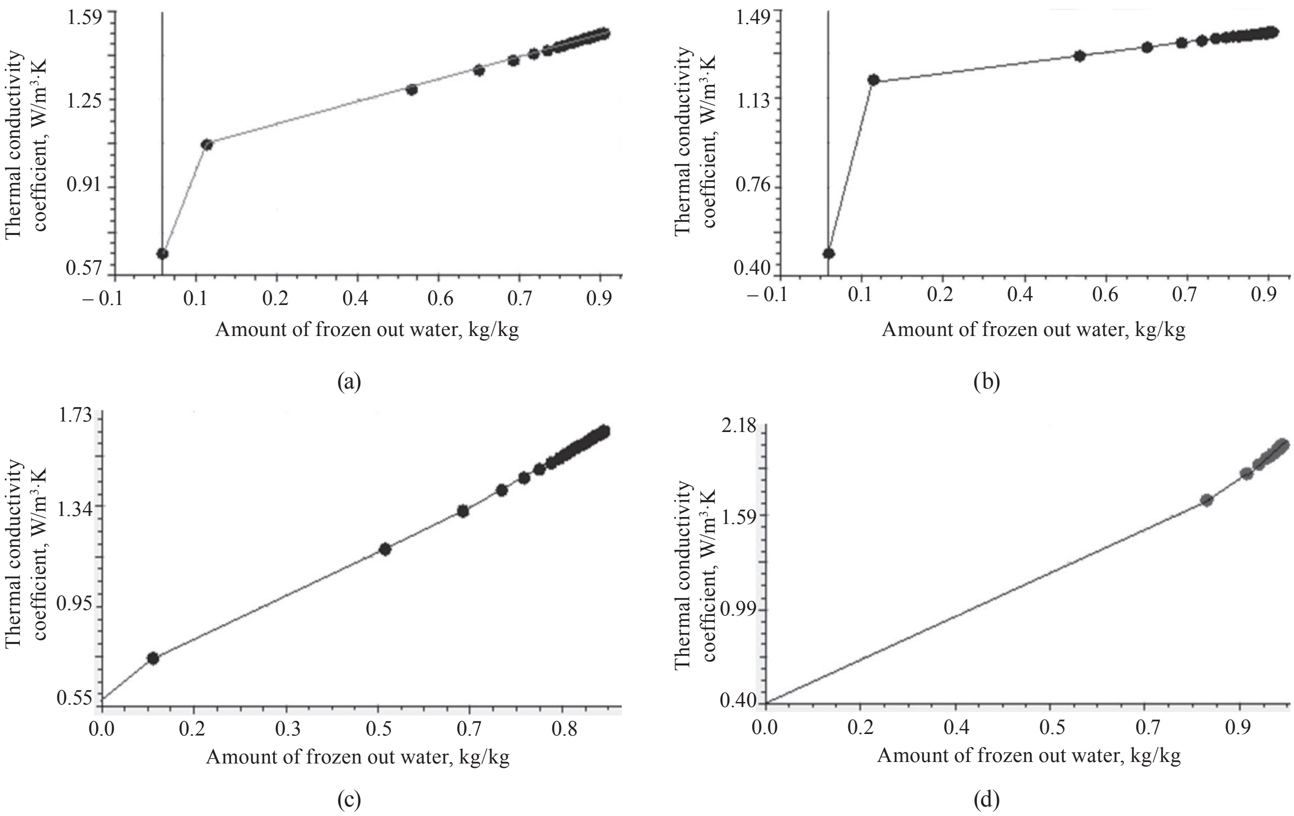

Figure 2 Relation between thermal conductivity coefficient and the amount of frozen out water for: (a) squid trunk; (b) milt of herring; (c) octopus skin; (d) cucumaria

samples before freezing, when the amount of frozen out water $\omega = 0$. The thermal conductivity coefficient of non-frozen seafood equals: squid – 0.52 W/m·K, milt of Pacific herring – 0.47 W/m·K, octopus skin – 0.63 W/m·K, cucumaria – 0.53 W/m·K. The values of thermal conductivity coefficients obtained correlate well with the data available in academic literature for fish raw materials: big-eyed tuna, Pacific cod, tilapia [26–28].

Formulae (15–18) correspond to the equation (6), which allows to conclude that for the studied samples $\Delta\lambda$ equals the following values, W/(m·K): squid – 1.02; milt of herring – 1.01; octopus skin – 1.07; cucumaria – 1.54. It is known that the value of $\Delta\lambda$ according to experimental data for food containing 70–80% of water varies within 0.928–1.16 W/m·K [23]. This range exceeds $\Delta\lambda$ of cucumaria, which can be explained by the peculiar structure and higher water content (88.9%) in its muscle tissue.

Formula (10) helps calculate the density of raw materials in the process of freezing and construct graphs of the relation between density and the amount of frozen out water (Fig. 3).

Analysing the graphs in Fig. 3 it should be noted that the considered relations are of the same type and close to linear. Density of frozen raw materials is reduced with the increase in the amount of frozen water. This happens due to the high water content in the studied objects. Water turns into ice which has a lower density index. Approximating data curves with the help of Curve Expert Professional 2.3, we get the formulae:

$$\rho_{f.m} = 1209.6 - 142.89\omega \quad (19)$$

$$\rho_{f.s} = 1226.74 - 149.08\omega \quad (20)$$

$$\rho_{f.os} = 1128.55 - 138.24\omega \quad (21)$$

$$\rho_{f.cu} = 1031.26 - 100.42\omega \quad (22)$$

These equations can be used to determine the density of the samples before freezing, with the amount of frozen water equals 0. Then the density of chilled milt of Pacific herring can be set to ρ_0 = 1209.60 kg/m³, ρ_0 squid = 1226.74 kg/m³, ρ_0 octopus skin = 1128.55 kg/m³, and ρ_0 cucumaria shell = 1031.26 kg/m³. These data correlate well with the calculated values of the density of unfrozen objects under study obtained by formula (10).

The derived formulae (19–22) can be used to calculate the relation between the density of herring milk of the Pacific, squid trunk, octopus skin, cucumaria shell and the amount of frozen out water with a correlation coefficient of 0.99. The results of calculations show that the decrease in the density of the studied hydrobionts' tissues during freezing, when the amount of frozen out water reaches, for example, 90% makes up for squid – 11.9%, milt – 9.0%, octopus – 11.0%, and cucumaria – 8.4%. It is known that during freezing the

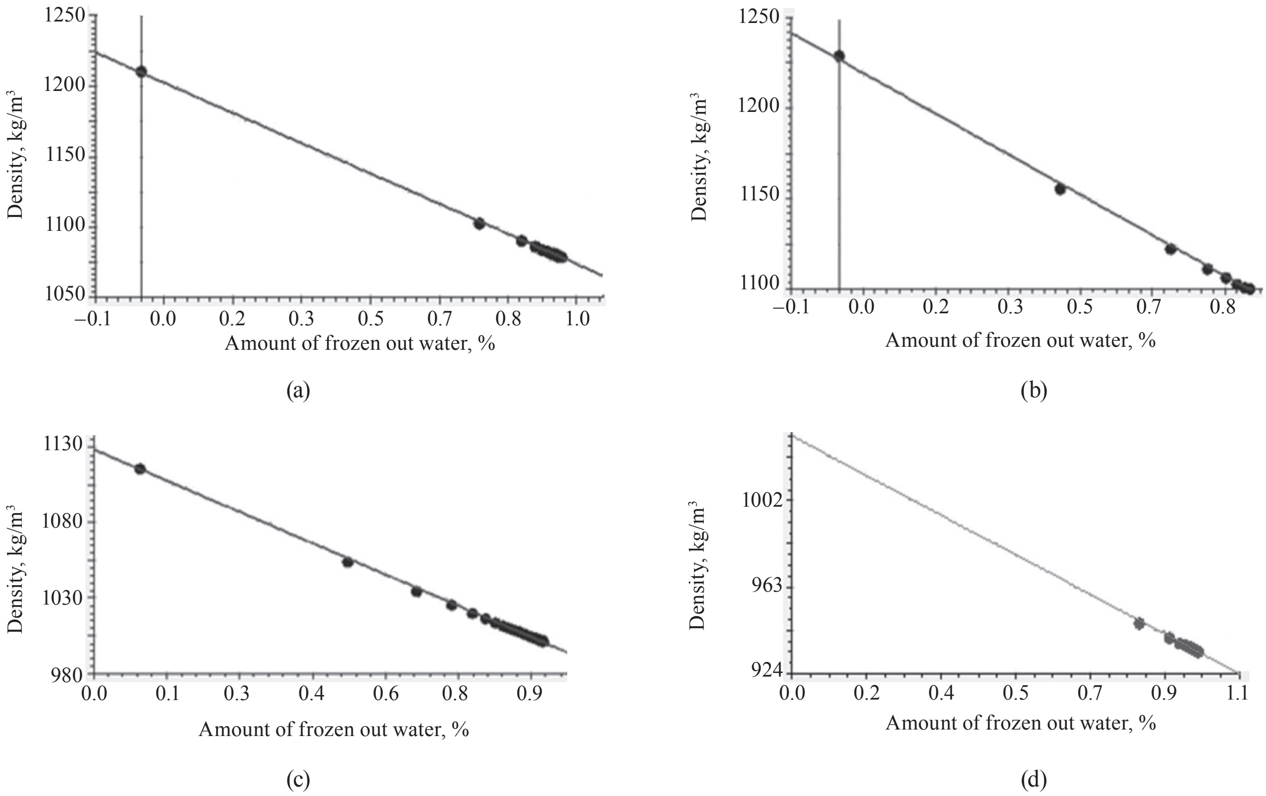

Figure 3 Relation between the density and the amount of frozen out water for: (a) milt of Pacific herring; (b) pallium of squid; (c) octopus skin; (d) cucumaria shell

density of Atlantic mackerel muscle tissue decreases by 9.3% [23].

Thus, studies of changes in thermal properties in the process of freezing Pacific squid, milt of Pacific herring, giant octopus, and muscle tissue of Japanese cucumaria were undertaken.

CONCLUSION

It was found that during freezing the change in total specific heat capacity of all the objects under study is of the same type: first, this figure increases due to the intensive ice formation in the tissues of hydrobionts, and then decreases due to a significant decrease in the content of the liquid aqueous phase in the objects under study. The transition point from growth to fall corresponds to the values of the amount of frozen water close to 50%.

The relation between the coefficient of thermal conductivity of the studied hydrobionts' tissues and the amount of frozen out water is close to linear. The thermal conductivity of tissue samples slowly increases with the proportion of frozen out water, approaching the thermal conductivity of ice.

The relation between the density index of hydrobionts' tissues in the freezing process is also close to linear. With the increase in the amount of frozen out water the density of the frozen raw material decreases, since the samples under study have a high content of water turning into the ice, which has a lower density index.

The obtained mathematical relationships of the studied thermophysical parameters also allow us to obtain the values of specific heat capacity, thermal conductivity and tissue density of fresh and chilled hydrobionts prior to freezing, when the amount of frozen out water equals zero.

The obtained digital values of total specific heat capacity, thermal conductivity, and density can be used by specialists for calculation, modeling and design of basic and derivative processes of non-fish commercial hydrobionts low-temperature processing, as well as refrigeration and process equipment.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest related to this article.

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