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ORIGINAL ARTICLE

Mathematical model and dependence of fish salting process duration to a given salt concentration

Modelo matemático para determinar a duração do processo de salgagem e a concentração de sal no peixe

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Abstract

The purpose of this scientific work is to build a diffusion model based on Fick's second law. The model is designed to determine the salting process duration and salt concentration in the fish muscle tissue (Atlantic herring). The method of photon correlation spectroscopy (PCS) by a certified Photocor FC correlator device was used. The device operating principle is based on Rayleigh radiation scattering, which allows us to directly determine the effective diffusion coefficients, taking into account all factors, without singling out a specific one. This research presents the spatiotemporal distributions of effective diffusion coefficients in a herring sample during brine salting. Formulas for calculating the salting duration and the average salt concentration in the thickness of fish muscle tissue are presented, and the time of reaching the sample center by salt and the average salt concentration in it were estimated. These parameters could be widely used for the finished product quality and the technical and economic indicators of production improvement. The average effective diffusion coefficient D_e through the sample thickness was used for calculation. The salting duration for a 30 mm thick herring sample with a saturated salt solution was 8.74 hours until the average salinity of the sample was 4.7%.

Keywords: Salting process mathematical model; Effective diffusion coefficient; Salting duration; Fish salting; Boundary layer; Spatiotemporal salt distribution; Salt concentration determination.

Resumo

O objetivo deste trabalho científico é construir um modelo de difusão baseado na segunda lei de Fick. O modelo é projetado para determinar a duração do processo de salga e a concentração de sal no tecido muscular do peixe, o arenque-do-atlântico. O método de espectroscopia de correlação de fótons foi utilizado nesta pesquisa. Para estudar amostras de peixes, foi usado um dispositivo correlacionador Photocor FC certificado. O princípio de operação do dispositivo é baseado na dispersão de radiação Rayleigh, que permite determinar diretamente os coeficientes de difusão efetivos, levando em consideração todos os fatores, sem destacar um específico. Esta pesquisa apresenta as distribuições espaço-temporais dos coeficientes de difusão efetivos em uma amostra de arenque durante a salga em salmoura. São apresentadas fórmulas para cálculo da duração da salga e da concentração média de sal na espessura do tecido muscular do peixe, e foram estimados o tempo de chegada do sal ao centro amostral e a concentração média de sal no mesmo. Esses parâmetros poderiam ser amplamente utilizados para a qualidade do produto acabado e os indicadores técnicos e econômicos de melhoria da produção. O coeficiente de difusão efetivo médio D_e através da espessura da amostra foi utilizado para o cálculo. A duração da salga para uma amostra de arenque de 30 mm de espessura com solução salina saturada foi de 8,74 horas até que a salinidade média da amostra fosse de 4,7%.

Palavras-chave: Modelo matemático do processo de salga; Coeficiente de difusão efetivo; Duração da salga; Salga de peixe; Camada limite; Distribuição espaçotemporal de sal; Determinação da concentração de sal.

Highlights

- A mathematical model of salting by the photon correlation spectroscopy (PCS) method was developed
- The spatiotemporal distribution of NaCl in the case of fish brine salting was investigated
- The mathematical model allows to predict the salt content in the muscle tissue of fish

1 Introduction

Among the wide variety of harvested hydrobionts, one of the important fishery objects is herring whose reserves are sufficient (Food and Agriculture Organization, 2020). The main method of herring preparation is salting which has been known since ancient times as the preserving process (Nummer & Brian, 2002; Holm et al., 2022). Currently, fish salting is still a relevant technological process. It is used to obtain either finished products with a specific aroma and taste, or semi-finished products, from which dried, smoked, cured, pickled, or other types of products are subsequently made (Turan & Erkoyuncu, 2012).

As an object of processing, herring is a traditional type of fish for salting, since it ripens during this process (Rahaman, 2014). Taking into account the positive dynamics of herring catch and its significant commercial stock, it seems relevant to conduct further research on herring salting, including mathematical modeling of the process, as well as using modern experimental methods.

Salting is a complex mass transfer process consisting of diffusive transfer of salt into fish and diffusion-osmotic transfer of the liquid phase from fish tissues to brine. During the salting process, physicochemical and biochemical changes occur. Salt diffusion is associated with changes in the salt concentration inside the product and in the brine, where the mass transfer rate does not stop until equilibrium occurs in the «fish-brine» system (Gómez-Salazar et al., 2015).

There are dry, brine and mixed types of salting. Dry salting consists of rubbing or coating the raw materials with solid salt. The brine salting is processing fish in a salt solution of a certain density. Barat et al. (2003) did not find any significant differences between dry and brine salting processes in terms of the physicochemical and sensory parameters but noted that dry salting is more favorable to dehydration, while brine salting favors the entry of salt. However, brine salting has several advantages over dry salting, including shorter processing times due to higher salt uptake and higher weight yields due to better control over the rate of salt uptake and water loss in the muscle (Andrés et al., 2005; Beraquet et al., 1983; Thorarinsdottir et al., 2004).

The most recognized empirical model of fish salting, experimentally confirmed for various fish species, is the model of Zugarramurdi and Lupín. This model is an exponential dependence created for salt absorption and moisture loss of products during mass transfer until the equilibrium state of the system is reached (Zugarramurdi & Lupín, 1980).

The duration of salting is of great scientific and practical importance in fish processing. Previously, methods for determining the duration of salting have been developed (Borgstrom, 1965; Dimova et al., 2006; Levanidov et al., 1987). The patterns obtained by these researchers require knowledge of diffusion coefficients. There is no consensus on how the diffusion coefficient changes during the salting process. Moreover, there is no single approach to determining its values. In most works (Zaitsev et al., 1969; Wheaton, Lawson, 1985; Levanidov et al., 1987), the diffusion coefficient is replaced by the salting coefficient. The two indexes are similar, but not identical.

The determination of the diffusion coefficient and the study of its changes during salting are of practical interest to clarify the quantitative dependencies characterizing the salting process and complementing the salting theory.

It is known that the diffusion coefficient in the salting process depends on several factors, such as water content, brine concentration, operating temperature, muscle tissue fiber direction, shrinkage and pH. For this reason, it is necessary to determine the effective diffusion coefficient $D_{e,}$ which covers all real factors (Gómez-Salazar et al., 2015). D_e characterizes the rate of transfer of the diffusing substance through a unit of the effective diffusion surface. Unlike the D diffusion coefficients, which can be obtained by calculation, the D_e effective diffusion coefficients are determined experimentally, as a rule, by one of the methods of continuous non-destructive testing.

To obtain information about the spatiotemporal nature of salt distribution within the product, existing chemical methods cannot be used since they do not provide accurate measurements of the salt and moisture content at each point of the product during processing (Barat et al., 2011).

In this direction, it is necessary to use non-destructive testing methods, such as X-ray tomography (Vestergaard et al., 2004), ultrasound (Fortin et al., 2003) and Nuclear Magnetic Resonance (NMR) (Bertram et. al., 2004; Liang et al., 2020).

To obtain quantitative patterns characterizing the diffusion of salts, it is necessary to propose a mathematical model and determine the values of effective diffusion coefficients by non-destructive testing - photon correlation spectroscopy (PCS).

The purpose of this research was to build a mathematical model that makes it possible to determine the optimal duration of the fish salting process to a given concentration, which in the future will allow to manufacture of products with specified consumer properties at minimal production costs.

2 Materials and methods

2.1 Materials

To study the effect of sodium chloride on the salting process, Atlantic herring (*Clupea harengus*), frozen for about 3 months at -18 °C, caught in summer, 30 ± 2 cm long and weighing 310 ± 10 g, was used. The laboratory-determined chemical composition of Atlantic herring tissues was: fat 18 ± 0.5%, protein 18 ± 0.5%, minerals 1.5 ± 0.1%.

Sodium chloride and potable water were used to prepare a brine of a given concentration of 25.9% (saturated salt solution).

2.2 Research methods

In this research, a modern optical method of PCS was used. It is based on Rayleigh scattering of radiation from substances (Pike & Abbiss, 1997). The PCS method consists in measuring the diffusion coefficient of dispersed particles by analyzing dynamic fluctuations in the intensity of scattered light.

The Photocor-FC correlator is a certified experimental device with Flex 5.3.3 software (Photocor Ltd.) working according to this method. The Photocor-FC is a multifunctional real-time device for auto- and cross-correlation measurements built on software configurable flex-logic integrated circuits. The capabilities of this device make it possible to measure diffusion coefficients both in solutions and in the muscle tissue of fish. The maximum value of the relative error of the diffusion coefficient is 5%.

The device radiation source is a single-mode helium-neon laser (power W = 15 mW, wavelength λ = 632.8 nm, beam diameter = 100 μm).

The scattered light was recorded by a photomultiplier tube (PMT) operating in the photon counting mode. The correlation function was calculated using a 32-bit 282-channel Photocor-FC correlator connected to a computer.

The PCS method is based on the fact that information about the particle diffusion coefficient is contained in a time-dependent correlation function of intensity fluctuations. The temporal autocorrelation function is described as follows Equation 1:

$$G(\tau) = I(\tau - \tau') = \lim_{\tau_m \to \infty} \frac{1}{\tau_m} \int_0^{\tau_m} I(\tau)\, I(\tau - \tau') d\tau \tag{1}$$

The autocorrelation function of intensity decays exponentially with time, and the relaxation time is related to the diffusion coefficient by the Equation 2:

$$G(\tau) = a \cdot exp\left(\frac{-2\tau}{\tau_s}\right) + b \tag{2}$$

where the regression correlation time according to the solution of the diffusion equation is as follows Equation 3:

$$\frac{1}{\tau_s} = Dq^2 \tag{3}$$

The wave vector of concentration fluctuations is described by the following Equation 4:

$$q = \frac{4\pi n}{\lambda} sin\, sin\, \frac{\vartheta}{2} \tag{4}$$

a and b are experimental constants;

n is the substance refractive index;

λ is the wavelength of the laser radiation;

ϑ is the scattering angle.

The program calculated the scattering correlation function (time resolution τ’ = 25 ns), then the particle size distribution function and the diffusion coefficient were determined from the correlation function.

The device scheme is shown in Figure 1.

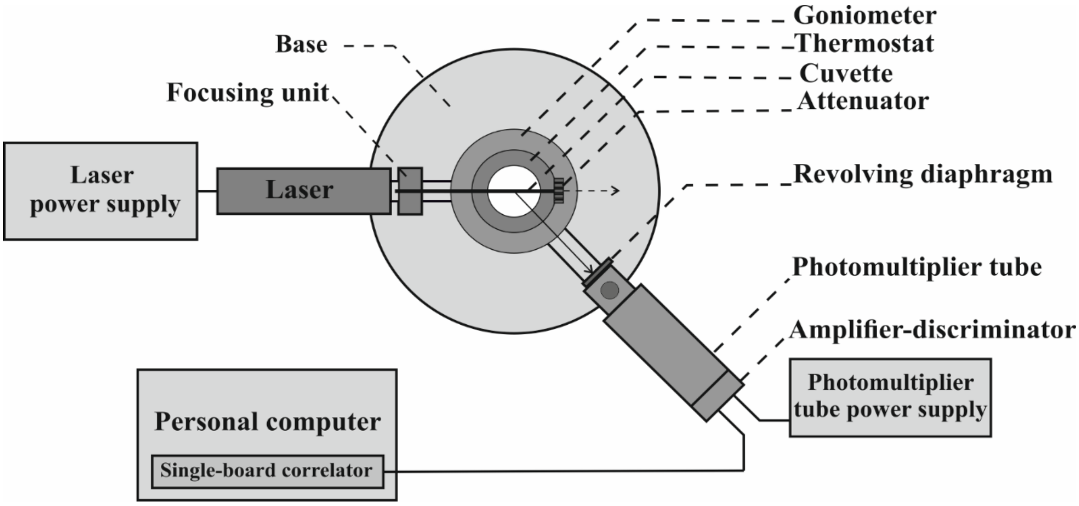

Figure 1. The device scheme.

2.3 Research methodology

Five herring samples were examined. The samples were cut with skin from the middle part of the herring back. The height of the samples was 15 mm. The surface area of the slice (28 x 16 mm) corresponded to the cross-sectional area of a quartz cuvette in the form of a parallelepiped with dimensions of 28 x 16 x 36 mm. Cuvette had the shape of a parallelepiped. The scheme of filling the cuvette is shown in Figure 2. The filling scheme of the radiated object was chosen in such a way that it was possible to produce a mathematical formulation of the studied salting processes.

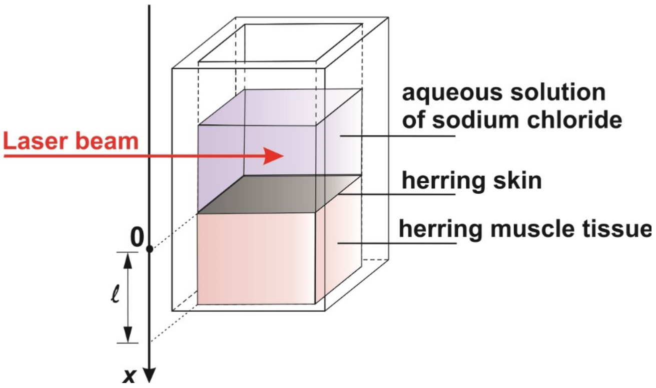

Figure 2. The scheme of filling the cuvette.

The samples were placed in the cuvette with the muscle tissue facing down and the skin up. A saturated salt solution was poured onto the skin of the fish. The volume of the sodium chloride solution corresponded to the size of the fish sample. The experiment was replicated five times.

The cuvette was placed in a thermostat at a temperature of 10 ± 0.1 °C. The x-axis was directed downwards along the height of the cuvette. The point on the fish skin was taken as zero. The positive values of the x-axis were going from the skin deep into the muscle tissue to the value of l (l is the thickness of the fish muscle tissue).

The laser beam fell on the side face of the quartz cuvette, which moved along its height with a path length of 1 mm using a specially installed micrometer device. Thus, the laser beam scanned the skin and muscle tissue of herring along the height of the cuvette.

Effective diffusion coefficients for the initial process time were measured through the entire thickness of the sample after every 1 mm ($\tau = 0$ min), then measurements were made every 30 min ($\tau = 30; 60; 90; 120; 150; 180; 210; 240$ min).

The coefficients of effective diffusion through the thickness at certain time points (0-240 min) were measured on five samples for the reliability of the results.

3 Results and discussion

3.1 Solution of the differential diffusion equation

All mathematical models describing the regularities of mass transfer during the salting process can be divided into empirical and theoretical. Most theoretical models are diffusion models based on Fick's second law by the Equation 5:

$$\frac{\partial c}{\partial \tau} = D \frac{\partial^2 c}{\partial x^2}. \tag{5}$$

Defining the sample in the form of a plate of finite dimensions ($0 < x < l$), initial and boundary conditions have been formulated by the Equation 6:

$$\left.\begin{aligned} C(x,0) &= C_{nat} \\ C(0,0) = C(l,0) &= C_0 \end{aligned}\right\}, \tag{6}$$

where C_{nat} is the natural salinity of fish muscle tissue, %; C(0,0), C(l,0) is the salt concentration at sample opposite sides at the initial time, %; C_0 is the solution concentration in the boundary layer at the edge of the sample, %.

The solution of the differential diffusion equation with boundary conditions was obtained by the method of separation of variables (also known as the Fourier method).

The solution of the diffusion equation was expressed as a series that slowly converges if $D\tau < l^2$ (where l is the thickness of the slice (m); D is the diffusion coefficient (m^2/s); τ is the time, (s); and x is the coordinate) by the Equation 7:

$$C(x,\tau) = C_0\left(1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{1}{2n+1}\cdot e^{\frac{-(2n+1)^2\pi^2 D\tau}{l^2}}\cdot \sin\frac{(2n+1)\pi x}{l}\right) \tag{7}$$

With $\tau \geq \tau' = 0.05\, l^2/D$, it is possible to limit ourselves to the first member of the series (n = 0) with a 1% accuracy by the Equation 8:

$$C(x,\tau) = C_0\left(1 - \frac{4}{\pi}e^{\frac{-\pi^2 D\tau}{l^2}}\cdot \sin\frac{\pi x}{l}\right) \tag{8}$$

D is the diffusion coefficient, (m^2/s); C_0 is the concentration of salt solution in the boundary layer near the fish skin (%).

The time for the salt to reach the center of the sample was found by the Equation 8. At $C(x;\tau) = C(l/2;\tau) = 0$ the following result is obtained by the Equation 9:

$$\tau' = \frac{4\ln\frac{\pi}{4}}{-\pi^2}\cdot\frac{l^2}{D} = 0.0979\frac{l^2}{D}. \tag{9}$$

The solution of Equation 8 will be as follows Equation 10:

$$C = C_0\left(1 - \frac{4}{\pi}\cdot e^{\frac{-2.4674 D\tau}{l^2}}\right) \text{ or } C = C_0\left(1 - 0.9998\cdot e^{-2.4674\left(\frac{D\tau}{l^2}-0.0979\right)}\right) \tag{10}$$

The coefficient before the exponent can be equated to 1.

Scientists Dimova et al. (2006) and Glazunov (1983) used a variational method for solving the differential diffusion equation and developed the following solution by the Equation 11 taking into account the natural salinity of fish:

$$C = C_{nat} + C_0\left(1 - e^{-2.4706\left(\frac{D\tau}{l^2}-0.0843\right)}\right) \tag{11}$$

Solutions of diffusion equations by different methods with Equations 10 and 11 are very similar and differ slightly in numerical coefficients. It is worth considering that the numerical coefficients in Equation 10 were obtained by the analytical method.

As a result, by separating the variables, a formula was obtained for calculating the salting time to the center of a fish sample with skin to a given concentration. This Equation 12 takes into account the natural salinity of the fish C_{nat}:

$$\tau = \frac{l^2}{D}\left(\frac{1}{2.4674}\ln\frac{C_0}{C_0 - C - C_{nat}} + 0.0979\right) \tag{12}$$

where D is the diffusion coefficient determined using the Photocor-FC correlator by photon correlation spectroscopy, m^2/s;

C is the concentration of salt in the center of the sample, %;

C_0 is the concentration of salt solution in an unlimited layer near fish skin, %;

$C_0 = \alpha C_b$;

C_b is the concentration of the salt solution away from the skin of the fish, %;

C_{nat} is the natural salinity of fish muscle tissue, %. (C_{nat} = 0.2 - 0.4%);

α is a correction factor indicating a decrease in the salt concentration in the boundary layer of the solution near the fish skin.

For herring, α = 0.67 (Dimova et al., 2006).

In Equation 12, the salt concentration near the skin of the fish C_0 (%) differs from the concentration of the solution away from the fish skin C_b (%). The correlation was established experimentally through the correction factor α: $C_0 = \alpha\, C_b$ (Dimova et al., 2006).

If the distribution of salt concentration through the thickness of the sample is known, it becomes possible to calculate the average concentration of salt in the muscle tissue of fish by the Equation 13:

$$< C(x,\tau) > \frac{2}{l}\int_0^{l/2} C(x,\tau)dx = \frac{2}{l}\int_0^{l/2} C_0\left(1-\frac{4}{\pi}\cdot e^{\frac{-\pi^2 D\tau}{l^2}}\cdot \sin\frac{\pi x}{l}dx\right) = C_0\left(1-\frac{8}{\pi^2}\cdot e^{\frac{-\pi^2 D\tau}{l^2}}\right) \quad (13)$$

Equation 12 is valid for the salting time τ > τ', where τ'= 0.0979 l^2/D is the time after which the salinity change begins in the center of the sample.

3.2 Dependences of the effective diffusion coefficient

The values of effective diffusion coefficients D_e through the thickness of herring muscle tissue at time points from 0 to 240 min at a temperature of 10 °C were experimentally determined by the PCS method. The values of D_e are shown in Table 1 and Table 2.

Table 1. Experimental effective diffusion coefficients depend on time and distance.

	$D_e \cdot 10^9$ (m²/s)							
τ (min)	**X (mm)**							
	0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
0	2.06 ± 0.09	1.61 ± 0.07	1.12 ± 0.06	0.91 ± 0.03	0.73 ± 0.02	0.69 ± 0.02	0.65 ± 0.03	0.61 ± 0.01
30	2.21 ± 0.11	1.89 ± 0.07	1.46 ± 0.03	1.07 ± 0.06	0.75 ± 0.05	0.38 ± 0.01	0.38 ± 0.03	0.37 ± 0.01
60	2.39 ± 0.12	2.05 ± 0.09	1.59 ± 0.02	1.07 ± 0.04	0.71 ± 0.04	0.47 ± 0.02	0.60 ± 0.04	0.55 ± 0.02
90	2.52 ± 0.11	1.87 ± 0.06	1.15 ± 0.06	0.69 ± 0.02	0.50 ± 0.03	0.51 ± 0.01	0,53 ± 0.04	0.49 ± 0.03
120	2.64 ± 0.13	2.15 ± 0.09	1.71 ± 0.04	1.32 ± 0.04	1.03 ± 0.06	0.82 ± 0.03	0.84 ± 0.03	0.89 ± 0.05
150	2.72 ± 0.12	2.11 ± 0.09	1.33 ± 0.04	0.89 ± 0.03	0.69 ± 0.04	0.63 ± 0.04	0.55 ± 0.03	0.51 ± 0.05
180	2.81 ± 0.14	1.95 ± 0.10	1.23 ± 0.05	0.81 ± 0.05	0.71 ± 0.04	0.63 ± 0.02	0.59 ± 0.03	0.57 ± 0.01
210	2.87 ± 0.11	2.16 ± 0.08	1.55 ± 0.04	0.89 ± 0.01	0.73 ± 0.05	0.65 ± 0.01	0.60 ± 0.01	0.50 ± 0.03
240	3.06 ± 0.09	2.22 ± 0.07	1.34 ± 0.06	0.82 ± 0.03	0.71 ± 0.01	0.69 ± 0.04	0.67 ± 0.04	0.65 ± 0.02

Table 2. Experimental effective diffusion coefficients depend on time and distance.

	$D_e \cdot 10^9$ (m²/s)							
τ (min)	**X (mm)**							
	8	**9**	**10**	**11**	**12**	**13**	**14**	**15**
0	0.59 ± 0.04	0.55 ± 0.01	0.48 ± 0.03	0.47 ± 0.02	0.44 ± 0.02	0.41 ± 0.01	0.39 ± 0.02	0.38 ± 0.04
30	0.32 ± 0.02	0.30 ± 0.02	0.27 ± 0.02	0.27 ± 0.01	0.27 ± 0.03	0.26 ± 0.02	0.26 ± 0.01	0.25 ± 0.01
60	0.44 ± 0.03	0.36 ± 0.04	0.29 ± 0.01	0.26 ± 0.03	0.23 ± 0.02	0.21 ± 0.01	0.19 ± 0.02	0.15 ± 0.02
90	0.37 ± 0.01	0.25 ± 0.01	0.21 ± 0.02	0.21 ± 0.02	0.20 ± 0.01	0.19 ± 0.02	0.16 ± 0.02	0.17 ± 0.02
120	0.79 ± 0.05	0.72 ± 0.02	0.55 ± 0.03	0.43 ± 0.05	0.41 ± 0.04	0.32 ± 0.03	0.32 ± 0.04	0.34 ± 0.02
150	0,45 ± 0.03	0.56 ± 0.07	0.32 ± 0.03	0.19 ± 0.03	0.19 ± 0.01	0.21 ± 0.02	0.22 ± 0.02	0.23 ± 0.03
180	0.55 ± 0.02	0,46 ± 0.03	0.59 ± 0.05	0.53 ± 0.03	0.34 ± 0.03	0.25 ± 0.02	0.21 ± 0.02	0.20 ± 0.02
210	0.46 ± 0.04	0.59 ± 0.03	0.42 ± 0.01	0.37 ± 0.03	0.33 ± 0.02	0.32 ± 0.02	0.32 ± 0.03	0.32 ± 0.01
240	0.64 ± 0.05	0.75 ± 0.04	0.53 ± 0.04	0.34 ± 0.05	0.32 ± 0.02	0.29 ± 0.01	0.27 ± 0.02	0.25 ± 0.03

Experimental dependences of the effective diffusion coefficient when salt penetrates into the herring muscle tissue from the distance x at different time points at a temperature of 10 °C are shown in Figure 3.

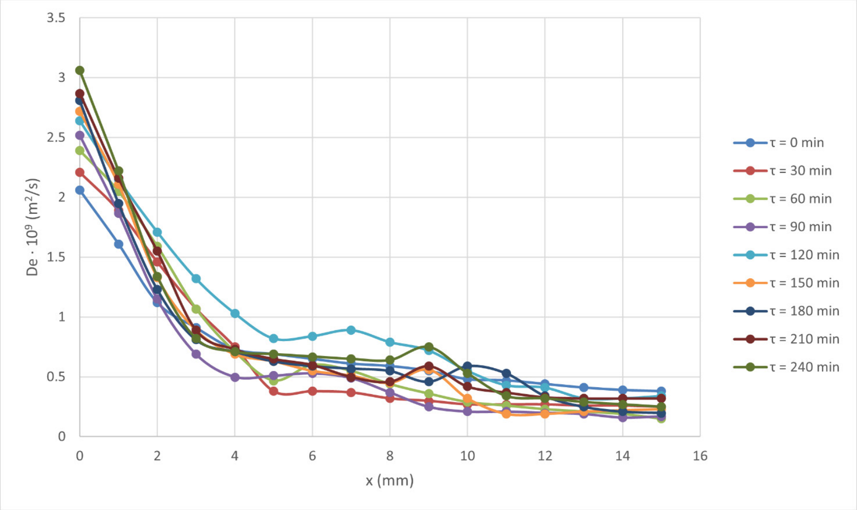

Figure 3. Experimental dependences of the effective diffusion coefficient D_e on the distance x.

According to the data obtained, an average dependence of the effective diffusion coefficient through the thickness of the sample was constructed for all time intervals (Figure 4).

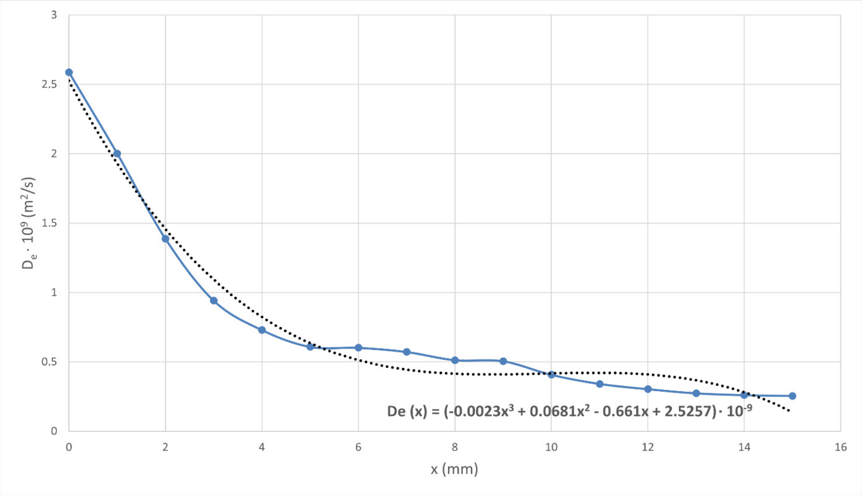

Figure 4. Average dependence of the effective diffusion coefficient D_e on the distance x.

The polynomial distribution function of the effective diffusion coefficient through the sample thickness $D_e(x) = (- 0.0023x^3 + 0.0681x^2 - 0.661x + 2.5257) \cdot 10^{-9}$ was determined by the averaged dependence.

To calculate the duration of salting to the sample center and the average salinity of the sample, the average value of the effective diffusion coefficient was obtained equal $\langle D_e \rangle = 0.73 \cdot 10^{-9}$ m²/s, which was determined as the average value of the continuous function $D_e(x)$ in the interval of [0,15].

Substituting the values of $D_e = 0.73 \cdot 10^{-9}$ m²/s, l = 30 mm, $C_b = 25.9\%$ into Equations 12 and 13, the duration of the salting process was $\tau = 8.74$ hours with an average salt concentration through the thickness of the sample $\langle C \rangle = 4.7\%$.

The obtained values turned out to be quite close and comparable with other researches on the salting process (Dimova et al., 2006).

For a visual representation of the change in salinity in fish during salting, the solution of the differential diffusion equation for the given parameters $D_e = 0.73 \cdot 10^{-9}$ m^2/s, $l = 15$ mm, $C_b = 25.9\%$ is presented graphically in Figure 5.

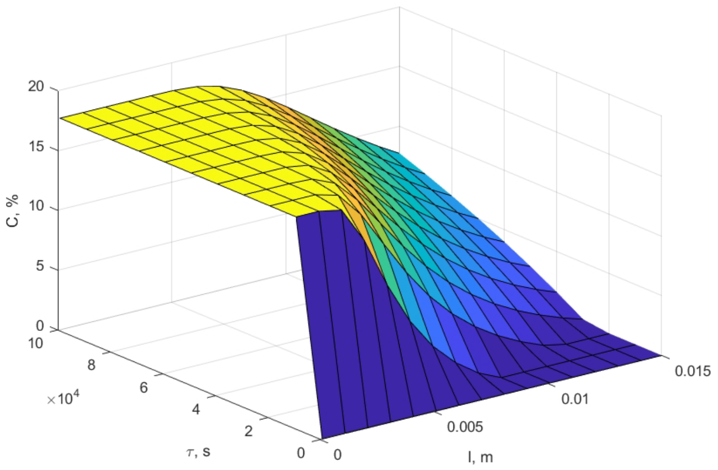

Figure 5. Spatiotemporal distribution of salt concentration in herring muscle tissue.

It can be seen from Figure 5 that the increase in the salt concentration in the center begins only at a certain point in the salting process ($\tau = \tau'$). This model is valid for the time $\tau' > 0.0979\ l^2/D$.

4 Conclusions

A theoretical diffusion model of the process of salting fish in brine without circulation was proposed, obtained by solving Fick's diffusion equation. The model is designed to determine the duration of the salting process and the concentration of salt in the fish muscle tissue.

Effective diffusion coefficients were obtained by the PCS method, taking into account all factors affecting the salting process.

Formulas for calculating the duration of salting and the average salt concentration in the thickness of the muscle tissue of fish were presented, based on which the time of reaching the sample center by salt and the average salt concentration in it were estimated.

Experimental dependences of the effective diffusion coefficient of salt penetration into the muscle tissues of fish have been obtained, which makes it possible to determine the average value of D_e used in the mathematical model.

Experimental results demonstrated that the effective diffusion coefficient has maximum values near the fish skin and decreases by thickness, which is explained by the rheological properties and structural composition of the fish muscle tissue.

A reliable mathematical model is necessary to establish reasonable parameters of the technological process to obtain a salted product with specified properties at minimal production costs.

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