Mathematical Description of Sodium Chloride Diffusion and Its Practical Impact on the Processing of Animal Fleshings

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***Abstract*—** Diffusion of sodium chloride plays key role in the pre-tanning treatment of raw hides, namely in the brine curing and soaking operations. Understanding the transport processes related to these operations gives ground for their optimization and thus to reduction of the adverse environmental impact as well as minimization of the consumption of sodium chloride, water and energies. Mathematical model of raw hide curing is presented and its modification for spherical coordinates applied on the description of the desalting of fleshings, a cheap raw material for the production of biodiesel and high quality gelatin. Desalting is the key step in the fleshings complex processing and the quality of the resulting products is highly dependent on the precise performance of the desalting operation. The experimental part includes determination of the effective diffusion coefficient of NaCl during brine curing on model porous material using color indication of the border area, determination of the diffusion coefficient of NaCl in the inner volume of fleshing during desalting, and pilot scale testing of the efficiency of fleshings desalting. The obtained diffusion coefficients were compared with the theoretical value at infinite dilution and factors affecting the diffusion coefficient values were discussed.

***Keywords*—** Diffusion, sodium chloride, fleshings, mathematical modeling, transport parameters

I. Introduction

TRANSPORT phenomena related to the transport of sodium chloride (NaCl) in raw hide plays significant role in the key operations during which raw hide is prepared for further processing, namely preservation and soaking. Transport of molecules inside the hide is closely related to the hide texture, which fundamentally affects not only the properties of final leather, but also predetermines the requirements for curing [1].

Curing with NaCl is so far the most common way of long-term raw hide preservation [2]. In spite of indisputable advantages, this curing method entails some serious issues the most important of which is negative impact on the environment, especially with huge amount of TDS (Total Dissolved Solids). This applies to both curing and soaking (desalting) operations. Therefore, great efforts are put into optimization of the said operations, which includes especially consumption of the preservation agent, water and energies.

Modern approach to optimization is application of theoretical tools of chemical engineering, *i.e.* mathematical modeling of the physical-chemical processes [3]. Detailed description of transport phenomena related to NaCl diffusion in the hides is available only for soaking. First mathematical description of non-stationary diffusion of NaCl during soaking of cattle hides was described in [4]. A comprehensive study on soaking was published on [5]. Among other conclusions, the authors state that the sorption of NaCl to the collagen mass can be neglected and the diffusion takes place along the real pore length which is not significantly affected by swelling during the soaking operation. The results of the said study were used for optimization and control of the soaking operation [6, 7]. Kolomaznik et al. [8] modeled soaking in tannery drum as a non-isotermic and non-adiabatic reactor using the model of continuous reaction [9]. This paper shows the importance of mathematical modeling in rationalization of tannery operations on the example of so-called concentration shock, which is a collapse of the hide texture resulting from dramatic concentration changes during removal of NaCl from the inner volume of the hide. Kolomaznik et al. [10] further extended their research and proposed three-dimensional model of the transport processes that take place during soaking.

In food industry, curing with NaCl has been mathematically described mostly in relation to cheese and meat salting, for example in 11, 12], with determination of the related diffusion coefficients and their dependence on temperature, NaCl concentration and time of curing. [13] dealt with the numerical solution of NaCl diffusion in potatoes and modeled concentration fields of NaCl in various times from the salting

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initiation. Very few reports on modeling of raw hide curing are available, with an exception of [2] where the concentration field and time concentration of NaCl in brine solution were modeled in dependence on the ratio of the volumes of brine solution and cured hide (soaking number – *Na*).

Great attention is also paid to experimental determination of diffusion coefficients of NaCl, the values of which are essential for determination of dependencies (time, concentration, temperature, *etc*.) and thus verifications of the proposed mathematical models. Conventional ways applicable to all the above mentioned materials (food, hides) are usually based on chemical or conductometric determination of NaCl concentrations (either its decrease during salting, or increase during soaking). However, as said above, the diffusion is strongly affected by the texture of the respective material. From this point of view, measurements of concentration give us no idea on the space distribution of NaCl in the material. This problem has been pointed out recently by several authors [2, 14, 15].

One of the solutions suggested by [15] is the use of SL-NVRK (solid liquid non-volatile release kinetic method), NMR (nuclear magnetic resonance) or possibly NMR using "pulsed field gradients". Special cases of NMR (^{23}Na magnetic resonance imaging MRI and ^{23}Na NMR relaxometry) have been applied in visualization of NaCl distribution during meat salting [14]. The only publication addressing this issue in raw hides is [2], who tracked NaCl diffusion into the hide inner volume with the use of epifluorescent microscope and the fluorescent dye CoroNa^{TM}.

Another "salt issue" is arising nowadays in the field of utilization of primary wastes from leather industry (especially fleshings) for the production of biofuels and other valuable products [16]. Animal fleshings contain considerable amount of both fat and protein. To reach the maximal profit from this raw material it is advantageous to process the said fractions simultaneously. Very few reports are available in the literature on complex processing of fleshings, with an exception of [17]. One of the major technological barrier is NaCl content in the fleshings which negatively affects the product quality, be it biodiesel from the fat fraction or gelatin from the protein. The quality of gelatin is given by so-called Bloom value, which is in direct proportion to ash content, in our case particularly NaCl. For this reason the fleshings desalting is one of the key operations in the complex technology of fleshing conversion into commercially promising products.

From the mechanical engineering point of view, the desalting operation can be performed in a similar way like the soaking, i.e. by washing in water enhanced by mechanical movement of the desalting device. The washing liquid (which may also include surfactants or other supporting agents) dissolves the salt and the established concentration gradient provides the flow of more liquid. During the desalting operation it is necessary to follow technological parameters the most important of which are the ratio of the washing liquid to the solid phase (so called soaking number), time of the desalting, temperature of the washing liquid and concentration of the supportive agents if involved.

The main objectives of our contribution were mathematical description of transport phenomena related to NaCl transport during raw hide curing and desalting of fleshings including experimental determination of the effective diffusion coefficients as a ground for determination of optimal conditions for the desalting of fleshings in the production of biodiesel and gelatin.

II. Theory

A. Mathematical model of raw hide preservation

The model of raw hide curing is based on the continuous reaction model [9]. We assume asymmetric diffusion of salt into the hide from the flesh towards the grain side. The model is represented by a partial differential equation of parabolic type (Fick's second law) (2.1) under specific initial and boundary conditions (2.1a-c).

$$\frac{\partial c}{\partial \tau}(x,\tau) = D\frac{\partial^2 c}{\partial x^2}(x,\tau) \quad 0 < x < b \quad \tau > 0 \tag{2.1}$$

$$\frac{\partial c}{\partial x}(0,\tau) = 0 \tag{2.1a}$$

$$c(b,\tau) = \varepsilon c_S \tag{2.1b}$$

$$c(x,0) = 0 \tag{2.1c}$$

Boundary condition (2.1a) stands for the fact that the problem is not solved symmetrically, in other words that the diffusion proceeds from the flesh towards the grain side only. Condition (2.1b) expresses ideal mass transfer between the brine solution and the hide surface (in practice ensured by intensive stirring). Initial condition (2.1c) says that NaCl in the hide is zero in $\tau = 0$, i.e. at the beginning of the curing process.

For the model solution it is advantageous to introduce the following dimensionless parameters:

$$C = \frac{c}{\varepsilon c_S} \quad X = \frac{x}{b} \quad Fo = \frac{D\tau}{b^2} \tag{2.2a,b,c}$$

The model in dimensionless form is expressed by (2.3) together with the initial and boundary conditions (2.3a-c):

$$\frac{\partial C}{\partial Fo}(X,Fo) = \frac{\partial^2 C}{\partial X^2}(X,Fo) \quad 0 < X < 1 \quad Fo > 0 \tag{2.3}$$

$$C(1,Fo) = 1 \tag{2.3a}$$

$$\frac{\partial C}{\partial X}(0, Fo) = 0 \tag{2.3b}$$

$$C(X,0) = 0 \tag{2.3c}$$

After Laplace transformation we get (2.4) describing a non-stationary concentration field of NaCl in the hide.

$$C = 1 - \sum_{n=1}^{\infty} \frac{\cos\left[X(2n-1)\frac{\pi}{2}\right] e^{-\frac{Fo(2n+1)^2\pi^2}{4}}}{(2n-1)\pi(-1)^{n+1}} \tag{2.4}$$

The average integral concentration for the calculation of the time necessary for proper curing is expressed by (2.5).

$$\overline{C} = \int_0^1 C(X, Fo) dX \tag{2.5}$$

After integration with the use of (2.5) we get the resulting (2.6).

$$\overline{C} = 1 - 2\sum_{n-1}^{\infty} \frac{(-1)^{2n+3} e^{-\frac{Fo(2n-1)^2\pi^2}{4}}}{(2n-1)^2\pi^2} \tag{2.6}$$

B. Mathematical model of fleshing desalting

In mathematical description of the desalting of fleshings we proceeded from the results of [5]. Besides other conclusions, the authors state that it is possible to neglect the sorption of NaCl to the collagen mass and the diffusion takes place along the real pore length which does not change significantly by swelling.

The fleshings are stirred in cold water resulting in the formation of small spherical pellets. The desalting model describes NaCl diffusion from the pellet inner volume into the washing water (2.7), under initial and boundary conditions of (2.7a-e).

$$\frac{\partial c(r,\tau)}{\partial \tau} = D\left[\frac{\partial^2 c(r,\tau)}{\partial r^2} + \frac{2}{r}\frac{\partial c(r,\tau)}{\partial r}\right] \tag{2.7}$$

$$\tau > 0 \quad 0 < r < R_1$$

$$\frac{\partial c(0,\tau)}{\partial r} = 0 \tag{2.7a}$$

$$c(r,0) = c_p \tag{2.7b}$$

$$c_0(0) = 0 \tag{2.7c}$$

$$c(R_1,\tau) = \varepsilon c_0(\tau) \tag{2.7d}$$

$$-SD\frac{\partial c(R_1,\tau)}{\partial r} = V_0 \frac{\partial c_0(\tau)}{\partial \tau} \tag{2.7e}$$

For the model solution it is advantageous to introduce the following dimensionless parameters:

$$C = \frac{c_p - c}{c_p} \quad C_0 = \frac{\varepsilon c_0}{c_p} \quad Fo = \frac{D\tau}{R_1^2} \quad R = \frac{r}{R_1} \tag{2.8a-d}$$

By integration of (2.7) we get the equation describing the dimensionless concentration field of NaCl in the pellets (2.9):

$$C = \frac{Na}{\varepsilon + Na} + \frac{2Na}{3\varepsilon} +$$

$$+ \sum_{n=1}^{\infty} \frac{\frac{\sin(R\, q_n)}{R\, q_n}\exp\left(-Fo\, q_n^2\right)}{\sin(q_n)\left[\frac{Na}{3q_n+\varepsilon} + \frac{1}{q_n} - \frac{1}{q_n^3}\right]\cos(q_n)\left[\frac{1}{q_n^2} + \frac{Na}{3\varepsilon}\right]} \tag{2.9}$$

$$\operatorname{cotg}(qn) = \frac{Na\, q_n}{3\varepsilon} + \frac{1}{q_n} \tag{2.10}$$

Equation (2.11) describes the dimensionless concentration of NaCl in the washing water.

$$C_0 = \frac{\varepsilon}{\varepsilon + Na} - \frac{2Na}{3\varepsilon}\sum_{n=1}^{\infty} \frac{\exp\left(-Fo\, q_n^2\right)}{1 + \frac{Na}{\varepsilon} + \frac{Na^2 q_n^2}{9\varepsilon^2}} \tag{2.11}$$

The washing efficiency can be calculated according to (2.12):

$$y = \frac{V_0 C_0}{C_p V} = \frac{NaC_0}{\varepsilon} = \frac{Na}{\varepsilon + Na} - \frac{2Na^2}{3\varepsilon^2}\sum_{n=1}^{\infty} \frac{\exp\left(-Fo\, q_n^2\right)}{1 + \frac{Na}{\varepsilon} + \frac{Na^2 q_n^2}{9\varepsilon^2}} \tag{2.12}$$

Where q_n stands for the roots of (2.10) and *Na* stands for the ratio between the volume of washing water and the solid phase (the soaking number). Graphical expressions of (2.9) and (2.12) are shown in the following Fig. 1 and Fig. 2, respectively.

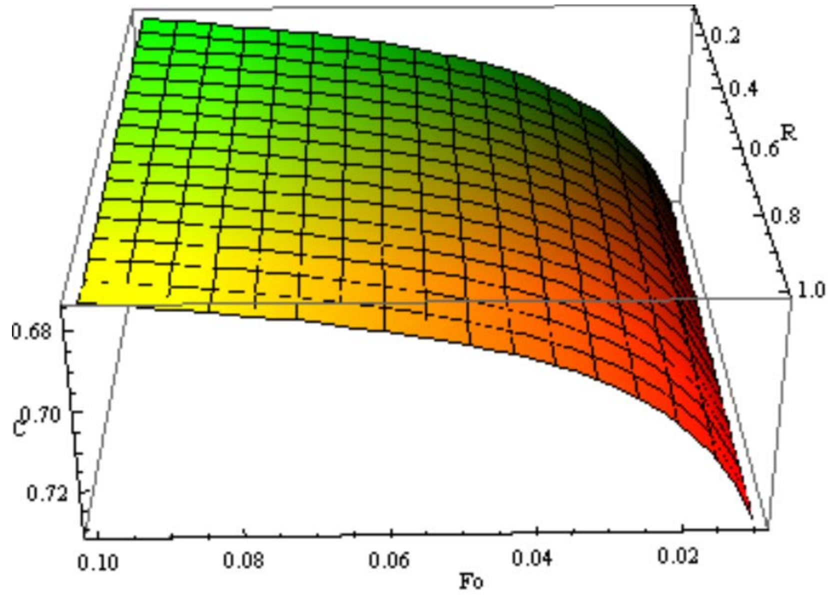

Fig. 1 Dimensionless concentration field of NaCl in the solid phase (the fleshings pellets).

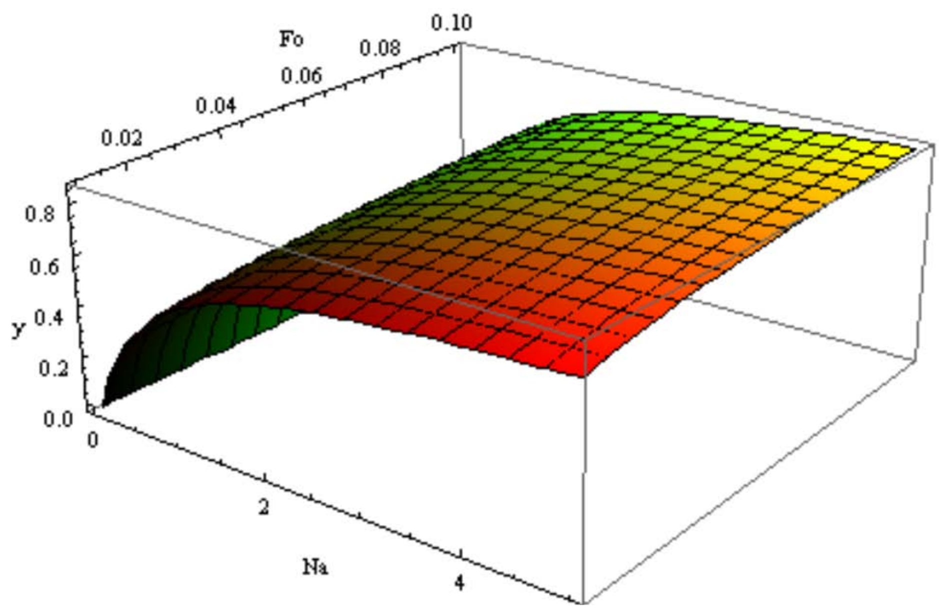

Fig. 2 Dependence of the desalting efficiency on the dimensionless time and consumption of washing liquid (*Na*).

C. Determination of the effective diffusion coefficient of NaCl during salt curing

For the calculation of the value of the effective diffusion coefficient from experimental data we proceeded from the assumption that the NaCl concentration within the solid phase decreases from its source (in practice from the raw hide surface with initial NaCl concentration c_p) towards the solid phase inner volume. In a certain distance from the source, NaCl concentration is practically zero. However, this critical concentration (a) shifts with time further into the inner hide volume. The speed of this shift is proportional to the diffusion flow of NaCl on the considered border. Physical interpretation of this process is built on the non-reacted nucleus model [9].

$$Sc_p \frac{da}{d\tau} = SD_{ef} \frac{(c_p - c_{\min})}{a} \tag{3}$$

Assuming quasi-stationary conditions when $c_{min} << c_p$, equation (3) can be expressed as follows:

$$\frac{da}{d\tau} = \frac{D_{ef}}{a} \tag{4}$$

where D_{ef} is the effective diffusion coefficient of NaCl in the solid phase [$m^2 \cdot s^{-1}$], τ stands for time [s] and a [m] denotes the distance from the source with the initial concentration of NaCl c_p.

III. Experimental

The experimental part comprises three main parts, namely determination of the effective diffusion coefficient of NaCl and its dependence on initial NaCl concentration during salt curing on model porous material using color indication of the border area, determination of the effective diffusion coefficient of NaCl in the inner volume of fleshing during desalting, and pilot scale testing of the efficiency of fleshings desalting.

A. Determination of the effective diffusion coefficient of NaCl during curing on model porous material

Effective diffusion coefficients for various initial concentrations of NaCl were determined as well as their dependence on the initial concentration in model porous material represented by cellulose (filtration paper). Visual identification of the critical border described in the theoretical part was based on Mohr determination of chloride ions and the principal of different precipitation products giving specific colors for different substances. This principle is described in detail in [18].

On moistened filtration paper was applied 0.05 ml of NaCl aqueous solution progressively of the following concentrations (w/v): 1%, 5%, 10%, 15%, 20%, 26% and 30%. Progress of the NaCl diffusion was monitored for each of the said concentrations after the following time periods (in minutes): 5, 15, 30, 60, 90, 120, 240, 360, 600, 960 and 1140. After the respective time period, the filtration paper was sprayed with 1% (w/v) solution of silver nitrate ($AgNO_3$) and subsequently with 1% (w/v) solution of sodium sulfide (Na_2S). The obtained color boundary was marked, photographed and processed in computer with the use of technical image analysis tool to determine the color area. The areas were assumed to possess circular shape, from which the critical distance a was calculated as its radius. An example of the graphs obtained is shown in Fig. 3.

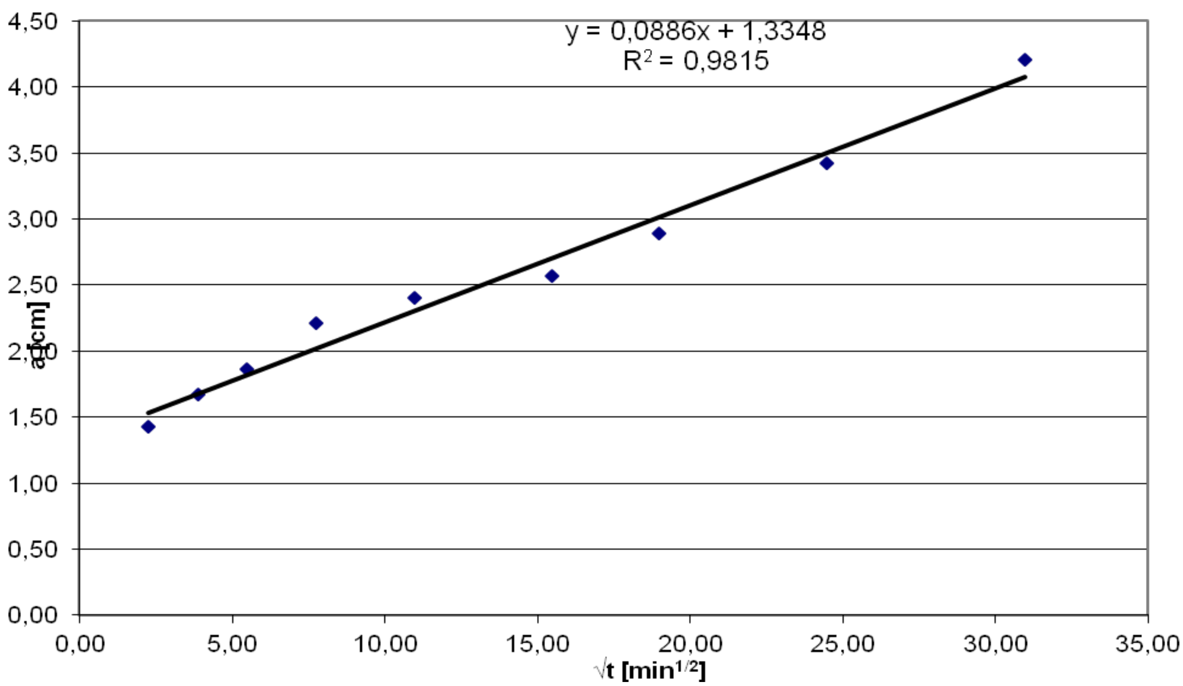

Fig. 3 Example graph of the dependence of critical distance a in cellulose on the square root of time. The graph corresponds to the initial NaCl concentration of 26% (w/v).

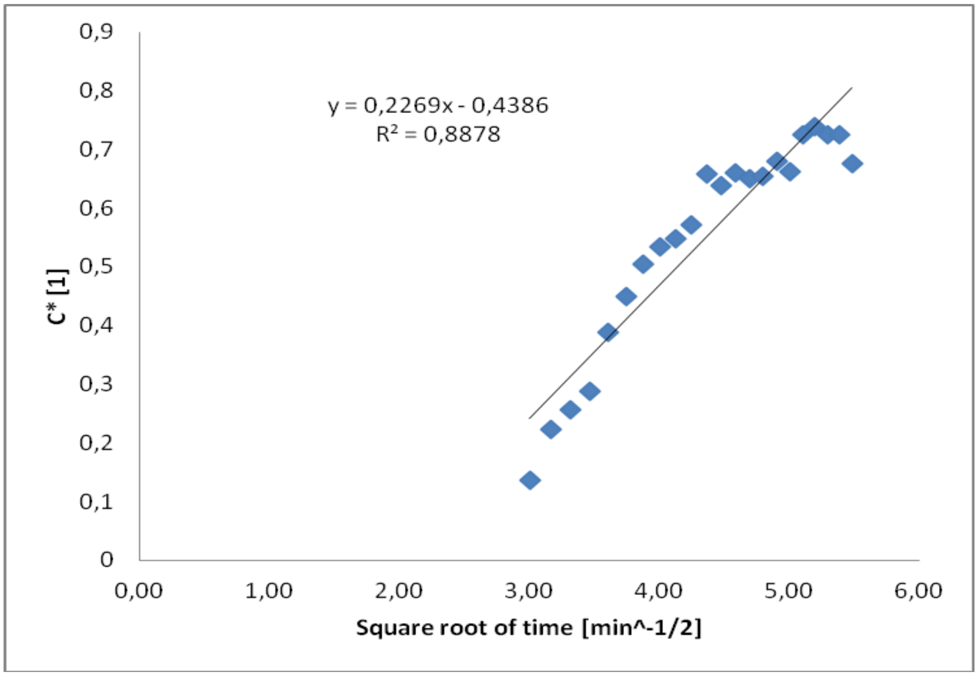

Fig. 4 Dependence of the dimensionless concentration of NaCl in the fleshings on the square root of time.

B. Determination of the effective diffusion coefficient of NaCl in the inner volume of fleshings

The effective diffusion coefficient was calculated from the dimensionless soaking number *Na* and transport parameter λ.

For the laboratory scale the data was the following:
Weight of water = 432.06 g
Density of water = 1 g cm^{-3}
Weight of fleshings = 172. 66 g
Density of fleshings = 0.90 g cm^{-3}

The dimensionless soaking number *Na* is calculated like the ratio of the volume of washing liquid to the volume of solid phase and is equal to 2.25.

For the determination of the diffusion coefficient it was necessary to determine the dependence of dimensionless concentration of NaCl in the fleshings of the square root of time. The dimensionless concentration was obtained by calculation from the conductance of the washing liquid.

The fleshings together with demineralized water were put into reaction flask and mixed intensively at laboratory temperature. Every minute, the conductance of the washing liquid was measured. By linear regression we obtained the slope of the straight line *k* (Fig. 4) needed for the calculation of the transport parameters λ according to Crank [19].

$$k = \frac{2}{\sqrt{\pi}} \frac{1+Na}{Na} \sqrt{\lambda} \tag{5}$$

Transport parameter λ according to (5) is:

$$\sqrt{\lambda} = \frac{0.2269 \cdot \sqrt{\pi} \cdot Na}{2 \cdot (1+Na) \cdot 60}$$

$$\lambda = \frac{0,0002^2 \pi}{240} \cdot \left(\frac{Na}{1+Na} \right) = 3.23 \cdot 10^{-4} s^{-1}$$

The diameter of the spherical particle was 1 mm, *i.e.* the radius *R* was 0.5 × 10^{-3} m.

The diffusion coefficient is calculated according to the following equation:

$$D = \lambda \cdot R^2 \tag{6}$$

The resulting diffusion coefficient of NaCl in the fleshings during the desalting operation is 8.08 × 10^{-11} m^2·s^{-1}.

C. Determination of the efficiency of fleshings desalting

The pre-fleshings were obtained in local tannery (Tarex, Ltd., Otrokovice, Czech Republic). The acid number of the fleshings was 2.43 mg KOH/g, dry matter content was 57.56 % w/w, sodium chloride content was 5 % w/w (related to the dry matter content). Demineralized water was used in all experiments. Sodium chloride determination was performed by conductometric titration. The equilibrium was detected from the graph of the dependence of the solution conductance on silver nitrate consumption. All chemicals used in various analyses were of analytical grade.

A total of 3 kg of fresh fleshings were loaded into reactor (washing machine) together with 12 kg of demineralized water. The reaction mixture was intensively mixed at laboratory temperature for one hour. After this period, the water layer (lower phase) was separated from the fat layer (upper phase) with the use of filtration cloth and samples were taken for sodium chloride content determination. This presents one cycle of decantation washing. In the next cycle, 12 kg of demineralized water were added to the filtration cake obtained from previous cycle and the procedure was repeated. Totally, four cycles were done.

IV. Results and Discussion

Unlike the mathematical model of raw hide curing, the mathematical model of desalting of fleshings is solved as a symmetrical problem in spherical coordinates. It can be explained by the properties of the raw material - in raw hide the diffusion proceeds from the flesh towards the grain side only and the hide is considered to be a "plate", while the fleshings particles take spherical shape and the diffusion of sodium chloride takes place from the center towards the margins.

A. Determination of the effective diffusion coefficient of NaCl during curing on model porous material

Theoretical value of the diffusion coefficient of NaCl was estimated with the use of Nernst-Haskell equation for electrolytes at infinite dilution (7) [20]. This equation is sometimes stated in the form when expression R/F^2 is given its numerical value, *i.e.* 8.9 × 10^{-10}, and $l_+^0 + l_-^0 = \Lambda^0$ [21].

$$D_0 = \frac{RT}{F^2}\left(\frac{l_+^0 l_-^0}{l_+^0 + l_-^0}\right)\left(\frac{z_+ + z_-}{z_+ z_-}\right) \quad (7)$$

where D_0 [$cm^2 \cdot s^{-1}$] is diffusion coefficient of calcium salt at infinite dilution, R [$J \cdot mol^{-1} \cdot K^{-1}$] stands for the universal gas constant, F [$C \cdot mol^{-1}$] denotes Faraday constant, $l_+^0 + l_-^0$ [$S \cdot cm^2 \cdot mol^{-1}$] represent the limit ionic conductance for the cations and anions, T [K] is the absolute temperature and z_+, z_- [1] are the valences of cations and anions. Diffusion coefficient of NaCl at infinite dilution is 1.58 × 10^{-9} $m^2 \cdot s^{-1}$.

Integration and rearrangement of (4) gives the following relation:

$$a = \sqrt{2 D_{ef} \tau} \quad (8)$$

where $\sqrt{2D_{ef}} = k$ and k represents the slope of the straight line obtained by linear regression of plotting radius a on the square root of time for various values of initial NaCl concentration as specified in experimental part. The resulting values of the effective diffusion coefficients of NaCl in the model porous material for various initial NaCl concentrations are summarized in Table I.

Table I Effective diffusion coefficients of NaCl in cellulose for various initial concentrations of NaCl.

Initial concentration of NaCl [% w/v]	**Effective diffusion coefficient [× 10^{-9} $m^2 \cdot s^{-1}$]**
1	1.56
5	2.73
10	5.81
15	6.04
20	4.55
26	6.54
30	4.00

As can be seen in Table I, the values of the effective diffusion coefficients do not differ significantly within the initial NaCl concentration range from 10-30% w/v, therefore from the viewpoint of the proposed mathematical model the dependence of effective diffusion coefficient on the initial NaCl concentration can be neglected. The table also shows that the lower the initial concentration, the more the values of the effective diffusion coefficients get closer to the theoretical value calculated according to (5).

B. Determination of the effective diffusion coefficient of NaCl in the inner volume of fleshings

The value of the effective diffusion coefficient differs from the theoretical value by one order of magnitude. According to [5], the value of the effective diffusion coefficient can be adversely influenced by many factors; in soaking of cured hides it can be for example the dry matter content of the hides or the content of fat in the remaining subcutaneous issue. The latter seems to be one of possible explanations since the fleshings contain high fat portion.

C. Determination of the efficiency of fleshings desalting

The values of filtrate and filtration cake weights and the content of sodium chloride in the filtrate after each cycle from the pilot scale tests are listed in Table II.

Table II Weights of filtrates and filtration cakes after each decantation washing cycle.

washing cycle	**filtrate [kg]**	**filtration cake [kg]**	**NaCl [g]**
1	11.44	2.34	53.66
2	11.86	2.42	7.69
3	11.32	2.42	1.54
4	12.20	2.10	0.68

The overall efficiency of the four washing cycles was calculated from the obtained amounts of sodium chloride:

$$\frac{54 + 7.6 + 1.54 + 0.68}{86.4} \cdot 100 = 73.6\%$$

The experimental determination of the efficiency of decantation washing was based on the results stated in [5]. The author of the paper suggests that soaking is a diffusion process which removes salt from the inner volume of cured hides. Other factors such as protein removal were not considered. It can be assumed from the results of the said work that determination of chlorides in the soaking (washing) bath can serve as a criterion for the evaluation of the quality of soaking process.

Preliminary experiments were carried out for the utilization of fleshings in the production of gelatin. The results showed that the gelatin quality (represented by the Bloom value) was significantly higher from the fleshings subjected to the desalting operation than from the fleshings on which the desalting procedure was not applied (158 and 32, respectively).

V. Conclusion

Mathematical description of NaCl diffusion in the hide during brine curing is presented and its modification for spherical coordinates is applied to the mathematical model of the desalting of animal fleshings. The desalting step determines the utilization of fleshings in the production of valuable products such as biodiesel and gelatin. The experimental part includes determination of effective diffusion coefficient of NaCl during brine curing on model porous material, determination of the effective diffusion coefficient of salt in the fleshings inner volume and pilot scale determination of the efficiency of the desalting process. The desalting process facilitated substantial reduction in sodium chloride content in the fleshings. As a result, protein fraction with low amount of ash and high gel strength was obtained. Furthermore, the losses of protein and fat during the desalting operation were negligible.

VI. List of symbols

a	Radius of the salt boundary	[m]
b	Raw hide thickness	[m]
c	NaCl conc. in hide/fleshings	[kg·m^{-3}]
c_{min}	NaCl boundary concentration	[kg·m^{-3}]
c_p	Initial NaCl conc. in cellulose	[kg·m^{-3}]
c_S	NaCl conc. saturated solution	[kg·m^{-3}]
D_{ef}	Effective diffusion coefficient	[m^2·s^{-1}]
S	Surface area of hide/fleshings	[m^2]
V	Volume of raw hide/fleshings	[m^3]
V_0	Volume of washing water	[m^3]
x	Position coordinate	[m]
c_0	NaCl conc. in washing water	[kg·m^{-3}]
r	Space variable	[m]
R_1	Radius of the fleshing pellet	[m]
y	Washing efficiency	[1]
ε	Porosity	[1]

Other symbols are explained directly in the text.

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