This study aimed at modeling sodium chloride (NaCl) diffusion in foods during thermal processing using heat and mass transfer, modeling, salt, storage, thermal processing.

Diffusion models can be used by the food industry when optimizing the amount of NaCl added to thermally processed, low-acid foods. Additionally, the results implied that when doing sensory evaluation, it is especially important to wait at least 14 d after processing to allow the salt to equilibrate within the products with less severe heat treatments, such as microwave assisted thermal processing.

Introduction

Worldwide, most people consume too much sodium and excessive sodium consumption has been linked to an increased risk of diseases, such as cardiovascular diseases and hypertension (Dotsch and others 2009). Sodium intake in Europe and North America is mainly from sodium chloride and 75% comes from processed foods (Brown and others 2009). Reduction of sodium chloride in processed foods is an important element of the overall strategy to reduce the amount of sodium in the diet.

Sodium reduction in thermally processed foods is not a simple task, as the addition of NaCl to food serves many purposes, including preservation, texture, and flavor (Doyle and Glass 2010). Retort processing used to produce shelf-stable, low-acid canned foods involves a lengthy exposure to high temperatures, which can increase off-flavors. The addition of salt helps mask bitter flavors developed during thermal processing and improve the overall taste of processed foods (Doyle and Glass 2010). Emerging technologies with shorter processing times to achieve pasteurization or sterilization have a potential benefit in the area of sodium reduction. Less severe thermal processing used in emerging technologies can improve food quality and generate fewer undesirable flavors, which could result in a product that tastes acceptable with less added salt. Microwave-assisted thermal sterilization (MATS) and the microwave-assisted pasteurization system (MAPS) are emerging technologies that can reduce overall processing time compared to conventional methods (Tang 2015).

Diffusion of sodium chloride within a product may be crucial when considering sodium reduction in food using novel technologies with shorter processing times. Many shelf-stable products are multiphase, with the solids and liquid having different initial sodium concentrations. The difference in sodium chloride content between the solids and liquid leads to sodium chloride diffusion during processing and storage. Salt that has diffused into a food is not as readily accessible to the taste receptors in the mouth compared to the salt on the surface of a food, yielding a decreased salty taste (Henney and others 2010).

Sodium chloride diffusion in food has been well researched for some applications, such as cheese and meat salting (Turhan and Kalentuc 1992; Wang and others 2000; Pajonk and others 2003; Telis and others 2003; Gallart-Jornet and others 2007a; Aursand and others 2009; Cierach and Modzelewski-Kapitula 2011; McDonnell and others 2013; Sharedeh and others 2015). However, cheese and meat salting typically use high concentrations of salt in liquid solutions (brine) at low temperatures for relatively long times, which is considerably different from the emphasis of this research. Previous research on salt diffusion during thermal processing has focused on temperatures below 100 °C, except for Liu (1992) who conducted diffusion experiments and modeling at temperatures above 100 °C. Most published research on NaCl diffusion during heat treatment utilizes special experimental set-ups to create one-dimensional diffusion in a slab in order to simplify modeling (Turhan and Kalentuc 1992; Wang and Sastry 1993; Pajonk and others 2003; Telis and others 2003; Gallart-Jornet and others 2007b; Hashiba and others 2007; Sarang and Sastry 2007; Volpato and others 2007; Alizadeh and others 2009; Dehkordi...
and others 2010; Cierach and Modzelewksa–Kapitula 2011). Published work using multiphysics modeling to couple heat and mass transfer for salt diffusion during heating is very limited.

There has been a lack of published data on salt diffusion at high temperatures, a lack of 3-dimensional diffusion models, a lack of multiphysics modeling for salt diffusion at high temperatures, and a lack of published data on salt diffusion during storage after thermal processing. Thus, the goal of this study was to model the 3-dimensional diffusion of sodium chloride in low-acid foods during high temperature thermal processing and subsequent storage. The objectives of this study were to: (I) model NaCl diffusion during thermal processing using an analytical solution, (II) develop and utilize a multiphysics model to perform a numerical simulation of NaCl diffusion during thermal processing, and (III) investigate the NaCl concentration of food products during storage after thermal processing.

Materials and Methods

Sample preparation

Diffusion experiments were conducted using white (Irish) potato (Solanum tuberosum), spring (European) radish (Raphanus sativus), and thawed, flash frozen wild sockeye salmon dorsal muscle (Oncorhynchus nerka) from Bristol Bay, Alaska obtained from a local grocery store. Prior to use in diffusion experiments, frozen salmon fillets were thawed overnight in a 5 °C refrigerator and the skin was removed. Potato, radish, and salmon were selected because of the ability to create a cylindrical shape without destroying the structure and the variation in compositions and structures. Potato had the greatest amount of carbohydrates, about 17.0% wet basis (wb), and it was the only starch containing food among the 3 selected, with about 13.5% wb starch (USDA 2012). Radish had the highest moisture content, measured as 94.1% ± 0.2% wb, and it had a lower amount of carbohydrates (3.4% wb) compared to potato (USDA 2012). Sockeye salmon had the lowest moisture content, measured as 72.0% ± 0.7% wb and highest protein and lipid contents, about 22.6% and 4.5%, respectively (USDA 2012). Salmon was selected as a high-protein food over other foods was minimal, measured to be less than 0.05% ± 0.005% wb or 0.73% ± 0.092% dry basis.

Moisture content and sodium chloride quantification

Sample moisture content was determined gravimetrically by drying with a vacuum oven (ADP-31, Yamato Scientific America Inc., Santa Clara, Calif., U.S.A.) at 70 °C with 60 cm Hg to a constant weight (Part 934.01, AOAC Intl. 1995). Sodium in liquid and food samples was measured using a calibrated ROSS® Sure-Flow® Sodium Combination Ion Selective Electrode (8611BNWP, Thermo Fisher Scientific, Beverly, Mass., U.S.A.) connected to an electrochemical analytical meter (SevenEasy™ S20, Mettler–Toledo AG, Schwerzenbach, Switzerland).

Sodium ion selective electrodes measure the sodium content in liquids and additional preparation steps are needed to measure food samples. Food samples (1.8 to 2 g) were crushed using a glass mortar and pestle with 4 mL deionized water, and filtered to remove large particles (Kindstedt and Kosikowski 1984; Biswal and Le Maguer 1989). Sodium chloride content for food samples was calculated taking into account the 4 mL dilution and reported on both a wet and dry basis using the sample mass and moisture content after cooking. NaCl concentration was reported in both wet and dry basis. Wet basis (wb) is more relevant to salt perception during consumption of a salty, high water content product because when a person consumes the product, they will simultaneously consume and taste the water and salt dissolved in the water (Henney and others 2010). However, to facilitate comparisons across the different food products with different water contents, dry basis (db) NaCl concentration was also used.

Experimental set-up

Diffusion during thermal processing. One cylindrical food sample and 20 mL of sodium chloride solution were placed in a custom-made aluminum test cell equipped with aluminum mesh in the center of the cell to suspend the food sample (Figure 1). A 20 mL solution volume was selected because it provided a sufficient amount of NaCl, but kept the size of the test cell small to facilitate heat transfer. A recirculating ethylene glycol bath (HAAKE DL 30, Thermo Fisher Scientific Inc., Newington, N.H., U.S.A.) was used to heat each test cell. Average come-up time for the geometric center of the food samples to reach within 1 °C of the target temperature was less than 5 min for all temperatures.

A completely randomized design with 3 factors was utilized to study diffusion during thermal processing; the 3 factors were food (potato, radish, or salmon), salt solution (1% or 3% NaCl), and temperature (90, 105, or 121 °C). Samples were heated at 90 °C for 0, 5, 10, 15, 30, 45, 60, 90, 120, 180, 210, and 240 min, and at 105 °C for 0, 3, 6, 10, 15, 30, 45, 60, 90, 120, 180, 210, and 240 min.
180, and 210 min, and at 121 °C for 0, 3, 6, 10, 15, 30, 45, 60, 90, 120, and 180 min. These treatment conditions cover the temperatures and times needed for sterilization and pasteurization by microwave (MATS and MAPS) processing and conventional thermal processing (Tang 2015), as well as the time needed for the salt concentration to reach the apparent equilibrium in the food. At lower temperatures, longer times were needed for the salt concentration in the food to reach the apparent equilibrium.

After heat treatment, test cells were cooled in ice for 3 min to reduce the temperature of the geometric center of the food samples to room temperature (22 °C). Food samples were removed from the NaCl solution immediately followed by dimension, weight, moisture, and sodium content measurement. Every experimental replicate utilized a separate food sample heated in a freshly prepared salt solution. Triplicates were performed for all treatments (that is, all combinations of food, salt solution, temperature, and heating time).

**Diffusion during storage.** Experiments to investigate the NaCl diffusion during storage after thermal processing used a completely randomized design with potato, radish, or salmon in 3% NaCl solution heated to 90 or 121 °C for 10 or 60 min followed by cooling in ice for 3 min. The samples heated at 90 °C were stored in refrigerated conditions (4 °C) and samples heated at 121 °C were stored in ambient conditions (22 °C) for 8 h to 28 d. These conditions were selected in order to study the diffusion of NaCl within a product during storage following heat treatment that simulated MATS (approximately 10 min at 121 °C followed by storage at 22 °C), conventional retort thermal sterilization (approximately 60 min at 121 °C followed by storage at 22 °C), and MAPS or conventional pasteurization (approximately 10 min at 90 °C followed by storage at 4 °C) (Tang 2015). During storage at 4 or 22 °C, food samples remained in the same 3% NaCl solution utilized during heating. After storage, the food samples were removed from the NaCl solutions and the weight, moisture content, and sodium content were measured. Each experimental replicate utilized a separate food sample and triplicates were performed for all treatments.

**Statistical analysis.** Statistical analysis was completed using SAS® 9.2. The general linear model for an analysis of variance and Fisher's least significant difference for a completely randomized design were utilized with a P-value for significance of less than 0.05. The model assumptions of the data following a normal distribution and constant variance were tested before performing the statistical analysis. Statistical analysis was performed for data from both the diffusion during heating and diffusion during storage studies. The analysis of the salt concentration data from the diffusion during storage study was done separately for each food.

**Analytical diffusion model setup**

Fick's second law was used to describe a diffusion driven change in concentration over time as (Crank 1956)

\[
\frac{\partial C}{\partial t} = D \nabla^2 C
\]

where \(C\) is the concentration of the diffusing component (% w/w), \(t\) is time (s), \(D\) is the diffusion coefficient (m²/s), and \(\nabla\) is the vector differential operator. Diffusion of sodium chloride into a cylindrical food sample was modeled using the unsteady state series solution of Fick's second law for a finite cylinder. This solution used an initial NaCl concentration of zero from a stirred solution of limited volume with uniform initial NaCl concentration and no surface resistance. The solution for a finite cylinder was calculated as the intersection of the solutions for an infinite cylinder and infinite slab (Crank 1956; Datta 2002):

\[
\frac{C_{\text{surf, food}}}{C_{\text{surf, solution}}} = \left( \frac{G_{\text{surf, food}}}{G_{\text{surf, solution}}} \right)_{\text{infinite, slab}} \times \left( \frac{C_{r, \text{food}}}{C_{r, \text{solution}}} \right)_{\text{infinite, slab}}
\]

(2)

where \(G_{\text{surf, food}}\) is the average sodium chloride concentration in the food (% w/w) at time \(t\) (s) and \(C_{\text{surf, food}}\) is the average equilibrium sodium chloride concentration in the food (% w/w) after infinite time.

The unsteady state series solution of Fick's second law for an infinite cylinder is defined as (Crank 1956)

\[
\left( \frac{C_{\text{surf, food}}}{C_{\text{surf, solution}}} \right)_{\text{infinite, slab}} = 1 - \sum_{n=1}^{\infty} \frac{4\alpha (1 + \alpha)}{4 + 4\alpha + \alpha^2 q_n^2} \exp \left( -D_{\text{eff}} q_n^2 t \frac{r^2}{\ell^2} \right)
\]

(3)

where \(r\) is the radius of the cylindrical food sample (m), \(t\) is the time (s), \(D_{\text{eff}}\) is the effective sodium chloride diffusion coefficient into the food (m²/s), and \(q_n\) is the positive, nonzero root of the expression (Crank 1956):

\[
\alpha q_n J_0 (q_n r) + 2 J_1 (q_n r) = 0
\]

(4)

where \(J_0\) is the Bessel function of the first kind of order zero and \(J_1\) is the Bessel function of the first kind of order one. The parameter \(\alpha (1/m)\) in Eqs. 3 and 4 is defined as (Crank 1956)

\[
\alpha = \frac{A}{rK}
\]

(5)

where \(A\) is the radius of the container, excluding the space occupied by the food (m) and \(K\) is the equilibrium distribution (partition) coefficient. The solute equilibrium distribution (partition) coefficient, \(K\) (dimensionless) was calculated for each sample using (Crank 1956; Wang and Sastry 1993):

\[
K = \frac{C_{\text{surf, food}}}{C_{\text{surf, solution}}}
\]

(6)

where \(C_{\text{surf, food}}\) is the average equilibrium sodium chloride concentration in the food (% w/w, wb) and \(C_{\text{surf, solution}}\) is the equilibrium sodium chloride concentration in the liquid (% w/w). For each temperature, food, and salt solution combination, the food and salt solution equilibrium concentrations were calculated as an average of triplicate concentration measurements at 3 time points for a total of 9 measurements (121 °C: 90, 120, and 180 min of heating, 105 °C: 120, 180, and 210 min, 90 °C: 180, 210, and 240 min). The distribution (partition) coefficient describes the maximum amount of NaCl diffusion that has occurred at a given temperature. A larger \(K\) indicates that more sodium chloride has diffused from the solution to the food.

The unsteady state series solution of Fick's second law for an infinite slab is defined as (Crank 1956):

\[
\left( \frac{C_{\text{surf, food}}}{C_{\text{surf, solution}}} \right)_{\text{infinite, slab}} = 1 - \sum_{n=1}^{\infty} \frac{2\alpha (1 + \alpha)}{1 + \alpha^2 q_n^2} \exp \left( -D_{\text{eff}} q_n^2 t \frac{r^2}{\ell^2} \right)
\]

(7)
where $\ell$ is the length of the cylindrical food sample or slab thickness (m), and $z_n$ is the positive, nonzero root of the expression (Crank 1956):

$$\tan (z_n) + \alpha_i (z_n) = 0 \quad (8)$$

The parameter $\alpha_i$ (1/m) in Eqs. 7 and 8 is defined as (Crank 1956)

$$\alpha_i = \frac{A_i}{\ell K} \quad (9)$$

where $A_i$ is the length of the container, excluding the space occupied by the food (m).

The diffusion model used a finite cylinder series solution (Eq. (2)), which combined the first 6 terms of both the infinite cylinder (Eq. (3)) and infinite slab (Eq. (7)) series solutions. Six terms were used in the series solution because preliminary testing showed that the addition of more than 6 terms did not improve the fit of the model to the experimental data. The effective diffusion coefficient was determined by fitting the finite cylinder solution (Eq. (2)) to the experimental data (triplicate measurements included separately, not averaged) using nonlinear least squares curve fitting in MATLAB 2012b. After the effective diffusion coefficient was determined, the predicted average NaCl concentration in the food was compared to the experimentally measured concentration at each time point. The same experimental data was used to fit the diffusion equation and to compare the experimental and predicted NaCl values. This approach provided sufficient credible data to achieve the objectives. The coefficient of determination ($R^2$) was calculated for each data fitting.

Arrhenius relationship for effective diffusion coefficients

The effect of temperature on the effective NaCl diffusion coefficients in food samples was described using the Arrhenius equation (Liu 1992):

$$D_{eff} = D_0 \exp \left( - \frac{E_a}{RT} \right) \quad (10)$$

where $D_0$ is the reference temperature constant (m$^2$/s), $E_a$ is the activation energy (kJ/mol), $R$ is the universal gas constant (0.008314 kJ/K/mol), and $T$ is the temperature (K). The reference temperature constant and activation energy were determined for each food and salt solution combination and the coefficient of determination ($R^2$) was calculated for each exponential regression.

Numerical diffusion model setup

**Governing equations and assumptions.** In addition to an analytical diffusion model, a numerical, multiphysics model was developed for salt diffusion during heating. There are numerous benefits of using a multiphysics model compared to an analytical model. Advantages of the multiphysics approach include a more detailed, realistic, and reliable model based on the laws of conservation, fewer experiments are necessary, the ability to predict various scenarios (for example, more complex system geometries), and the ability to utilize the model for process optimization (Datta 2008). Additionally, the use of a multiphysics model allowed for coupling of the heat and mass transfer phenomena occurring during the salt diffusion process.

A numerical model was developed to simulate NaCl diffusion during heating of fresh potato, fresh radish, and thawed, frozen sockeye salmon samples in 1% NaCl solution at 121 °C for 120 min. The model was developed for a 2-dimensional axisymmetric geometry in COMSOL Multiphysics 4.2a. The food and the test cell were defined as solids and the salt solution was defined as a fluid. The geometry was meshed using triangular elements with rectangular boundary layers at the food-salt solution interface and test cell-salt solution interface. The model was developed using the conjugate heat transfer module and transport of diluted species module.

Momentum transfer in the salt solution was defined as non-isothermal flow with a Boussinesq approximation using (Bird and others 2007):

$$\rho \frac{\partial u}{\partial t} + \rho u \cdot \nabla u = -\nabla p + \nabla \cdot \left( \mu \left( \nabla u + (\nabla u)^T \right) - \frac{2}{3} \mu (\nabla \cdot u) I \right) + \rho \cdot g \cdot \beta (\nabla T - T) \quad (11)$$

where $\rho$ is the fluid density (kg/m$^3$), $t$ is time (s), $u$ is the velocity vector (m/s), $\mu$ is the fluid dynamic viscosity (Pa s), $T$ is the absolute temperature (K), $p$ is the pressure (Pa), $I$ is the unit tensor, $g$ is the acceleration due to gravity (m/s$^2$), $\beta$ is the fluid volumetric thermal expansion coefficient (1/K), and $T_i$ is the initial temperature (K). Fluid flow was assumed to be incompressible and there was a no slip boundary condition ($\nabla u = 0$) at the wall of the test cell and the surface of the food. Pressure and density were both assumed constant. Over the temperature range of this model (20 to 121 °C) the density of pure water decreases by approximately 5.6%; this change is relatively small, thus the constant density assumption is reasonable.

Heat transfer in the salt solution was defined as (Bird and others 2007):

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p u \cdot \nabla T = \nabla \cdot (k \nabla T) \quad (12)$$

where $C_p$ is the specific heat capacity at constant pressure (J/kgK), $k$ is the thermal conductivity (W/mK). Heat transfer in the solid sections was defined as (Bird and others 2007)

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) \quad (13)$$

where variables are as defined above. The heat source for this system was an oil bath, which was defined in the model as an inward heat flux from the outside surface of the test cell using (Bird and others 2007):

$$- n \cdot (-k \nabla T) = h \cdot (T_{ext} - T) \quad (14)$$

where $n$ is the normal vector of the boundary, $h$ is the heat transfer coefficient (W/m$^2$K), and $T_{ext}$ is the external temperature (oil bath temperature) (K). The heat transfer coefficient at the interface between the test cell and the oil bath was determined experimentally (Table 1) using a solid aluminum test cell with the same dimensions as the test cell and a lumped parameter heat transfer modeling method (Datta 2002). At the internal wall boundaries separating the solid and liquid (that is, test cell and solution, food and solution) a temperature continuity boundary condition was utilized.

Mass transfer in the liquid portion of the transport of diluted species module was defined as (Bird and others 2007)
Table 1—Constants used in the COMSOL computer simulation.

<table>
<thead>
<tr>
<th>System parameter and property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial velocity (m/s)</td>
<td>0.0</td>
</tr>
<tr>
<td>Pressure (atm)</td>
<td>1.0</td>
</tr>
<tr>
<td>Initial temperature (°C)</td>
<td>20</td>
</tr>
<tr>
<td>External (oil) temperature (°C)</td>
<td>121</td>
</tr>
<tr>
<td>Heat transfer coefficient (W/m²K)</td>
<td>533.2</td>
</tr>
<tr>
<td>Volumetric thermal expansion coefficient for water (m²/s)²</td>
<td>2.14 × 10⁻⁴</td>
</tr>
<tr>
<td>Effective diffusive coefficient: NaCl in water at 121 °C (m²/s)⁶</td>
<td>4.97 × 10⁻⁶</td>
</tr>
<tr>
<td>Stiff-spring velocity (m/s)⁷</td>
<td>1 × 10⁴</td>
</tr>
<tr>
<td>Initial NaCl solution concentration (% w/w)</td>
<td>0.01</td>
</tr>
<tr>
<td>Density of water (kg/m³)</td>
<td>998.2</td>
</tr>
</tbody>
</table>

Food dimension and property

<table>
<thead>
<tr>
<th>Potato</th>
<th>Radish</th>
<th>Salmon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (mm)</td>
<td>14.27</td>
<td>14.28</td>
</tr>
<tr>
<td>Length (mm)</td>
<td>11.51</td>
<td>11.43</td>
</tr>
<tr>
<td>Density (kg/m³)³</td>
<td>1066</td>
<td>959</td>
</tr>
<tr>
<td>Thermal conductivity (W/mK)⁴</td>
<td>0.571</td>
<td>0.582</td>
</tr>
<tr>
<td>Specific heat capacity, constant pressure (J/kgK)⁵</td>
<td>3340</td>
<td>3950</td>
</tr>
<tr>
<td>Initial concentration (% w/w NaCl)</td>
<td>0.11</td>
<td>0.13</td>
</tr>
</tbody>
</table>

¹From (Cengel and Boles, 2008).
²From (Li and Gregory 1974; Cussler 1997).
³From (COMSOL 2011).
⁴From (Rahman 2009).
⁵From (Souma and others 2004).

The best fit numerical solution was determined using a nonlinear, least-squares approach by comparing the experimental and predicted average salt concentration in the food. Both the effective diffusion coefficient for NaCl in food (D_eff) and the NaCl equilibrium distribution coefficient (K) were varied in the process of finding the best fit numerical solution.

**Convergence study and simulation validation.** A solution convergence study was conducted to examine the influence of the number of elements (mesh size) and time step size on the diffusion coefficient of NaCl in water (m²/s). For the food sample portion of the model, the mass transfer was described by

\[
\frac{\partial c_{\text{food}}}{\partial t} = \nabla \cdot \left( D_{\text{eff,food}} \nabla c_{\text{food}} \right)
\]

(16)

where \(c_{\text{food}}\) is the concentration of the species (NaCl) in the food, and \(D_{\text{eff,food}}\) is the effective diffusion coefficient of NaCl in the food (m²/s). The Stiff-Spring method for boundary conditions was used to account for the continuous flux over the phase boundary between the salt solution and food (COMSOL 2011; Shirkhande 2014). There was a no flux condition at the boundary between the liquid and the aluminum test cell.

Additional system parameters and properties that were assumed constant were the heat transfer coefficient, food sample dimensions, food specific heat capacity, food thermal conductivity, oil bath temperature, volumetric thermal expansion coefficient for water, and the effective diffusion coefficients in water and food. The initial temperature and velocity were assumed uniform for the entire system, as well as the initial NaCl concentrations of the liquid and food. COMSOL default values were used for aluminum and any physical properties of water not listed in Table 1.

The best fit numerical solution was determined using a nonlinear, least-squares approach by comparing the experimental and predicted average salt concentration in the food. Both the effective diffusion coefficient for NaCl in food (\(D_{\text{eff}}\)) and the NaCl equilibrium distribution coefficient (\(K\)) were varied in the process of finding the best fit numerical solution.

**Convergence study and simulation validation.** A solution convergence study was conducted to examine the influence of the number of elements (mesh size) and time step size on the
NaCl diffusion during heating & storage...

Simulation results for both heat and mass transfer. Both the heat and mass transfer had converged on a solution (<1% maximum difference) when 3849 elements were used in the geometry mesh. The convergence study to determine an appropriate simulation time step showed both the heat and mass transfer models had converged on a solution (<1% maximum difference) with either a 0.5- or 0.25-min step.

The simulation was validated by comparing both the heat and mass transfer portions of the model to experimental data. Both the heat and mass transfer validation experiments were done in triplicate and utilized the same scenario: potato in 1% NaCl solution with a target oil bath temperature of 121 °C. For the heat transfer portion, the average absolute value of the temperature difference between the experimental and simulation results during the first 5 min was 4.1 °C and decreased to about 0.3 °C after 10 min. These deviations can be explained by experimental variations, such as test cell movement at the beginning of the experiments and varying initial food temperatures. For the mass transfer portion, the numerical solution showed good agreement with the experimental results, with an average absolute concentration difference of 0.02% w/w NaCl (wb).

**Response time of dimensionless temperatures and concentrations.** The relative rates of heat transfer and salt diffusion were examined by calculating dimensionless temperatures and concentrations for a simulation from 0 to 60 min using a 0.25-min time step for potato in a 1% NaCl solution at 121 °C. The dimensionless temperature ($T_{\text{dimen}}$) was calculated by (Datta 2002)

$$T_{\text{dimen}}(t) = \frac{T - T_{\infty}}{T_i - T_{\infty}}\quad (17)$$

where $T$ is the temperature (°C) at time $t$ (s), $T_{\infty}$ is the temperature at infinite time (°C), and $T_i$ is the initial temperature (°C). All temperatures were determined at the geometric center of the solid. The dimensionless concentration ($C_{\text{dimen}}$) was defined similarly as

$$C_{\text{dimen}}(t) = \frac{C - C_{\infty}}{C_i - C_{\infty}}\quad (18)$$

where $C$ is the concentration (% w/w NaCl (wb)) at time $t$ (s), $C_{\infty}$ is the concentration at infinite time (% w/w NaCl (wb)), and $C_i$ is the initial concentration (% w/w NaCl (wb)). All concentrations were calculated as the average solid NaCl concentration (% w/w wb).

Response time is typically used to describe the lag time of a temperature measurement sensor to assessing a substance’s true temperature (Heldman 2003). Response time can be defined as the time for a sensor to achieve a 63.2% change in temperature (substance temperature minus the initial temperature of the sensor) (Heldman 2003). The concept of response time can be applied to the dimensionless temperatures and concentrations defined above to describe the speed of the parameters’ change. For this research, the response time of the dimensionless parameters was defined as the time it takes for a dimensionless parameter to change by 63.2% of the initial value.

**Results and Discussion**

**Diffusion during thermal processing.** Average NaCl content of heated potato, radish, and salmon samples in 1% and 3% NaCl

![Figure 3–Experimental dry basis NaCl concentrations (3 replicates) with 95% confidence intervals for radish, potato, and salmon in 3% NaCl solution and radish, potato, and salmon in 1% NaCl solution. Predicted concentrations using the analytical diffusion model are shown for each food in 1% and 3% NaCl solutions (___). Heating temperatures are A: 90 °C, B: 105 °C, C: 121 °C.](image-url)
solutions increased with increasing time until an equilibrium concentration was reached (Figure 2). Potato, radish, and salmon samples heated in 3% NaCl solution had significantly higher ($P < 0.05$) average NaCl concentrations (wb and db) than those in 1% NaCl solution throughout the heating time (Figure 2 and 3). The average NaCl equilibrium concentration (% wb) for samples in 1% NaCl solution ranged from 0.52% to 0.61% ± 0.03% for potato, 0.62% to 0.67% ± 0.04% for radish, and 0.55% to 0.61% ± 0.02% for salmon; samples in 3% NaCl solution had average NaCl equilibrium concentrations about 3 times greater, ranging from 1.52% to 1.68% ± 0.06% for potato, 1.81% to 2.04% ± 0.08% for radish, and 1.53% to 1.71% ± 0.06% for salmon.

At each temperature and NaCl solution, the dry basis equilibrium NaCl concentration for radish was significantly larger ($P < 0.05$) than that of potato, followed by salmon with the smallest concentration (Figure 3). The average NaCl equilibrium concentration (db) for samples in 1% NaCl solution ranged from 20.3% to 23.4% ± 1.5% for radish, 3.6% to 4.5% ± 0.2% for potato, and 1.6% to 1.9% ± 0.1% for salmon. For samples in 3% NaCl solution, the average NaCl equilibrium concentrations ranged from 37.3% to 42.5% ± 2.1% for radish, 9.1% to 11.3% ± 0.3% for potato, and 4.8% to 5.6% ± 0.2% for salmon. This trend could be explained by moisture content differences between the food samples. At equilibrium, radish had the greatest average moisture content, ranging from 95.1% to 97.2% ± 0.3% wb followed by potato (82.7% to 86.4% ± 1.1% wb) and salmon (66.7% to 69.3% ± 0.8% wb). Differences in salt content within the 3 foods were amplified by expressing the NaCl concentration on a dry basis because the 3 foods all had different moisture contents.

**Equilibrium distribution coefficient.** All average equilibrium distribution (partition) coefficients ranged between 0.58 and 0.76, which means that the NaCl diffusion process did not eliminate the concentration gradient between the food and solution (Figure 4). The equilibrium distribution coefficients for potato are comparable with those determined by Wang and Sastry (1993) for fresh white potato at 25 °C (0.41 to 0.74).

The average equilibrium distribution coefficient for radish was greater than potato and salmon, indicating that a greater amount of NaCl diffused into the radish (Figure 4). On a molecular level, this trend indicates that more Na$^+$ and Cl$^-$ ions entered the radish before saturation was reached, which led to a larger equilibrium concentration and equilibrium distribution coefficient. Radish also had the greatest moisture content among the 3 foods and this might have facilitated a greater amount of NaCl diffusion into the radish samples. Salmon samples generally had smaller NaCl equilibrium concentrations and equilibrium distribution coefficients, which can be explained in-part by the lower moisture content and greater amount of lipids than in radish or potato. Salmon has a higher concentration of lipids, which are hydrophobic. The lipid content of a food has been shown to slow the diffusion of water soluble, hydrophilic ions (for example, Na$^+$ and Cl$^-$) into the food (Gallart-Jornet and others 2007a; Alizadeh and others 2009; Cierach and Modzelewska-Kapitula 2011).

Neither the salt solution concentration nor temperature significantly impacted the equilibrium distribution coefficient. This implies that for a given food, the equilibrium amount of NaCl in the food relative to the amount in the liquid was similar for all temperatures. These results agree with Sarang and Sastry (2007), who concluded that the equilibrium distribution coefficients for water chestnut in 5% to 10% NaCl solutions at 25 to 80 °C was not significantly influenced by temperature or salt concentration.

**Analytical diffusion model results**

Average sodium chloride content in the food during heating was predicted using the analytical diffusion model for wet basis (Figure 2) and dry basis (Figure 3) concentration data. The analytical diffusion model matched the trend of the experimental data, as the predicted NaCl concentration in the food increased with increasing time until the equilibrium concentration was achieved. The analytical diffusion model fit well to both the wet and dry basis concentration data, with an average $R^2$ among all treatments of 0.94 and 0.99, respectively.

Effective diffusion coefficients were determined using the analytical diffusion model and ranged from 0.7 × 10$^{-8}$ to 2.6 × 10$^{-8}$ m²/s for the wet basis concentration data (Table 2) and 0.2 × 10$^{-8}$ to 2.6 × 10$^{-8}$ m²/s for the dry basis concentration data (data not shown). If the moisture content was not significantly affected by heating time, changing the input data from wet to dry basis was simply a transformation or unit conversion and did not change the calculated rate of diffusion. The use of dry basis concentration data resulted in lower effective diffusion coefficients compared to the wet basis data for radish because the radish moisture content increased with increasing heating time. Dry basis concentration data and models also showed the differences between foods much more clearly than wet basis.

For all temperatures and salt solutions, salmon had the largest effective diffusion coefficients (db), followed by potato and radish. Salmon samples had faster rates of diffusion, but smaller equilibrium salt concentrations; in other words, the salmon samples reached a lower equilibrium NaCl concentration faster. The faster diffusion rate in the salmon could be explained partly by the fact that the samples were purchased frozen and thawed before the experiments and contain a higher amount of protein that was interacting with the salt. Frozen, thawed salmon was used in this study because the results are more applicable to the food industry in which most manufacturers use individual quick frozen (IQF) fish and meats in preparing ready-to-eat meals and food products. Frozen, thawed Atlantic salmon has been shown to have a larger effective NaCl diffusion coefficient compared to unfrozen samples due to the structure change (Alizadeh and others 2009; Aursand and others 2009). The addition of salt to high protein foods has been shown to influence the water-holding capacity and solubility of proteins (Albarracín and others 2011; McDonnell and others 2013). Protein solubility is affected by ions in solution (for example, Na$^+$ and Cl$^-$) and at low salt concentrations, such as those
used in this study, protein solubility increases due to a decrease in electrostatic interactions (salting-in) (Albarracin and others 2011).

ArRHENius relationship for effective diffusion coefficients

The Arrhenius equation was used to describe the influence of temperature on the wet and dry basis effective diffusion coefficients (Figure 5). Similar to the concentration profiles discussed previously, the use of dry basis data in determining the Arrhenius relationship facilitated differentiation between foods.

Arrhenius equations fit well to potato and radish wet and dry basis effective diffusion coefficient data with coefficient of determinations above 0.93 (Figure 5). However, Arrhenius equations did not fit well to the salmon data, with much lower R² values of 0.70 and 0.65 for 1% and 3% NaCl solutions, respectively (wet basis data). This indicates that the effect of temperature on the effective diffusion coefficient for salmon did not follow an Arrhenius relationship, which could be attributed to multiple processes occurring simultaneously. These processes, such as protein denaturation and precipitation, may have led to structural changes that affected the NaCl diffusion rate.

Reference temperature constants and activation energies for potato and radish in 1% salt solution were determined using wet basis effective diffusion coefficients (Table 3) and dry basis effective diffusion coefficients (data not shown). Reference temperature constants and activation energies for salmon samples were not included in Table 3 due to an unsatisfactory fit of the Arrhenius equation to the salmon data (R² less than 0.75), as they could result in inaccurate values and incorrect interpretations.

Radish samples in 1% NaCl solution had a significantly greater activation energy and reference temperature constant compared to potato in 1% and 3% NaCl solutions and radish in 3% NaCl solution (Table 3). The larger activation energy suggests that the rate of NaCl diffusion in radish in 1% salt solution was more sensitive to changes in temperature than the other treatment conditions. The larger reference temperature constant suggests that as the temperature approaches infinity, the effective diffusion coefficient for radish in 1% salt solution would be larger than the other treatment conditions.

Numerical diffusion model results

The numerical diffusion model was developed to simulate both heat and mass transfer simultaneously. The relative rates of heat transfer and salt diffusion were examined by calculating the response time of dimensionless temperatures and concentrations for potato in a 1% NaCl solution at 121 °C. Results showed that heat transfer was a much faster process than mass transfer. The response time, or time it takes for a dimensionless parameter to change by 63.2% of the initial value, was 2.5 and 11 min for the dimensionless temperature and concentration, respectively. This result was expected because diffusion is slow compared to most heat or momentum transfer processes. During high temperature (121 °C) processing, the amount of diffusion that occurred during heating & storage . . .

Table 2–Effective diffusion coefficients (D eff) with estimated 95% confidence intervals and coefficients of determination (R²) from the analytical diffusion model using wet basis NaCl concentration data.

<table>
<thead>
<tr>
<th>NaCl solution (% w/w)</th>
<th>Temperature (°C)</th>
<th>D eff (10⁻⁸ m²/s)</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Potato</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>90</td>
<td>0.9 ± 0.3</td>
<td>0.77</td>
</tr>
<tr>
<td></td>
<td>105</td>
<td>1.5 ± 0.3</td>
<td>0.91</td>
</tr>
<tr>
<td></td>
<td>121</td>
<td>1.8 ± 0.3</td>
<td>0.98</td>
</tr>
<tr>
<td>3</td>
<td>90</td>
<td>0.8 ± 0.2</td>
<td>0.99</td>
</tr>
<tr>
<td></td>
<td>105</td>
<td>1.4 ± 0.2</td>
<td>0.99</td>
</tr>
<tr>
<td></td>
<td>121</td>
<td>1.8 ± 0.3</td>
<td>0.99</td>
</tr>
<tr>
<td>Radish</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>83</td>
<td>0.93</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>155</td>
<td>0.99</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>13300</td>
<td>0.99</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>76</td>
<td>0.94</td>
</tr>
<tr>
<td>Salmon</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>83</td>
<td>0.93</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>155</td>
<td>0.99</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>13300</td>
<td>0.99</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>76</td>
<td>0.94</td>
</tr>
</tbody>
</table>

Table 3–Reference temperature constants (D₀), activation energies (Ea), and coefficients of determination (R²) for NaCl diffusion in the temperature range of 90 to 121 °C on a wet basis. Salmon was excluded because of an unsatisfactory fit of the Arrhenius equation.

<table>
<thead>
<tr>
<th>Food</th>
<th>NaCl Solution (% w/w)</th>
<th>D₀ (10⁻⁸ m²/s)</th>
<th>Eₐ (kJ/mol)</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Potato</td>
<td>1</td>
<td>83</td>
<td>27.5</td>
<td>0.93</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>155</td>
<td>29.6</td>
<td>0.99</td>
</tr>
<tr>
<td>Radish</td>
<td>1</td>
<td>13300</td>
<td>43.7</td>
<td>0.99</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>76</td>
<td>27.3</td>
<td>0.94</td>
</tr>
</tbody>
</table>

Figure 5–Effective NaCl diffusion coefficients from wet basis (A) and dry basis (B) data for salmon (●), potato (▲), and radish (●) in 1% NaCl solution and salmon (●), potato (●), and radish (●) in 3% NaCl solution. The Arrhenius relationships are shown for each food in 1% and 3% NaCl solutions (●●●●).
the temperature come-up time was relatively small, which implied that a constant D-value assumption was appropriate.

The predicted salt concentration (mass average) was determined from the numerical model for potato, radish, and salmon in 1% NaCl solution heated at 121 °C from 0 to 120 min; these predictions were compared to the analytical diffusion model and experimental results (Figure 6). From the numerical model, the effective diffusion coefficient for potato ($0.60 \times 10^{-8}$ m$^2$/s) was the largest, followed by radish ($0.45 \times 10^{-8}$ m$^2$/s), and salmon ($0.30 \times 10^{-8}$ m$^2$/s). The equilibrium distribution coefficients from the numerical solution for potato, radish, and salmon were 0.64, 0.68, and 0.60, respectively. The numerical solutions fit better than the analytical solutions for potato, radish, and salmon, with $R^2$ values of 0.99, 0.98, and 0.97 for the numerical solutions and 0.98,
0.97, and 0.90 for the analytical solutions, respectively. The superior fit of the numerical diffusion model could be due to the more sophisticated modeling approach compared to the analytical diffusion model. The numerical model had coupled heat and mass transfer, which was not present in the analytical model. Both the effective diffusion coefficient for NaCl in food and the NaCl equilibrium distribution coefficient were optimized to find the best fit numerical solution. This is a different approach than used in the analytical model, where only the effective diffusion coefficient was optimized and the equilibrium distribution coefficient was constant (experimentally determined value).

Effective diffusion coefficients determined using the numerical solution were smaller than those determined using the analytical solution and closer to $D_{eff}$ values reported in previous diffusion research. For example, Liu (1992) reported an effective diffusion coefficient (wet basis) for potato in 3% NaCl solution at 120 °C of $0.42 \times 10^{-6}$ m²/s. Effective diffusion coefficients for fresh radish determined by Hashiba and others (2007) using dual mode diffusion for fresh Japanese radish in 3% NaCl solution at 98 °C ranged from $0.18 \times 10^{-8}$ to $0.31 \times 10^{-8}$ m²/s, which was smaller than those found in this study, likely because of the lower heating temperature. Previous work on salt diffusion in salmon was conducted at low temperatures, at or below 10 °C.

The COMSOL numerical simulation can also be employed to visualize the distribution of NaCl within the food and liquid samples over time. The numerical solution was used to generate pictorial representations of the NaCl concentration after 5, 10, 30, 60, and 120 min of heating in 1% NaCl at 121 °C (Figure 7). At all of the time points, radish samples had the largest average NaCl concentration, followed by potato, and salmon samples. These results are helpful in understanding how much salt diffusion has occurred after different amounts of time during thermal processing. It is especially interesting to compare the salt diffusion that would occur during a shorter sterilization process (for example, MATS, approximately 10 min) and a longer sterilization process (for example, retort, approximately 60 min). After 10 min of heating, the NaCl concentration inside the food was not uniform and became more uniform after 60 min of heating at 121 °C. This implies that at the end of a short process, such as MATS, the system is likely not at steady state and salt would continue to diffuse during storage. This finding agrees with the results from this study for diffusion during storage.

Diffusion during storage

NaCl concentrations in the food after thermal processing during storage are summarized in Figure 8. Similar to the results for salt diffusion during heating, throughout the storage radish had the highest NaCl concentration, followed by potato and salmon. Immediately after heating (0 d storage) potato and radish heated for 60 min at 121 °C had the greatest salt concentration, followed by 60 min at 90 °C, 10 min at 121 °C, and 10 min at 90 °C with the smallest concentration ($P < 0.001$). Immediately after heating (0 d storage) salmon samples heated for 60 min at 121 °C or 60 min at 90 °C had the greatest NaCl concentrations, followed by those heated for 10 min at 121 °C or 10 min at 90 °C ($P < 0.0001$). The NaCl concentration in the liquid during the storage study ranged from 2.4% to 2.8% for all samples. Thus, the NaCl concentration in the liquid was always greater than the concentration in the food samples, which further supports the use of the distribution (partition) coefficient in this study.

The NaCl concentration in the food had not reached equilibrium after 10 or 60 min of heating at 90 or 121 °C and salt diffusion continued during storage (Figure 8). After 14 d of storage, potato (1.57 ± 0.03% NaCl wb) and radish (1.81% ± 0.04% NaCl wb) samples did not have significantly different NaCl concentrations, meaning that heating time and temperature had no significant effect on the concentration ($P > 0.05$). After 7 d of storage, results for the salmon samples (1.49% ± 0.04% NaCl wb) showed the same trend that heating time and temperature had no significant effect on the NaCl concentration ($P > 0.05$). The results imply that it would be advisable to wait 14 d after processing before doing sensory evaluation to allow the salt to equilibrate within multiphase, high moisture, low-acid thermally processed products.

This study suggests that a shorter processing time (for example, 10 min) led to less NaCl diffusion during the processing compared to a longer time (for example, 60 min), but during storage most samples equilibrated to a similar NaCl concentration regardless of the thermal process severity. For example, the percent change in potato NaCl concentration from day 0 to 28 was 126, 63, 23, –2%.

![Figure 8–Average solid NaCl concentration (n = 3, wet basis) with 95% confidence intervals for potato (A), radish (B), and salmon (C) samples heated at 90 °C for 10 min (–) or 60 min (–) and stored at 4 °C and samples heated at 121 °C for 10 min (–) or 60 min (–) and stored at 22 °C.](image-url)
for samples heated for 10 min at 90 °C, 10 min at 121 °C, 60 min at 90 °C, and 60 min at 121 °C, respectively. These results can be applied to salt diffusion during storage after thermal processing of MATS, (approximately 10 min at 121 °C), conventional retort thermal sterilization (approximately 60 min at 121 °C) and MAPS or conventional pasteurization (approximately 10 min at 90 °C).

Conclusions

The NaCl content of potato, radish, and salmon samples in salt solutions increased with increasing heating time until an equilibrium concentration was reached. For each temperature and NaCl solution, the equilibrium NaCl concentration (db) and equilibrium concentration was reached. For each temperature and NaCl concentration on Atlantic salmon fillet salting. J Food Eng 80(1):267–75.


References

