Heat and Mass Transfer Modeling of Part-Baked Sangak Bread Packaged in Modified Atmosphere

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Abstract—*Modified atmosphere packaging is a dynamic system that the inevitable heat and mass transfer phenomena occur during their storage. In order to study changes in a MAP system, heat conduction and convection, gas solution and diffusion, gas permeation in the polymeric film, evaporation and condensation phenomena were modeled. The model validated against experimental data of part-baked Sangak bread packaged in 100% CO_2. The high coefficient of correlation and low root mean square error between experimental and predicted data confirmed the developed mathematical model and its numerical solution. Results showed that the storage temperature has an important effect on the headspace gas composition and volume of package. In this study, because of the important effect of CO_2 solubility on equilibrium gas composition, CO_2 solubility for part-baked Sangak bread was measured experimentally. This model is a suitable tool for optimizing the MAP of non-respiring products packed in flexible plastic packages.*

Keywords-Modified atmospheres packaging; Mathematical model; Heat and mass transfer; Part-baked Sangak bread

I. INTRODUCTION

In recent years, there has been increasing demand for minimally processed and near-fresh quality products. Baked-Off Technology (BOT) not only reduces economic losses due to staling, but also provides the possibility of fresh bread for consumers at any time [1]. However, the shelf life of part-baked products is very short and it is a problem to maintain their quality during prolonged storage [2]. Modified atmosphere packaging (MAP), is packaging of a perishable product under a different composition gas than air, which can increase product shelf life while maintaining its quality [3], [4]. The most important gas which is used in MAP is carbon dioxide, because it is a non-toxic gas that its anti-microbial effects well known [5]. Also commonly nitrogen is used in MAP in combination with carbon dioxide. N2 is an inert gas, without taste and without any anti-microbial activity that mainly is used for replacement with oxygen and to prevent package collapse [3].

Maintaining an optimum internal atmosphere in package is the main factor in quality preservation of products in MAP [6]. Mathematical modeling of MAP systems is a suitable tool for identifying and describing the interactions between food, head space and ambient. So it can predict behavior of these phenomena under hypothetical conditions. Therefore, using mathematical models, optimization system with maximum quality and shelf life will be possible without requiring costly and time-consuming tests.

So far, many models have been proposed in MAP respiring products [7]–[10]. The main mechanisms of gas exchange in these cases, is the respiration product and gas transfer through the packaging material [3], [11]. Despite this, modeling studies in non-respiring products is limited [11]–[14]. The most important mass transfer phenomena in these products are gas absorption in food and gas permeability of package [5]. In addition, storage temperature is an effective parameter on quality stability of product packaged in MAP. So that the reactions such as respiration rate of product, gas solubility, permeability of packaging material, the growth of microorganisms and product moisture phase change are affected by the storage temperature. Therefore, due to the inevitable temperature fluctuations that occur during transport and storage, heat transfer modeling is essential to estimate the product temperature as a function of time and place [15], [16]. Sangak bread is one of the most widely used flat bread in Iran that due to preparation from whole wheat flour has a high nutritional value. The objective of this study was to develop and validate a mathematical model to describe heat and mass transfer phenomena in part-baked Sangak bread packed under modified atmosphere.

II. MATHEMATICAL MODEL

A. Model assumptions

The package containing part-baked Sangak bread is modeled as an infinite slab and the cross-section of the infinite slab is divided into a fixed grid system.

To simplify the modeling, the following assumptions were made in this work:

- The food and package were considered as an infinite slab. Thus heat and mass transfer were done one-dimensional.
- Concentration of gas in the head space was considered homogeneously and without convective flux.
- Gas exchange mechanisms are gas absorption by food, permeability through the packaging film, water evaporation from food surface and condensation within the package. Changes of gas composition due to microbial respiration were neglected.
- Heat transfer between package and air and also in the head space occurs through convection and in the food occurs through conduction.
- Thermophysical properties of food were assumed constant during storage.

Nomenclature and units used for modeling are given in table 1.

B. Governing equations

- *Heat balance equation:*

Heat transfer in the part–baked Sangak bread was described by second Fourier's law:

At 0<x<L, $$\rho_F Cp_F \frac{\partial T}{\partial t} = k_F \frac{\partial^2 T}{\partial x^2} \quad (1)$$

For describing heat transfer in boundaries of part-baked bread, the following equations were applied:

At x=0, $$\frac{\partial \mathrm{T}}{\partial \mathrm{x}} = 0 \quad (2)$$

At x=L, $$k_F A_{FS} \frac{\partial T_{FS}}{\partial x} = \mathrm{h_H}(\mathrm{T_H} - \mathrm{T_{FS}}) + \lambda \dot{m}_e \quad (3)$$

TABLE I. NOMENCLATURE

Symbol	Quantity	Unit	Symbol	Quantity	Unit
a	Klein constant	dimensionless	V	Volume (m^3)	m
A	Area	m^2	W	Mass (kg)	
a_w	Water activity,	dimensionless	X	Moisture fraction	kg water/kg dry material
C	Gas concentration in food or Water concentration in food	kmol/kg or kgW/kgDM	x	Distance in the x axis	m
c	Gas concentration in headspace	kmol/kmol	y	Molar volumetric fraction	kmol/kmol
cp	Specific heat	J/kg/K	λ	Latent heat of evaporation	J/kg
D	Diffusion coefficient	m^2/s	ρ	Density	kg/m^3
h	Heat convection coefficient	$W/m^2/K$	ρ_s	Density of dry matter	kg/m3
$\bar{H}$	Specific enthalpy	J/kg	φ	Permeability	kmol/m s Pa
k	Thermal conductivity	W/m/K	***Subscripts***		
k_g	Mass convection coefficient	$kmol/m^2/s/Pa$	*0*	Initial	
L	Thickness	m	*a*	Air	
$\dot{m}_{con}$	Condensation rate	kg/s	*aq*	aqueous solution	
$\dot{m}_e$	Evaporation rate	kg/s	*F*	Food	
$\dot{m}_P$	Permeation rate for H_2O	kmol/s	*FS*	Food surface	
Mw	Molecular weight	kg/kmol	*H*	Headspace	
n	Number of moles	kmol	*P*	Package	
P	Pressure	Pa	*sat*	Saturated vapor	
p	Partial pressure	Pa	*exp*	Experimented	
R	Ideal gas constant	Pa m^3/kmol K	*pre*	Predicted	
S	Solubility	kmol/kgPa	CO_2	Carbon dioxide	
T	Temperature	K	O_2	Oxygen	
t	Time	s	N_2	Nitrogen	
v	Partial volume	m^3	H_2O	Water	

Equation (4) estimates temperature evolution in headspace. It considers convective heat transfer between package and headspace, food surface and headspace, energy terms associated to water evaporation from food surface, condensation on the inner surface of package and water vapor permeation through the package. Energy related to gas transport was ignored because of their low specific enthalpy and low permeability of film for gases.

$$\frac{\partial T_H}{\partial t} = \frac{RT[h_H A_P(T_P - T_H) - h_H A_{FS}(T_H - T_{FS}) + \dot{m}_e \bar{H}_{H20} - \dot{m}_{con} \bar{H}_{H20} - \dot{m}_P \bar{H}_{H2O}]}{P[(cpMwv)CO_2 + (cpMwv)O_2 + (cpMwv)N_2 + (cpMwv)H_2 0]} \quad (4)$$

Package temperature was estimated by considering convective heat transfer in outer and inner surface of package, energy terms related to condensation on inner surface of package and water vapor permeation through the package.

$$\frac{\partial T_P}{\partial t} = \frac{h_a(T_a - T_P) - h_H(T_P - T_H) + \lambda \dot{m}_{con} + \dot{m}_P \bar{H}_{H2O}}{(cp\rho x)_P} \quad (5)$$

$$\dot{m}_P = \left(\frac{\varphi_{H2O} A}{l}\right)_P (p_{H(H2O)} - p_a) M_{W_{H_2 0}} \quad (6)$$

Temperature was supposed constant and uniform in the package containing bread at time zero.

$$T = T_0 \quad (7)$$

Mass balance equation:

Fick's second law was applied to describe mass transfer in the part–baked Sangak bread.

At 0<x< L, $\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$ (8)

For describing mass transfer in boundaries of part-baked bread, the following equations were applied:

At x=0 $\frac{\partial C}{\partial x} = 0$ (9)

At x=L $C = Sp_H$ (10)

$$D \frac{\partial C_{H_2O}}{\partial x} = k_g M_W (p_{H(H2O)} - p_{\mathrm{sat,T_{FS}}} a_w) \frac{1}{\rho_s}$$

Gas absorption by the food was obtained by calculating of gas concentration in the bread and using the following relation:

$$[\frac{\partial n}{\partial t}]_F = W_F [\frac{\partial \bar{C}}{\partial t}] \quad (11)$$

Evaporation at the product surface was estimated by the following expression:

$$[\frac{\partial n_{H2O}}{\partial t}]_F = k_g A_{FS}[p_{H(H2O)} - p_{sat,T_{FS}} a_w] \quad (12)$$

Because of near-saturation conditions within the package and fluctuations in ambient temperatures, condensation on the inside of the package film was estimated by the following correlation:

$$\left[\frac{\partial n_{H2O}}{\partial t}\right]_{con} = k_g A_P\left(p_{H(H20)} - p_{sat.T_P}\right) \quad (13)$$

Gas transfer through the package due to permeability of the package film to gases was estimated by:

$$[\frac{\partial n}{\partial t}]_P = \left[\frac{\varphi A}{l}\right]_P (p_H - p_a) \quad (14)$$

A mass balance was written for calculating concentration of each component (CO_2, O_2, N_2 and water vapor) in headspace. Because of flexibility of package film, ideal gas law with considering variable volume and constant pressure was used to estimate the number of gases moles in headspace. Thus, the evolution of gas and water vapor concentrations in headspace was expressed by "(15) and (16)", respectively:

$$-\left[\frac{\partial n_i}{\partial t}\right]_P - \left[\frac{\partial n_i}{\partial t}\right]_F = \frac{P}{R}[\frac{1}{T_H}\frac{\partial v_i}{\partial t} + v_i\frac{\partial[\frac{1}{T_H}]}{\partial t}] \quad (15)$$

$$-\left[\frac{\partial n_{H20}}{\partial t}\right]_P - \left[\frac{\partial n_{H20}}{\partial t}\right]_F - \left[\frac{\partial n_{H20}}{\partial t}\right]_{con} = \frac{P}{R}[\frac{1}{T_H}\frac{\partial v_{H2O}}{\partial t} + v_{H2O}\frac{\partial[\frac{1}{T_H}]}{\partial t}] \quad (16)$$

Finally, headspace volume is calculated by "(17)" and then gas composition headspace was estimated by "(18)";

$$V_H = v_{CO2} + v_{O2} + v_{N2} + v_{H20} \quad (17)$$

$$c_i = \frac{v_i}{V_H} \quad (18)$$

Concentration was supposed constant and uniform in the package containing bread at time zero.

$$0 \leq x \leq L \qquad C = Sp_a \qquad C = C_{0_{H2o}} \quad (19)$$

C. Numerical method

A finite difference method was used to solve the heat and mass transfer equations [17]. Then, the mathematical model developed was implemented using a computer program written in MATLAB (version 7.1).

D. Model parameters

Film permeability for CO_2, O_2 and N_2 at 25 °C was measured by using a gas permeability tester (Brugger, GDP-C, Germany). A water vapor permeability tester was used for determining permeability of water vapor on film (Brugger, WDDG, Germany). Table 2 shows these parameters.

TABLE II. Film permeability for CO_2, O_2, N_2 and H_2O at 25°C (kmol/m s Pa).

	CO_2	O_2	N_2	H_2O
Film permeability	9.333×10^{-21}	1.309×10^{-20}	7.536×10^{-21}	6.047×10^{-19}

Thermophysical properties of the part-baked Sangak bread have been shown in table 3. Density was determined at 25°C using the rapeseed displacement [18]. Water activity was measured by a water activity meter (Novasina ms1-aw, Axair Ltd., Switzerland). Specific heat was calculated based on a parallel model and heat conductivity was calculated with krisher model [19].

TABLE III. Physical properties of the part-baked Sangak bread.

	ρ (kg/m³)	a_w	*cp* (J/kg K)	*k* (W/m K)
25 °C	0.495	0.91	3231.506	0.275
50 °C	0.462	0.94	3258. 065	0.292

CO_2 solubility in part-baked Sangak bread was determined by using the data of changes of headspace volume, as described in detail by [20].

Gas solubility and diffusivity in part-baked Sangak bread were determined from values in water and by using the following equations [21].

$$S = S_{aq}(1 - X) \quad (20)$$

$$D = D_{aq}\, exp(-ay) \quad (21)$$

Water vapor diffusivity in part-baked Sangak bread was estimated by using a model for predicting the mass diffusivity (Ozisik, 1985). Tables 4 and 5 shows parameters utilized.

TABLE IV. Solubility for CO_2, O_2 and N_2 in the part-baked Sangak bread (kmol/kg Pa).

	CO_2 [a]	CO_2 [b]	O_2 [b]	N_2 [b]
25 °C	1.973×10^{-10}	1.747×10^{-10}	6.543×10^{-12}	3.184×10^{-12}
50 °C	-	1.135×10^{-10}	4.014×10^{-12}	2.847×10^{-12}

[a] measured value
[b] estimated value

TABLE V. Diffusivity for CO_2, O_2, N_2 and H_2O in the part-baked Sangak bread (m²/s).

	CO_2	O_2	N_2	H_2O
25 °C	1.686×10^{-10}	2.032×10^{-10}	1.601×10^{-10}	3.803×10^{-10}
50 °C	2.652×10^{-10}	3.831×10^{-10}	3.123×10^{-10}	4.463×10^{-10}

The convective heat and mass transfer coefficients on package surface and headspace were measured by using the transient temperature measurement method. For this purpose, an aluminum block with dimensions similar to package was placed in the incubator at 25°C and also cabinet drier at 50°C with 1m/s air velocity. Surface and center temperatures of aluminum block were measured with thermocouples.

The convective heat transfer coefficient on headspace was determined by means of packaging an aluminum block with dimensions of 15×15×1 cm in a bag plastic the same part-baked breads. The packages were placed in incubator and drier with 25 and 50°C temperature respectively. Center of aluminum block and headspace temperatures were measured by thermocouples.

The convective mass transfer coefficient on headspace was obtained from correlation between heat and mass transfer coefficient and using Chilton – Colburn analogy. These results have been shown in table 6.

TABLE VI. Convective heat and mass transfer coefficients.

	25°C	50°C
h_a (W/m² K)	11.863	13.444
h_H (W/m² K)	6.101	9.377

E. Statistical analysis

The model validation was done by comparing the values predicted with experimental results. The comparison criteria used to evaluate goodness of fit, were the coefficient of correlation (*r*) and the root mean square error (*RMSE*).

$$RMSE = [\frac{1}{N}\sum_{i=1}^{N}(\exp_i - pre_i)^2]^{\frac{1}{2}} \qquad (22)$$

III. EXPERIMENTAL METHODOLOGY

A. Bread making and packaging

Whole wheat flour containing 11.037% protein, 1.32% ash, 14% water and 1.55% lipid was used for production of Sangak Bread (Baharestan Co., Isfahan, Iran). The formula consisted of 100 g of flour, 100 g of water, 1 g of salt and 1 g of dried active yeast (Kelarmaye Co., Shahrekord, Iran). All ingredients were mixed with a stand mixer (HOBART mixer, model C-100, USA) for 15 min, and then dough was fermented at 30° C for 60 min and 75-85% relative humidity in a fermentation chamber. Next, prepared dough was cut in 120 g pieces and was sheeted to 5 mm thickness. Part baking was done in a forced convection oven (BOSCH, HBA73B550, Germany) at 250°C for 6 min. Part-baked Sangak bread samples were cooled down at room temperature to 25° C and were cut with dimensions of 15×15×0.8 cm. Each loaf had average weight of 109.46 g and volume of 221.94 cm^3. Each sample was packaged in a plastic bag PA/PE with dimensions of 20×25 cm and 100µm thickness (Nadi Plastic Arya, Tehran, Iran) with 80.33 cm^3 of CO_2 (98.5% CO_2, 1.17 N_2 and 0.31 O_2) by a vacuum packaging machine (HENKOVAC, E-153, Netherlands). All samples were stored at 25° C for 21 days in an incubator (Wisecube, WIG-105, Korea).

B. Temperature profile

Package, headspace and product (surface and center) temperatures were measured with K-type thermocouples with 0.3 mm diameter (Omega, Stamford, USA) during heating. Heating was carried out in a forced convection incubator (Trans-Nik, Iran) at 50°C with an air flowed perpendicular to the package surface at a velocity of 1 m/s. The temperature data were recorded using a data logger (Delta-T, DL2e, U.K.).

C. Gas composition and relative humidity in headspace

The evolution of gas composition in headspace was analyzed by a gas analyzer (model 6.0, Oxybaby, Germany) during storage of packaged samples at 25°C. The headspace relative humidity was logged throughout the experiment with a hygrometer (Hygrolog, Rotronic, Swiss).

D. Moisture content of part-baked Sangak bread

Moisture content of part-baked Sangak bread samples was evaluated during storage according to the AACC method 44-15A with an air oven (UNB400, Memmert, Germany).

E. Headspace volume

Headspace volume was measured by submerging the package under water and determining the resultant force of gravity and buoyancy forces using a UTM (Instron1140, Instron Ltd., High Wycombe, U.K.) at 0, 1, 2, 4, 6 and 8 hours. Immersing probe was made of stainless steel, 120 mm high and 130 mm wide. The container used for immersion had volume of 6.5 liter. Submerging was done at a rate of 100 mm/min. Initial headspace volume was determined by packaging with 100% N_2 [20].

IV. RESULT AND DISCUSSION

A. Heat transfer

Fig. 1 shows changes in package, headspace and product (surface and center) temperature of a MA-packed part-baked Sangak bread during heating at 50°C with air velocity of 1m/s. Large temperature difference between package and food temperatures is because of the gas layer in the headspace.This layer due to low convection and conduction heat transfer works like a thermal insulator between the package and the product. This effect may be an advantage or a disadvantage depending on the initial temperature of the products. Also, it is observed that the temperature difference between the center and surface temperature of the part-baked Sangak bread is very low. Low thickness of the product and small Biot number leads to this negligible temperature difference [22]. Fig. 2 compares the measured and predicted temperatures. Coefficients of correlation (r) and the root mean square error (RMSE) between experimental and predicted values for package were 0.946 and 1.948°C, respectively. For headspace were 0.992 and 0.982°C and for center of product were 0.998 and 0.453°C, respectively. So, the developed heat transfer model can predict the temperature in a MAP system the change storage temperature.

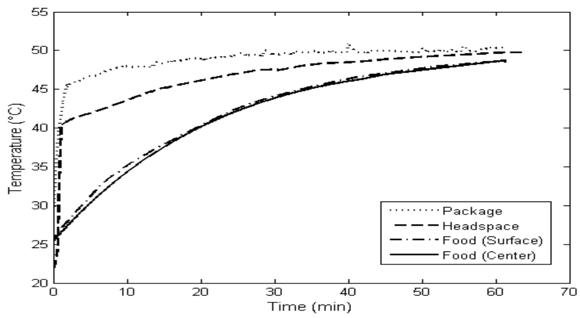

Figure 1. Experimental value of package, headspace and food temperatures during heating at 50°C.

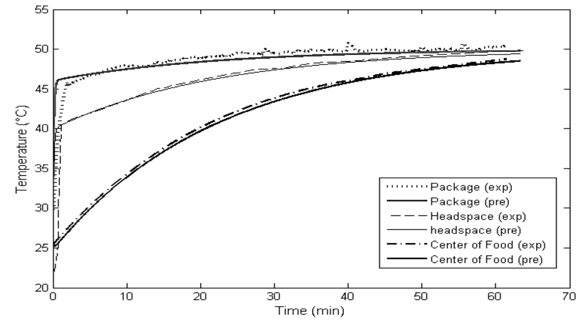

Figure 2. Measured and predicted temperatures during heating at 50°C.

B. Mass transfer

Fig. 3 shows the changes in CO_2 concentration in the headspace during storage at 25°C. Reduction in CO_2 concentration occurs in two stages; in the early hours of the storage it occurs faster due to the gas solubility in aqueous phase of part-baked bread and headspace reaches an equilibriumCO_2 concentration. In long-term storage, permeability of packaging film leads to reduced concentration slightly. *r* and *RMSE* between experimental and predicted values were 0.947 and 2.42% respectively. By increasing in storage time (especially after 15 days), it causes difference between experimental and predicted values due to microorganism's respiration.

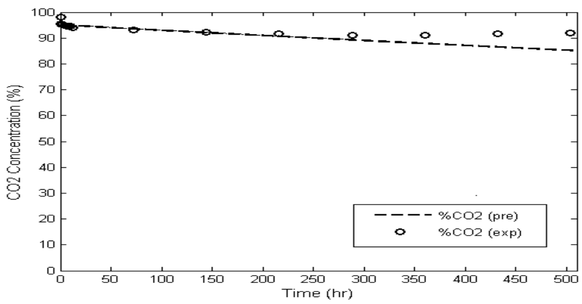

Figure 3. Changes in headspace CO_2 concentration during storage time.

Fig. 4 shows the changes in concentration of oxygen in the headspace during storage. Increasing oxygen concentration in the early hours of storage time occurs due to air present in the porous texture of bread. In addition, influx of O_2 through the package will increase its concentration during 21 days slowly. *r* and *RMSE* between experimental and predicted values were 0.873 and 0.262% respectively. Relatively high error value is probably due to low oxygen level in the package and is related to measurement error. Also O_2 consumption by the microorganisms has increased difference between measured and predicted values at the final days of storage period.

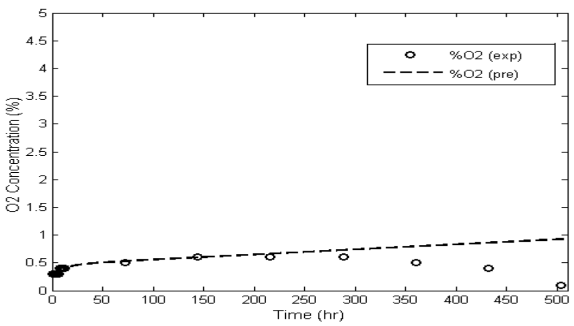

Figure 4. Changes in headspace O_2 concentration during storage time.

Fig. 5 shows that water activity of product and relative humidity in headspaces reaches equilibrium after 2 hours at 25°C. Because of high water activity in part-baked Sangak bread, a high equilibrium relative humidity is obtained. *r* and *RMSE* between experimental and predicted values were 0.998 and 0.35% respectively.

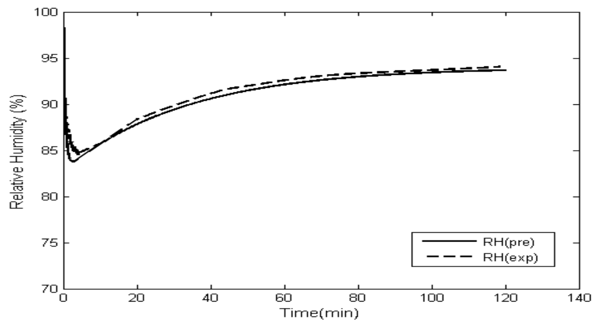

Figure 5. Changes in headspace relative humidity concentration during storage time.

Fig. 6 shows that the bread moisture decreased slowly over the time. It is because of the low permeability of packaging film to water vapor. The results indicate a good agreement between experimental and predicted values. *r* and *RMSE* for these data were 0.998 and 0.0006 kgW/kgDM respectively.

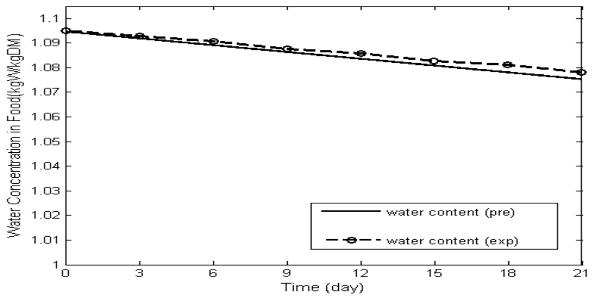

Figure 6. Changes in moisture content of part-baked Sangak bread during storage time.

Fig. 7 shows the changes in volume of the package during storage. Increased volume in the first hour is related to the moisture evaporation from the surface of bread and increase the water vapor partial volume in the headspace. After that, the solubility of gases, specifically carbon dioxide in aqueous phase of product decreases the volume of package until it reaches equilibrium after 6 hours. Using the values of the initial and equilibrium volumes and by applying ideal gas law at constant pressure was calculated dissolved CO_2.Then with Henry´s law CO_2 solubility in part-baked Sangak bread was obtained at 25°C.

r and *RMSE* between experimental and predicted values were 0.970 and 1.102cm^3respectively.

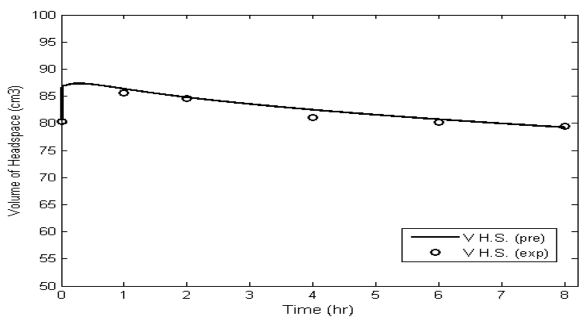

Figure 7. Changes in headspace volume during storage time.

Finally, the results of the validated model showed that the changes in storage temperature will have the significant effect on the headspace gas composition and volume of package, but it has no considerable effect on moisture content of product and relative humidity.

V. CONCLUSION

A mathematical model was developed to estimate heat and mass transfer in part-baked Sangak bread packaged under modified atmosphere and validated against experimental data. The results showed that changes in gas composition were substantially affected by the gas solubility in the food at the beginning of the storage time. From simulation of data, it was observed that changes in storage temperature had a significant effect on the characteristics of system such as the headspace gas composition and volume of package. This revealed the importance of storage temperature fluctuations in the MAP products.

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