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Article

The Effect of Initial Moisture Content on Moisture Migration in a Big Bag of Sugar in the Presence of Cyclic Temperature Changes: Numerical Analysis

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Abstract. The study of moisture migration in a big bag of sugar in which undergoes the fluctuation of temperature as in shipping condition was investigated by a mathematical model. The mathematical model was derived from the conservation of mass and energy, solved by numerical method of lines, validated with the experimental data reported in the previous literature, and used to predict moisture migration for 30 days. It has revealed that the initial moisture content before shipping has the significant effect on the moisture migration. It was found that the large amount of moisture diffuses into the interfacial big bag and the periodical adsorption and desorption takes place when the initial moisture content is high; hence, caking of moist sugar is likely to occur. The result greatly yields useful information for studying caking formation due to moisture migration.

Keywords: Sugar, caking, moisture migration, mathematical modeling.

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1. Introduction

The caking of sugar or the occurrence of large chucks of sugar is currently a serious problem, especially for the exported sugar, since the caked sugar may be rejected at destination ports which would result in an extra cost for waste management of caked sugar. Consequently, it is of significant importance to identify the tendency of caking in sugar in order to avoid this phenomenon during shipping or storage.

The occurrence of caking in sugar during transportation is a result of the fluctuation of temperature. Because of the low heat conductivity of sugar, the fluctuation of ambient temperature results in the temperature gradient within sugar and also the fluctuation of air relative humidity. Then, the water adsorption occurs in a region of low temperature and high relative humidity whereas the water desorption occurs in a region of high temperature and low relative humidity. As the sugar continually adsorbs moisture, the surface of sugar grain is then dissolved, and the liquid will also touch with the other grains - this phenomenon is generally known as the “liquid bridging”. After the evaporation of water in the liquid bridging when the relative humidity of air is lower, sugar in this liquid bridging recrystallizes and solid sugar is formed between adjacent sugar grains - the phenomenon is known as “solid bridging” [1]. This repeated cycle of liquid bridging and liquid bridging is the major cause of caking in sugar.

Since the cause of liquid bridging and solid bridging and, hence, caking is repeated adsorption and desorption and moisture migration within sugar under transportation or storage, several publications have focused on the moisture migration and adsorption and desorption of moisture in sugar. For example, Leaper [2] measured and simulated the moisture migration in a caking box of sugar and also presented some experimental data and an empirical equation of sorption isotherm. They found that the caking box can be applied instead of bulk solids in investigating the moisture migration. Wang [3] simulated moisture migration in a sugar bag or big bag in which the outside temperature changes in cyclic form with only one initial condition. The partial differential equations were discretized using the finite volume and solved numerically by overrelaxed Jocobi and overrelaxed Gauss-Seidal. The high intensity of moisture adsorption and desorption at the interface of the big bag was found. Christakis [4] simulated moisture migration in a small cylinder of sugar in which the top side was exposed to the air of fluctuating relative humidity at the constant temperature. The sharpest changes near the interface were found. In their work, they also took advantage of the moisture migration results to determine the tensile strength which is then used as an indicator to express the intensive of caking. Billings and Paterson [5] simulated and measured the moisture migration in a packed bed of sugar which the temperature difference was applied to both sides by using mathematical model developed by Bronlund and Paterson [6] which was originally developed for lactose .

In this article, the mathematical model describing moisture migration was presented for the case of a big bag of sugar. The model was adopted from the one developed for moisture migration in lactose by Bronlund and Paterson [6]; however, the one dimensional infinite cylinder is applied instead of the one dimensional slab as reported in the original paper since the big bag of sugar would be closer to the cylinder than the slab. The mathematical models were then solved by numerical method of lines that is the effective method for solving the parabolic partial differential equations. In order to include the effect of temperature on adsorption isotherm, the GAB(T) equation [7] was used in the modelling instead of ones previously used in the literature because the sorption isotherm equations previously used in the literature are somewhat debatable. For example, Bronlund and Paterson [6] disregarded the effect of temperature on sorption isotherm although some previous works have reported the strong effect of temperature [2, 8]. The empirical sorption isotherm equation that include the effect of temperature used by Leaper [2], Wang [3], and Christakis [4] strangely yields the non-zero moisture content of sugar when the relative humidity in air is zero. For this reason, the GAB(T) equation is used instead.

The main objective of this article is to study the effect of initial moisture content on moisture migration in the big bag of sugar in the presence of cyclic temperature changes by including the effect of temperature on water sorption isotherm. The amount of initial moisture content that was investigated is divided into two conditions, high moisture (at 0.040% w/w) and low moisture (at 0.015% w/w). These two conditions were chosen based on the real observation reported by a Thai sugar factory that the caking of sugar occurs when the initial moisture content of sugar of 0.040% was shipped to the cold countries but does not occur with the moisture content of 0.015%. The change of temperature during shipping and storage is represented by a square wave form in order to simulate the temperature of night and daytime. The finding in this article would be useful in understanding on caking of sugar since the moisture migration is a main mechanism of liquid bridging, solid bridging and, hence, caking of sugar.

2. Model Development

2.1. Conceptual Model Development

A big bag of sugar with 0.5 meters of radius and 1.20 m in height was a system of interest, and was simplified to be an infinite long cylinder so that the system can be simplified to the one-dimensional in r-direction. A material used to make the big bag is polyethylene which has the very low water permeability; hence, the flux of moisture is presumably zero at the interface of big bag. From conservation of mass and energy, the mathematical models were derived. The phenomena of mass and heat transfer in big bag are shown in Fig. 1. Based on the similar approach to the work of Bronlund and Paterson [6], the assumptions of local thermal equilibrium, local moisture equilibrium, negligible heat convection, and negligible moisture diffusion through particle were applied in this article. As a result, there are only the heat conduction and moisture diffusion that transfer throughout the big bag; heat conduction in both solid and gas phases and the moisture diffusion in gas phase.

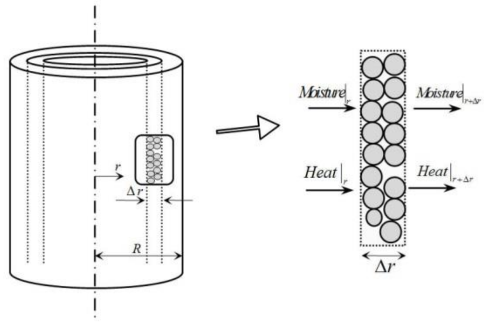

Fig. 1. Schematic diagram of system for shell balance.

2.2. Mathematical Models

The conservation of mass can be written as:

$$\frac{\partial W}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(D_{eff}\, r \frac{\partial}{\partial r}\left(\rho_{da} Y\right)\right) \tag{1}$$

$$W = \varepsilon Y \rho_{da} + \left(1-\varepsilon\right) M \rho_s \tag{2}$$

where W is the total amount of moisture in gas and solid phases in the corresponding shell.

The conservation of energy can be written as:

$$\frac{\partial H}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r D_{eff} \frac{\partial}{\partial r}\left(\rho_{da} Y\right) h_v\right) + \frac{k_{eff}}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) \tag{3}$$

$$H = h_g \varepsilon \rho_{da} + h_s\left(1-\varepsilon\right)\rho_s \tag{4}$$

where H is the total enthalpy of gas and solid phases in the corresponding shell.

2.3. Constitutive Equations

The effective diffusivity can be defined from vapor diffuse through dry air in a porous bed;

$$D_{eff} = \varepsilon D_a \tag{5}$$

where D_a is equal to $1.7255\times10^{-7}\left(T+273.15\right)-2.552\times10^{-5}$ as reported by Shah [9] and ε is equal to 0.365 [4].

By using the parallel pore model, the effective thermal conductivity for bed of sugar can be evaluated from Tsao [10]. The value of thermal conductivity of sugar is 0.208 W m^{-1} K^{-1} [4].

$$k_{eff} = \varepsilon k_a + \left(1-\varepsilon\right)k_s \tag{6}$$

The density of dry air is assumed to be the ideal gas because the pressure and temperature are the ambient condition. Furthermore, the mole fraction of moisture in gas phase is quite small; hence, the density of dry are can be calculated from;

$$\rho_{da} = \frac{273.15}{22.4\left(273.15+T\right)\frac{1}{29}} \tag{7}$$

The enthalpy can be written as shown in the following equations using saturated liquid water at 0°C as the reference state;

$$h_s = C_{ps}T + MC_{pw}T \tag{8}$$

$$h_v = C_{pv}T + h_{fg} \tag{9}$$

$$h_g = C_{pa}T + Yh_v \tag{10}$$

The relationship of absolute humidity and relative humidity at equilibrium can be expressed as shown in Eqs. (11) to (13), which is taken from Rastikian and Capart [11];

$$Y_{eq} = \frac{18}{29} \times \frac{ERH \times P_{sat}}{760 \times 100 - ERH \times P_{sat}} \tag{11}$$

$$ERH = a_{w,eq} \times 100 \tag{12}$$

$$\log_{10} P_{sat} = 7.96681 - \frac{1668.21}{228+T} \tag{13}$$

2.4. Sorption Isotherm

In this article, the GAB(T) equation [7], which is an extension of the Guggenheim-Anderson-de Boer (GAB) model by including the effect of temperature, was used to describe the adsorption isotherm of water. The GAB model is widely used for sorption isotherm of water in foods [7, 12, 13]. The GAB(T) equation is expressed in Eqs. (14) to (17). It should be pointed out that the GAB(T)equation becomes the GAB model when M_0, C and k are constants. In this articles, the parameters of GAB(T) model were determined by fitting the water sorption isotherm reported by Leaper [2].

$$\frac{M_{eq}}{M_0} = \frac{Cka_{w,eq}}{\left(1-ka_{w,eq}\right)\left(1-ka_{w,eq}+Cka_{w,eq}\right)} \tag{14}$$

$$M_0\left(T\right) = M_0^{'} e^{\frac{\Delta H}{(RT)}} \tag{15}$$

$$C\left(T\right) = C^{'} e^{\frac{H_l - H_m}{(RT)}} \tag{16}$$

$$k\left(T\right) = k^{'} e^{\frac{H_l - H_q}{(RT)}} \tag{17}$$

where M_{eq} is moisture content in solid phase in %, $a_{w,eq}$ is water activity, M_0 is the monolayer water content, C is the Guggenheim constant, and k is the constant correcting properties of multilayer molecules with respect to bulk liquid.

2.5. Initial and Boundary Conditions

For initial condition, the thermal and moisture equilibrium of the moist air and sugar are assumed;

$$T\left(r, t=0\right) = T_0 \tag{18}$$

$$Y\left(r, t=0\right) = Y_0 \tag{19}$$

Because the temperature and relative humidity is symmetric at $r = 0$ and no moisture can be diffused through the PE bag, the boundary conditions for conservation of mass are shown in Eqs. (20) and (21). On the other hands, the heat transfer between a PE bag and environment exists; therefore, the boundary conditions for conservation of energy are shown in Eqs. (22) and (23).

$$D_{eff} \left.\frac{\partial\left(Y\rho_{da}\right)}{\partial r}\right|_{r=0} = 0 \tag{20}$$

$$-k_{eff} \left.\frac{\partial T}{\partial r}\right|_{r=0} = 0 \tag{21}$$

$$D_{eff}\left.\frac{\partial(Y\rho_{da})}{\partial r}\right|_{r=R}=0 \tag{22}$$

$$-k_{eff}\left.\frac{\partial T}{\partial r}\right|_{r=R}=h\left(T_{outside}-T_{r=R}\right) \tag{23}$$

where h is heat transfer coefficient and was calculated from the correlation reported by Churchill and Chu [14] for heat transfer by natural convection between vertical wall and ambient as shown in Eq. (24). Nu is the Nusselt number, Ra is the Rayleigh number, and Pr is the Prantdl number.

$$\mathrm{Nu}=\left(0.825+\frac{0.387\mathrm{Ra}^{1/6}}{\left[1+\left(0.492/\mathrm{Pr}\right)^{9/16}\right]^{8/27}}\right)^2 \tag{24}$$

During the shipping and storage, the temperature is usually fluctuating between high and low temperatures as a result of the day time and night time. In order to understand this effect on the moisture migration, the ambient temperature is assumed to be a square-wave function that oscillates between 40 and 10°C every 12 hours with 30 cycles or 30 days as shown as in Fig. 2.

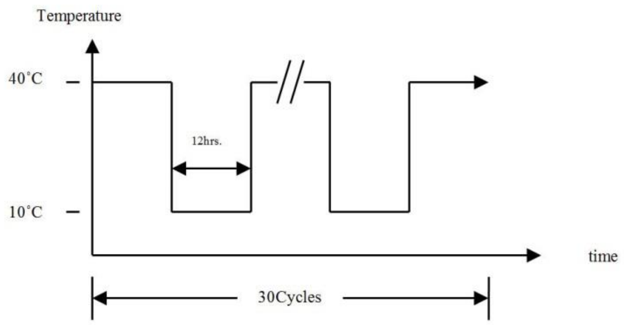

Fig. 2. The fluctuation of ambient temperature in square-wave form.

3. Numerical Method

The mathematical models were solved using numerical method of lines [15], which is the powerful method for solving the parabolic partial differential equations. In numerical method of lines, partial differential equations and boundary conditions were discretized using finite difference approximation. In this article, the second-order forward, centered, and backward differences with the total number of interval of 20 internal nodes were used in the numerical analysis. After discretizing, the PDEs are transformed to a system of ordinary differential algebraic equations, which can then be solved by *ode15s*- a build-in function in MATLAB for solving DAEs by backward differentiation formulas [16, 17]. The numerical analysis can be completed within three minutes for 720 hours simulation of moisture migration.

4. Results and Discussions

4.1. Fitting of Water Sorption Isotherm with GAB(T)

The result of fitting GAB(T) equation was shown in Table 1 and Fig. 3. Table 1 is the list of fitted parameters equation. Figure 3 shows the comparison of the results of sorption isotherm data, GAB(T) equation, and empirical equation by Leaper [2]. It is clearly apparent that GAB(T) equation fits experimental data very well and is also better than the empirical equation. Moreover, the coefficient of determination, R^2, is close to 1.

Table 1. Fitted parameters of GAB(T) equation of sugar grain.

$M_0^{'}$	$C^{'}$	$k^{'}$	$\frac{\Delta H}{RT}$	$\frac{H_l-H_m}{RT}$	$\frac{H_l-H_q}{RT}$	R^2
9.16×10^{2}	5.53×10^{22}	1.20×10^{2}	3.70×10^{2}	-1.54×10^{-4}	-150×10^{2}	0.9706

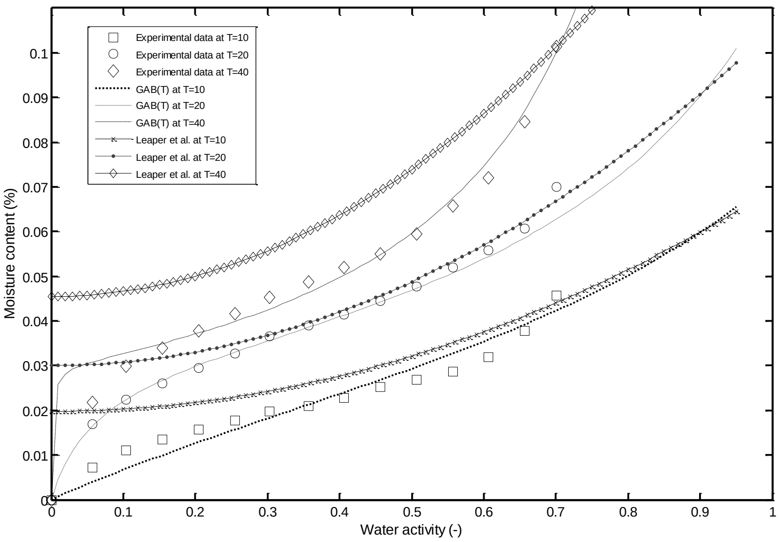

Fig. 3. Sorption isotherm of sugar grain; experimental data by Leaper [2], Fitted GAB(T) equation, and empirical equation by Leaper [2].

4.2. Model Validation

The mathematical model was validated by comparing the numerical result of this model with the experimental data reported by Leaper [18]. In their experiment, sugar initially was kept at 18˚C in the caking box with the height of 0.3 m before one side of the box were maintained at 30 ˚C. The reported initial moisture content was 0.068%. The experiment were conducted for 4 hours. Both experimental and numerical results at 4 hours are shown in Fig. 4. It can be seen that numerical result agrees with experimental data quite well.

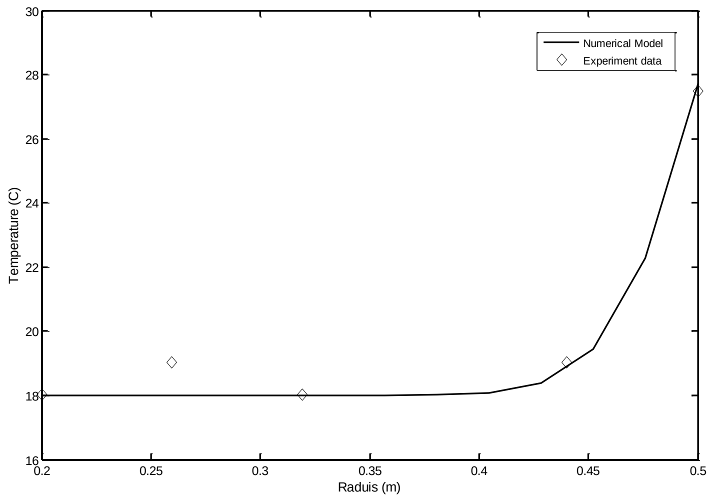

Fig. 4. Comparison of temperature distribution between experimental data by Leaper [18] and numerical result at T_0=18˚C and M_0=0.068% and the outside temperature is 30˚C.

4.3. Effect of Initial Moisture Content on Moisture Migration

As discussed previously, two conditions of initial moisture content of sugar, high moisture (M_0=0.040% and T_0=40˚C) and low moisture (M_0=0.015% and T_0=30˚C), were investigated numerically. These conditions were chosen to investigate the effect of initial moisture content on the moisture migration under the fluctuation of temperature between 40 and 10°C throughout shipping which is approximately 30 days.

The result was considered into two perspectives; short time (less than 36 hours) and longer time perspectives. The variables monitored were consisted of the profiles of temperature, absolute humidity, and moisture content of sugar.

4.3.1. Short time (less than 36 hours)

The numerical result *for high initial* moisture content during the first 36 hours is shown in Fig. 5. It should be pointed out that the profiles of temperature, absolute humidity, and moisture content were obviously not changed for the first 12 hours. This is due to that the initial temperature is the same as the outside temperature during this period. As a result, the temperature and the moisture of gas phase and solid phase is still in equilibrium as in the initial condition.

During the period of 15 to 24 hours, the outside temperature decreased and remained at 10˚C. It has been seen that the temperature at the interface, as well as the absolute humidity, was gradually decreasing with time; however, the moisture content was increasing. This result indicates that there is the adsorption near the interface; since the decreasing temperature, in fact, results in increasing relative humidity, moisture is then adsorbed onto the surface of sugar. Furthermore, it can be observed that the temperature has been affected only 0.15 meters from the interface because of the low thermal conductivity of sugar, whereas both absolute humidity and moisture content have been affected and were decreasing in almost the whole range of bag ($r=0$ to 0.5 meter for absolute humidity, $r=0$ to 0.42 meter for moisture content). This result indicates that the moisture desorbs and diffuses to the interface.

During the period of 27 to 36 hours, the outside temperature increased and remained at 40˚C. It can be observed that the temperature near the interface, as well as absolute humidity, was gradually increasing with time while moisture content was decreasing. This observation means that there is desorption near interface, which was in the reverse direction from the previous period. This phenomenon can be explained by the fact that the increasing temperature results in decreasing of the relative humidity and moisture is subsequently desorbed into the gas phase. Furthermore, it can be observed that both of absolute humidity and moisture content near the core were still decreasing continually from previous the period. This decreases separated the absolute humidity profile into two zones and the moisture content profile into three zones. The result means that moisture was continually desorbed and continually diffused away from core region. According to the temperature profile, it was observed that the range of temperature affected by fluctuation of ambient temperature was gradually increasing with time, 0.20 meter comparing with0.15 meters in the first period.

The numerical result *for low initial* moisture content sugar during the first 36 hours is shown in Fig. 6. It should be pointed out that the profiles of absolute humidity and moisture content in the core region have not been affected. The increase and decrease of moisture content with the temperature near the interface also differed from the case of high initial moisture content. As seen on Fig. 6, the moisture content increases when the temperature increases instead of decreasing as previously believed. The reason for this phenomenon is that the sugar is the hygroscopic material, which would become more hygroscopic as temperature increases [19]; hence the adsorption of water at higher temperature is more important than moisture diffusion. The higher water sorption isotherm at higher temperature shown in Fig. 3 clearly supports this claim.

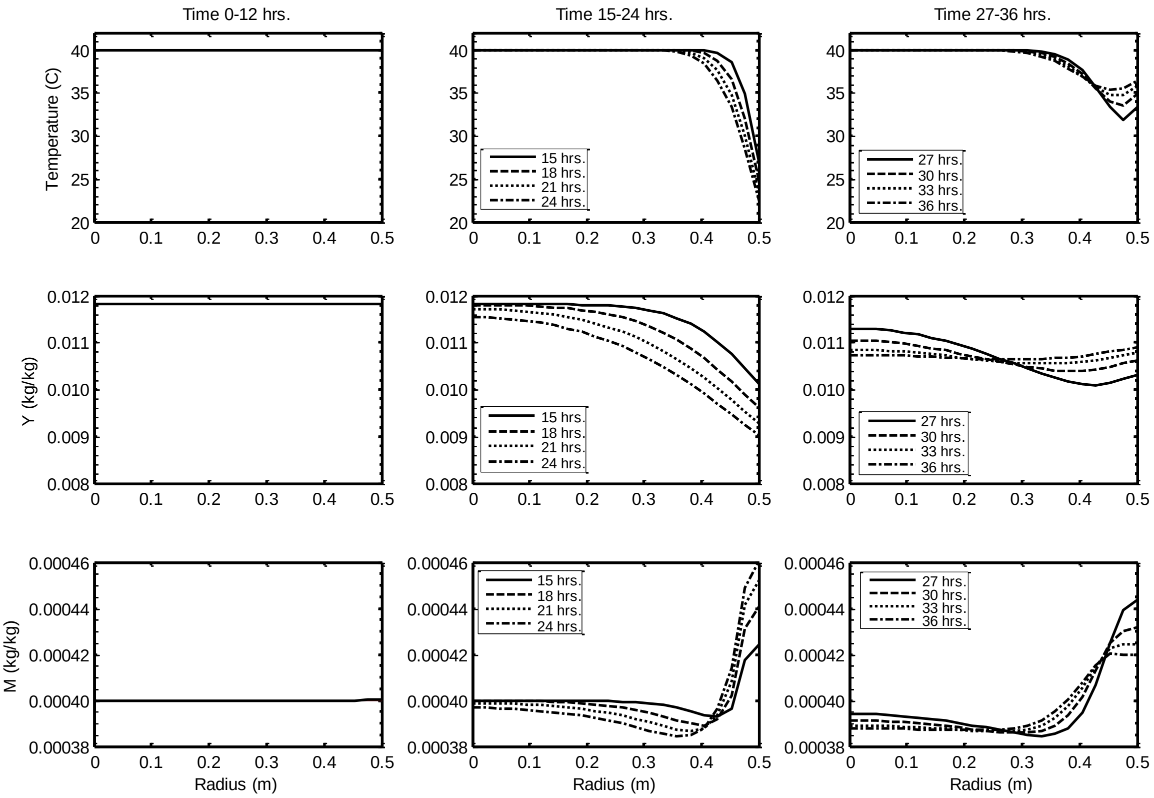

Fig. 5. Dynamics of numerical results for high initial moisture content in first 36 hours.

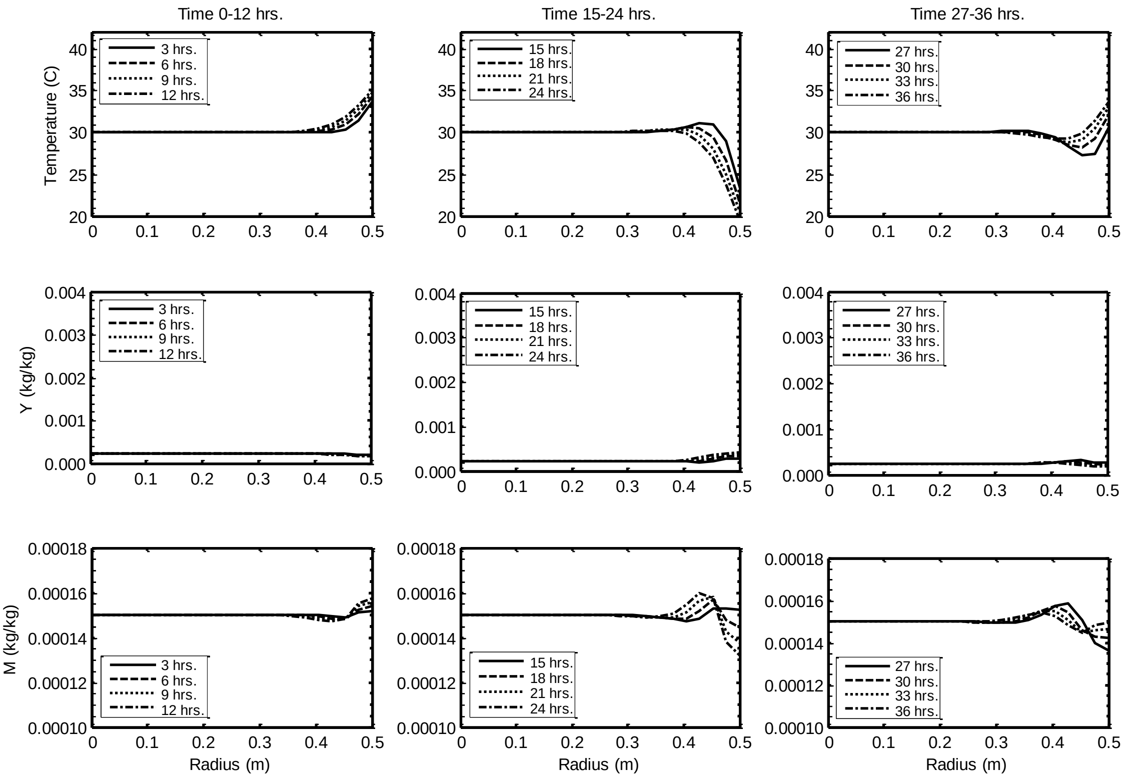

Fig. 6. Dynamics of numerical results for low initial moisture content in first 36 hours.

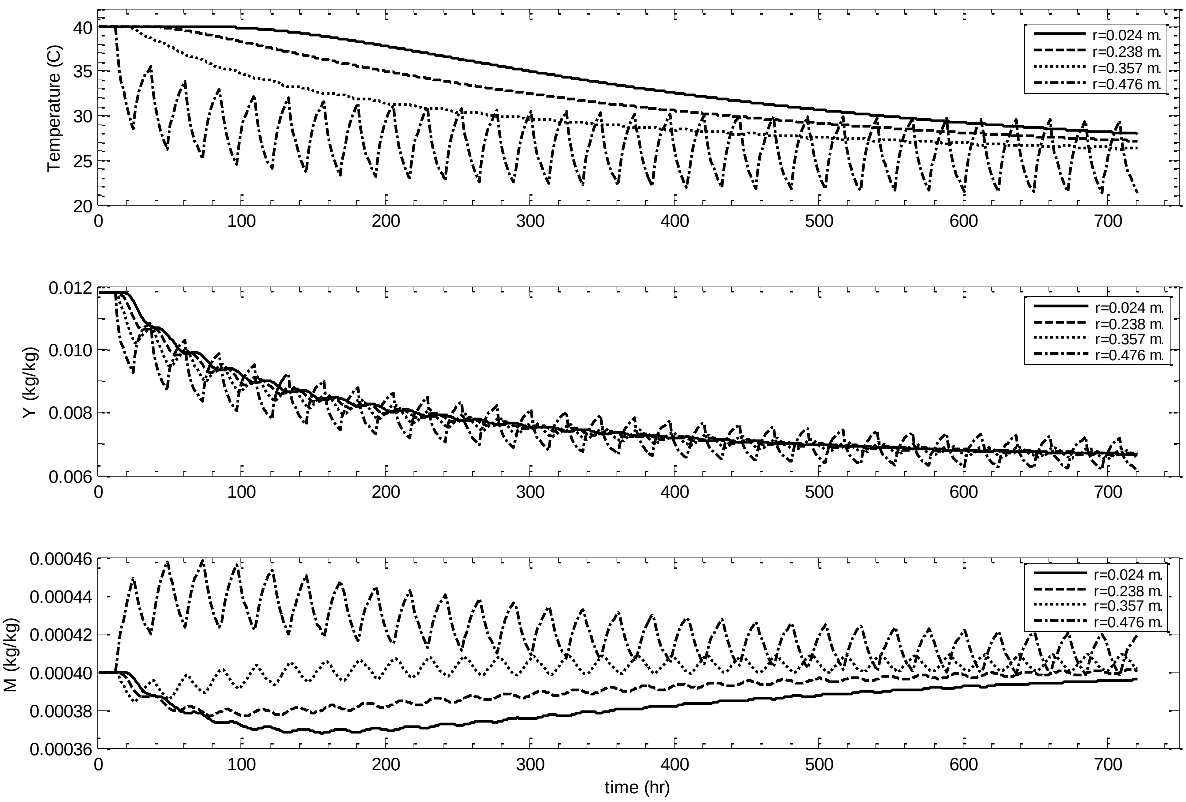

Fig. 7. Numerical result for high initial moisture content through 720 hours.

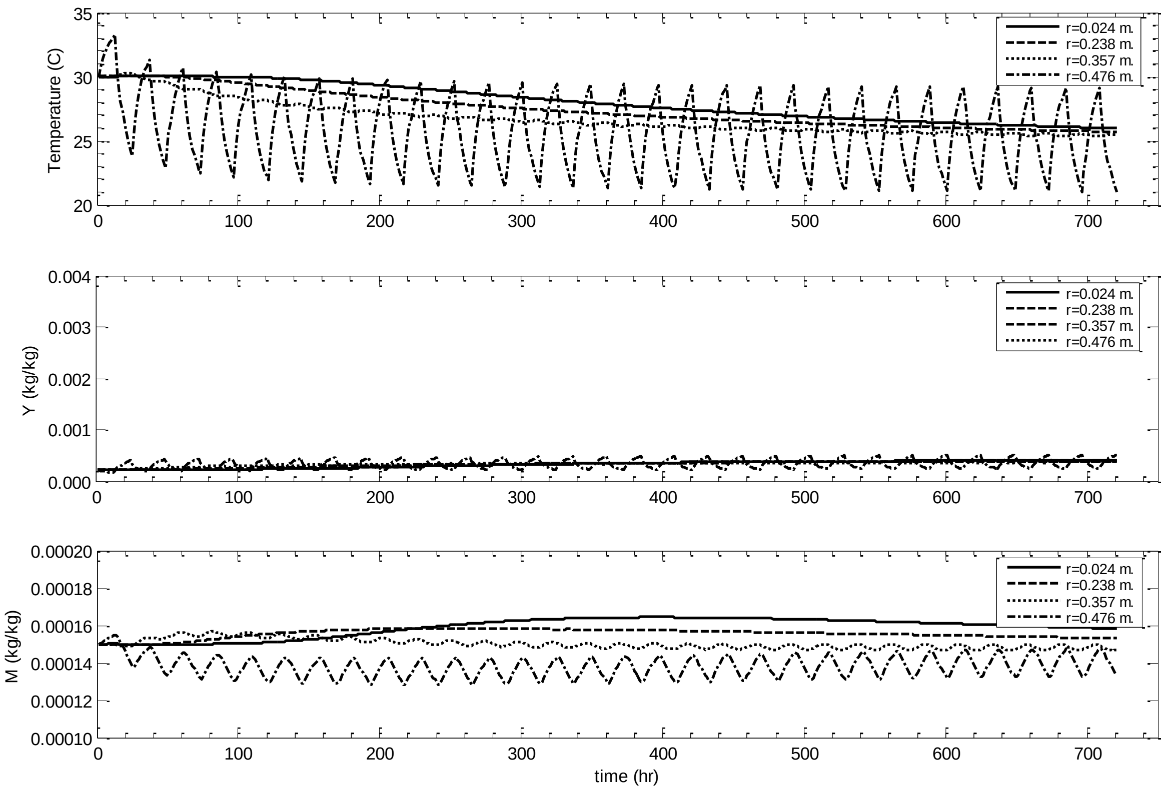

Fig. 8. Numerical result for low initial moisture content through 720.

4.3.2. Long time (through 36 hours)

The numerical result for high initial moisture content through 720 hours is shown in Fig. 7. It can be seen that all profiles oscillate highly near the interface and its oscillations gradually decrease as the distance from the interface increases. It was observed that temperature oscillated around 25˚C near the interface and converged to 25˚C near the core, which is the averaging temperature. For the absolute humidity, it can be seen that the humidity decreased with time; this result indicates that there is decreasing amount of moisture in gas phase. The result also indicates that there is the repeated adsorption and desorption near the interface of big bag and, hence, caking is more likely to happen there. This finding also agrees with Excell and Stone's finding [20], which found the wetted sugar occurred in near the door of a container.

The numerical result for low initial moisture content through 720 hours is shown in Fig. 8. It can be seen that the oscillation of this case is similar to the previous case, especially for temperature profile. The absolute humidity was almost unchanged at the core region while it was fluctuated slightly near the interface. The moisture content near the core region increased with time while the moisture content near the interface region decreased with time along with the repeated adsorption and desorption – these phenomena differ from the high initial moisture content. However, the amplitude of moisture content changes near the interface was quite smaller when being compared with the high moisture content case.

5. Conclusions

The model derived and solved numerically in this article is used to predict the moisture migration and temperature distribution in a big bag of sugar for a shipping or storage and the GAB(T) equation used as a constitutive equation is also correlate the experimental data very well. In this work, it has shown that the moisture diffusion through the interstitial air in big bag of sugar plays an important role in moisture migration when there is the temperature gradient for the case of sugar with high initial moisture content. The interesting observation is that there are the increasing amount of moisture content and periodical adsorption and desorption with high amplitude near the big bag. This phenomenon may induce that the area near the bag is where the liquid and solid bridging and, eventually, caking likely to occur. On the other hand, the prediction of sugar with low initial moisture content has shown that the periodical adsorption and desorption is smaller in magnitude and caking, hence, is probably less serious.

Nomenclature

$a_{w,eq}$	water activity at equilibrium
C	Guggenheim constant
C_{pa}	heat capacity of dry air, J kg^{-1} K^{-1}
C_{ps}	heat capacity of solid sugar, J kg^{-1} K^{-1}
C_{pv}	heat capacity of water vapor, J kg^{-1} K^{-1}
C_{pw}	heat capacity of water liquid, J kg^{-1} K^{-1}
D_a	diffusivity of water vapor in dry air, m^2 s^{-1}
D_{eff}	effective diffusivity of water vapor, m^2 s^{-1}
ERH	equilibrium relative humidity, %
H	total enthalpy, J m^{-3}
h_g	enthalpy of gas, J kg^{-1}
h_s	enthalpy of sugar, J kg^{-1}
h_v	enthalpy of water vapor, J kg^{-1}
h_{fg}	latent heat of vaporization of water, J kg^{-1}
k	constant correcting properties of multilayer molecules with respect to bulk liquid
k_a	thermal conductivity of gas phase, W m^{-1} K^{-1}

k_s thermal conductivity of solid phases, W m^{-1} K^{-1}
k_{eff} effective thermal conductivity, W m^{-1} K^{-1}
M moisture content in solid phase, kg of water/ kg of dry solid
M_{eq} moisture content in solid phase at equilibrium, %
M_0 monolayer water content
Nu Nusselt number
Pr Prantdl number
P_{sat} vapor pressure of water, mmHg
Ra Rayleigh number
r radius from the center, m
T temperature, °C
$T_{outside}$ ambient temperature, °C
t time, s
W total moisture in bed, kg of water/m^3
Y absolute humidity in gas phase, kg of water/ kg of dry air
Y_{eq} absolute humidity in gas phase, kg of water/ kg of dry air

6. Greek symbols

ε porosity of bed
ρ_{da} density of dry air, kg m^{-3}
ρ_s density of sugar, kg m^{-3}

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