Application of Empirical Peleg Model to Study the Water Adsorption of Full Cream Milk in Drying Process

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Abstract. The ability of spray dryer in producing full cream milk at different inlet temperatures and the effectiveness of empirical model used in order to interpret the drying process data is evaluated in this study. In this study, a lab-scale spray dryer was used to dry full cream milk into powder with inlet temperature from 100 to 160°C with a constant pump speed 4rpm. Peleg empirical model was chosen in order to manipulate the drying data into the mathematical equation. This research was carry out specifically to determine the equilibrium moisture content of full cream milk powder at various inlet temperature and to evaluate the effectiveness of Peleg empirical model equation in order to describe the moisture sorption curves for full cream milk. There were two conditions set for this experiments ; in the first condition (C1), further drying process of milk powder in the oven at 98°C to 100°C while the second condition (C2) is mixing the milk powder with different salt solutions like Magnesium Chloride (MgCl), Potassium Nitrite (KNO₂), Sodium Nitrite (NaNO₂) and Ammonium Sulfate ((NH₄)₂SO₄). For C1, the optimum temperature were 160°C with equilibrium moisture content at 3.16 % weight dry basis and slowest sorption rates (dM/dt) at 0.0743% weight dry basis/hr. For C2, the best temperature for the mixture of dry samples with MgCl is at 115°C with equilibrium moisture content and sorption rates is -78.079 % weight dry basis and 0.01 % weight dry basis/hr. The best temperature for the mixture of milk powder with KNO₂ is also at 115°C with equilibrium moisture content and sorption rates at -83.9645% weight dry basis and 0.0008% weight dry basis/hr respectively. For mixture of dry samples with NaNO₂, the best temperature is 160°C with equilibrium moisture content and sorption rates at 84.1306% weight dry basis and 0.0013% weight dry basis/hr respectively. Lastly, the mixture of dry samples with ((NH₄)₂SO₄ where the best temperature is at 115°C with equilibrium moisture content - 83.8778% weight dry basis and sorption rates at 0.0021% weight dry basis/hr. The best temperature selected best on the lowest moisture content formed and also the slowest sorption rates.

1. Introduction

According to [1] the model implied that the moisture equilibrium was somewhat higher than that determination on the assumption that the sample reached constant weight, but there was no conclusive
evidence that this was really the case. This is because, the weight of the full cream milk powder under two conditions which are soaked in different salt solutions and being dried inside the oven at 98°C to 100°C never reached a constant weight. The reading was taken every 30 minutes within 270 minutes. The weight of the full cream milk powder under this two conditions were decreasing but never achieved the constant weight that can be indicated the moisture equilibrium was achieved.

Not only the weight of the full cream milk samples was investigated, but after plotting a graph of moisture content Wet Basis (%) versus moisture content Dry Basis (%), the graph obtained was in linear regression form. This prove that the statement from M.Peleg, 1988 where published sorption curves in the form of moisture versus time relationship of milk powder and rice, exposed to moist atmosphere or soaked in water, were fitted by a two parameter, non-exponential empirical model was true. Unlike other agriculture products like bananas [13], green beans and olive cake [4] the equilibrium of moisture content can be investigated by using Mathematical Model like Lewis, Henderson and Pabis, Wang and Singh. Since the graph of moisture content (wet basis) (%) versus moisture content (dry basis) (%) obtained was in the exponential form, therefore, Peleg equation was chosen in order to determine the equilibrium moisture content for full cream milk powder under two different conditions since this model enabled prediction of moisture contents after long exposure from experimental data obtained in relatively short time exactly well before the moisture level appeared to reach a plateau.

The challenges that rise up regarding this research are the ability of the spray dryer to dry up the full cream milk at different temperatures and the effectiveness of empirical model used in order to interpret the drying process data for the description of moisture sorption curves. In order to overcome these problems, the lab-scale spray dryer was used to dried up the full cream milk into powder with the selection of temperatures of 100°C, 115°C, 130°C, 145°C and 160°C with a constant pump speed which is 4 rpm while the Peleg empirical model was chosen in order to manipulate the drying data into the mathematic equation.

The objectives of this study is, to determine the equilibrium moisture content of full cream milk powder at various inlet temperature and evaluate the effectiveness of Peleg empirical model equation in order to describe the moisture sorption curves for full cream milk powder

1.1. Drying process of full cream milk

As we know, the pathogens like bacteria, gems and bugs growth rapidly in a moisture condition. Therefore, drying will solve the problem. Drying is the process designated in order to remove the water or any types of solvents by evaporation from a solid, semi-solid or liquid surface. While drying process takes place, water molecule was migrate from the interior surface of the product to the outer surface of the product by a molecular diffusion. Following the Second Law of Thermodynamics, the water will remove from the higher moisture content to the area of lower moisture content. For the drying process, moisture content plays the important role for the drying process to take place. Milk can be present as powder to solve the following above problems. The milk powder can be produced by using spray drying and roller drying. Spray Drying Process

The process of spray dryer is starting from the introduction of concentrated milk into a chamber (usually a fine mist) through which hot air is circulating. The milk droplets soon will lose their water and fall to the floor as fine powder after finish the process. Spray-dryer can be used to dry many form of milk such as whole milk, skim milk, whey and fat-enriched milk. Increased demand for milk powder especially in food industry has led the spray-dryer to widening its application. A milk spray-dryer is usually used in a large capacity with a million liters of milk per day therefore make it more economic. Usually, the inlet temperature chose around 200°C (400°F) to maintain the powder quality. The important properties for the final products of milk spray-dryer are flow properties, wettability,
sinkability, dispersability and solubility. For whole milk powder, it demands for fast reconstitution ability by the addition of additives called lecithin.

1.2. Mathematical models for drying process

The results from the drying process of full cream milk was determined for further steps in order to determine the best fit mathematical model for this product. There are many mathematical models used in agricultural field to identify the best fit mathematical models that can be used to ensure the effectiveness of the drying process. Many researches for agriculture applied several types of mathematical models to interpreted and explained each of them was the best fit for the drying process besides claimed the calculation for the convection drying kinetics.

The examples of mathematical models that are usually used for drying process of agriculture products are Lewis, Henderson and Pabis, Wang and Singh, Peleg, Page and Silva et al. Table 1 below shows varies empirical models to describe drying kinetics.

| Mathematical Model       | Empirical Expression |
|--------------------------|----------------------|
| 1 Lewis                  | $X^* = e^{at}$       |
| 2 Henderson and Pabis    | $X^* = ae^{bt}$      |
| 3 Wang and Singh         | $X^* = 1 + at + bt^2$|
| 4 Peleg                  | $X^* = 1 - t(a+bt)$  |
| 5 Page                   | $X^* = e^{at+b}$     |
| 6 Silva et al            | $X^* = e^{at+b+1}$   |

From Table 1, it can be seen that all types of mathematical model have always been used together with the result from the drying process of the agricultural products. The examples of drying process that cluster with these variations of mathematical models are Mathematical models to describe thin-layer drying and to determine drying rate of whole bananas [11]. Modelling of olive cake thin-layer drying [15], thin-layer modeling of black tea drying process and drying behavior of green beans [15] are the proof of all types of mathematical models such as Lewis, Page, Modified Page, Henderson and Pabis and also Logarithmic are reasonable and logical to be used.

Among several journals stated before, a thin-layer drying model for agricultural products was used where it described well that the drying process of agricultural materials mainly fall into three categories. The three categories involved namely as theoretical, semi-theoretical and the last one is empirical. Theoretical takes into account only the internal resistance to moisture transfer while for both semi-theoretical and empirical consider only external resistance to moisture transfer between product and air. All the types of mathematical models stated in Table 1 were declared as semi-theoretical thin layer drying models. As they were applied this models to the agricultural products, it might also be suitable for the milk drying process. The steps of drying process and also the calculation involved the moisture ratio can be applied also to the drying process of milk.
2. Methodology

2.1 Materials

The equipment used in this research is laboratory scale spray dryer. Besides that, the materials involved in this experiment are full cream milk (liquid form), Magnesium Chloride (MgCl), Potassium Nitrite (KNO$_2$), Sodium Nitrite (NaNO$_2$) and Ammonium Sulfate ((NH$_4$)$_2$SO$_4$). The apparatus used in this experiment are 50mL beaker, 1000mL beaker, 1000mL conical flask, Cryovac® plastic bag, aluminum foil and electronic weighing balance.

2.2 Moisture content measurement

Full cream milk was stored in a refrigerator prior to spray drying. The inlet temperature of the spray dryer was set up with ranges from 100°C to 160°C. The amount of the powder collected in cyclone collector (cyclone recovery) and manual sweeping the spray dryer’s wall was packed and stored in the desiccator. Empty beaker was weighed on electronic weighing balance and recorded as W1. Then 2g of full cream milk powder were added and recorded as W2.

2.3 Moisture Content Calculation

The initial weight of the full cream milk powder, $W_o$ was calculated by using Equation 2.1:

$$W_o = W_2 - W_1$$ (1)

For condition one, the samples were left inside the oven at 98°C to 100°C until final weight is achieved. The final weight, $W_D$ of these samples was recorded for every 30 minutes. The moisture content in wet basis, $M_{wb}$ and moisture content in dry basis, $M_{db}$ was calculated by using this Equation 2.2 and 2.3.

$$M_{wb} = \frac{(W_0-W_D)}{W_0}$$ (2)

$$M_{db} = \frac{(W_0-W_D)}{W_D}$$ (3)

For condition two, full cream milk powder was mixed with four types of salts ; Magnesium Chloride (MgCl), Potassium Nitrite (KNO$_2$), Sodium Nitrite (NaNO$_2$) and Ammonium Sulfate ((NH$_4$)$_2$SO$_4$). All samples were stored at the evacuated desiccators over P$_2$O$_5$.

2.4 Empirical Peleg Model equation

The graph $t/(Mt-Mo)$ (hour/M) against time (hour) was constructed from tabulated data and constant $k_1$ and $k_2$ was determined. With the value of $k_1$ and $k_2$ constant, initial moisture content (Mo) (% weight dry basis), moisture content at time $t$ ($M(t)$) (% weight dry basis), equilibrium moisture content (ME) (% weight dry basis), the sorption rate ($dM/dt$) and ratio (R) was calculated and extracted from the graph. The time taken ($t_R$) required to achieved ratio, R was calculated using Equation 2.4- 2.8 :

$$M(t) = Mo + t/(k_1 + k_2t)$$ (4)

$$ME = Mo + 1/k_2$$ (5)

$$dM/dt = k_2/(k_1 + k_2t)^2$$ (6)

$$R = (Mt-Mo)/(ME-Mo)$$ (7)

$$t_R/t_{1/2} = R/(1-R)$$ (8)
3. Results and Discussion

For most of the results, at different each temperature, the final weights, WD was in a range between 1.8532g to 1.9776g. The highest final weight, WD value was found to be at 1.9776g for sample number 1 at temperature 160°C while the lowest final weight, WD was found to be at 1.8532g for sample number 10 at 100°C. In addition, the average dry basis moisture content, MDB % also shows the lowest value with 1.1314% weight dry basis. Overall, the trend whereas the temperature increases, the value for final weight, WD are also increases. At high final weight, WD, the value for average dry basis moisture content, MDB % will decreases at best temperature 115 °C and 160 °C.

Figure 1. Graph of t/[M(t)-Mo] (hour/M) versus Time (hour) for (a) temperature 160°C and (b) temperature 115°C.

The graph of t/[M(t)-Mo] (hour/M) versus Time (hour) for dry sample of full cream milk powder mixed with MgCl showed in Figure 2 (a) and (b) at temperature 160 °C and 115 °C respectively resulted in linear regression line. In (a), the value for t/[M(t)-Mo] (hour/M) was suddenly drop due to sudden increased of dry samples weight at time 4.5 hours. This is because by the exposure of the sample number 10 towards the environment thus make it adsorb the moisture content or humidity from the environment. Therefore, it is very important to tighten up the samples beaker. From the equation, the value of constants k1 (hr/%weight) and also k2 (1/%dry weight basis) are 16.488 and 16.55 respectively. In (b) the weight dry samples of full cream milk powder mixed with MgCl decreases, with the increases of t/[M(t)-Mo] (hour/M) within the time 0.0 hour to 4.5 hours. The value of constants k1 (hr/%weight) and also k2 (1/%dry weight basis) directly extracted from the linear equation were 116.49 and 161.37 respectively. Both k1 and k2 constant values can be further used in Peleg equation in order to determine the equilibrium moisture content (ME) and also the sorption rate (dM/dt) for every sample from 0.0 hour to 4.5 hours.

Figure 2. Graph of t/[Mt-Mo] (hour/M) versus Time (hour) at temperature (a) 160 °C and (b) 115°C in MgCl solution.
Figure 3 shows the graph of \( t/[M_t-M_0] \) (hour/M) versus Time (hour) for dry samples of full cream milk powder mixed with NaNO2 at temperature 160°C and 115°C. The graph plotted was in a best fit linear regression line. In graph (a) the equation of the graph is \( y = 9.9408x + 3.0498 \). Therefore, where the constants \( k_1 \) (hr/%weight) and also \( k_2 \) (1/%dry weight basis) are 3.0498 and 9.9408 respectively. In graph (b) \( y = 40.664x + 24.198 \). Thus, the constant values required in Peleg equation can be extracted directly from the equation where the constants \( k_1 \) (hr/%weight) and also \( k_2 \) (1/%dry weight basis) are 24.198 and 40.664 respectively.

4. Conclusion

In this research, empirical Peleg model was selected in order to describe the moisture sorption curves of full cream milk powder under certain condition. The graph of \( t/(M_t-M_0) \) versus time (hour) formed a linear regression thus make it suitable to use the empirical Peleg model. The equilibrium moisture content (ME) was easily calculated by using this model as compared with the Page and Lewis model. This is because, the data sets of full cream milk powder after drying process was differ with other agriculture products like whole banana, olive cake, ginger and peach slice as mentioned in many journals.

Following steps in determination of equilibrium moisture content (ME) by using Page and Lewis model, a graph of moisture content wet basis % versus moisture content dry basis % was illustrated. Unfortunately, the graph obtained was not in exponential form due the weight of dry samples of full cream milk at certain conditions is changed with time. There is no constant weight was recorded in during 270 minutes experiment running. The graph of moisture content wet basis % versus moisture content dry basis % was in a linear regression form. This make it difficult in order to determine the equilibrium moisture content (ME) by using Page and Lewis model since the constant required in the equation is absence.

Acknowledgement

Support for this research was provided by Research Acculturation Grant Scheme (RAGS/1/2014/TK05/UiTM/4) from the Ministry of Higher Education of Malaysia and Research Management Institute (RMI), Universiti Teknologi Mara (UiTM) Shah Alam. Thanks to all who may contribute directly or indirectly towards making this research successful.
References

[1] Peleg M, An Empirical Model for the Description of Moisture Sorption Curves, 1216 J. of Food Sci.-Volume 53 No. 4, 1988
[2] Kockel T K, Allen S, Hennigs C, Langrish T A G, An experimental study of the equilibrium for skim milk powder at elevated temperatures, 51 291-297.
[3] Wang S, Langrish T, A review of process simulations and the use of additives in spray drying, 42 13-25.
[4] Chen X D and Lin S X Q, Air Drying of Milk Droplet under Constant and Time-Dependent Conditions, 51 1790-1799.
[5] Gharsallaoui A et al 2007 Applications of spray-drying in microencapsulation of food ingredients: An overview, 40 1107-1121.
[6] Midilli A , Kucuk H & Yaper Z, A new model for single-layer drying, 20 1503-1513.
[7] Zafer E, Filiz I, A Review of Thin Layer Drying of Foods: Theory, Modeling, and Experimental Results, 50 441-464.
[8] Langrish T A G, Chan W C, Kota K, Comparison of maltodextrin and skim milk wall deposition rates in a pilot-scale spray dryer, 179 84-89.
[9] Chiou D, Langrish T A G, 2007 Development and characterisation of novel nutraceuticals with spray drying technology, 82 84-91.
[10] James P, George A.K, Datta, Development and validation of heat and mass transfer models for freeze-drying of vegetable slices, 52 89-93.
[11] Silva W P et al 2013 Mathematical models to describe thin-layer drying and to determine drying rate of whole bananas 13 67-74.
[12] Mezhericher M, A. Levy a & I. Borde, Model of Single Droplets Containing Insoluble or Dissolved Solids, 25 1025-1032.
[13] Zhu A S, Shen X Q, The model and mass transfer characteristics of convection drying of peach slices, 72 345-351.
[14] Athanasia M G, Thodoris D K, Dimitris S A, Konstantinos G A 2008 Water sorption isotherms and glass transition temperature of spray dried tomato pulp, 85 73-83.
[15] Nalan A A, Doymaz I, Modelling of olive cake thin-layer drying process, 68 455-461.