Shelf-life Prediction of Micro-encapsulated Shrimp Oil in Different Packages using Empirical Models

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Received: 3 May 2019, Revised: 10 September 2019, Accepted: 7 October 2019

Abstract

Micro-encapsulated shrimp oil (MSO) can be used to fortify food products and may be especially useful in the development of new functional foods. The shelf-life of packaged MSO was studied and sorption isotherms were examined. Moisture sorption isotherms and moisture sorption kinetics of MSO were evaluated at 30°C for water activity (a_w) values ranging from 0.113 to 0.923 by a static gravimetric method. Empirical models were determined to predict the experimental data. The initial stage of moisture sorption by the MSO was relatively rapid and it decreased with time. All the sorption curves were found to be type III. The most suitable model for predicting the moisture sorption isotherm of MSO was the GAB model because it had the lowest percentage root mean square error (RMSE). The shelf-life of MSO packaged in polypropylene (PP), Nylon/linear low density polyethylene (Nylon/LLDPE) and metalized polyethylene terephthalate (metalized PET) pouches stored at 30°C and either 75% or 80% relative humidity (RH) was predicted by the GAB equation and the longest shelf-life of MSO (507 days in 80% RH and 725 days in 75% RH) was found in metalized PET packages. The empirical models can be useful for predicting the shelf-life of MSO.

Keywords: micro-encapsulated shrimp oil, moisture sorption isotherm, empirical model, shelf-life, packaging
DOI 10.14456/cast.1477.3

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1. Introduction

Hepatopancreas is a by-product generated from the manufacturing of hepatopancreas-free whole shrimp. Shrimp oil from hepatopancreas is one of the major sources of polyunsaturated fatty acids (PUFAs) such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) and it also contains the antioxidant astaxanthin. PUFAs are known for their health benefits, which include their ability to prevent lipotoxicity, cardiovascular diseases, human breast cancer, inflammatory diseases and Alzheimer's disease [1]. The use of shrimp oil in food products has been limited owing to the high susceptibility of PUFAs to oxidation. Encapsulation can be used to delay or inhibit oxidation and mask unpleasant flavors and odors in the resulting products [2] and the micro-encapsulation of oil can be achieved by spray drying [3]. Good quality microcapsules, with low water activity (a_w) and ease of storage and handling, can protect the active material against undesirable reactions. However, many food ingredients that are added in the form of powders tend to have sticking and caking problems due to moisture adsorption during storage. Water molecules permeate into the package, leading to an increase in moisture content and a decrease in the product stability and dispersibility over time [3]. Moisture sorption isotherms are related to the hydration of food products [4] and represent a correlation between a_w and equilibrium moisture content (EMC) in food products at a constant temperature [5]. Therefore, moisture sorption isotherms are a useful means of monitoring the drying process of products and the shelf-life stability of packaged products. They are usually classified into Types I, II, III, IV and V [6]. In general, dried food products have Type II or III isotherms [7]. Sorption isotherms have previously been described by mathematical models validated by statistical methods. Theoretical mathematical models were developed to describe sorption mechanisms. Some mathematical models are just empirical, or simplifications of more elaborate models [7, 8]. There are some semi-empirical equations with two or three fitting parameters to describe moisture sorption isotherms [7]. The BET equation, the GAB model, the Peleg model, the Lewicki model, the Oswin model and the Smith model are widely used to describe sorption in food products. Thus, it is a requirement to choose a suitable equation of moisture sorption isotherm for a specific product. This study aimed to investigate moisture sorption kinetics in order to select an empirical model for the moisture sorption isotherm and to evaluate the shelf-life of micro-encapsulated shrimp oil (MSO) packaged in bags made from three materials; (polypropylene (PP), nylon/linear low density polyethylene (Nylon/LLDPE) and metalized polyethylene terephthalate (metalized PET)).

2. Materials and Methods

2.1 Materials

Sodium caseinate was procured from Vicchi Enterprise Co., Ltd. (Bangkok, Thailand). Fish skin gelatin (bloom strength of 230-250 g) was procured from Lapi Gelatine S.p.A. (Milano, Italy). Glucose syrup was obtained from Charoenworrakit Co., Ltd. (Samut Prakan, Thailand). All the plastic bags were purchased from a supermarket. Polypropylene (PP), Nylon/linear low-density polyethylene (Nylon/LLDPE) and metalized polyethylene terephthalate (metalized PET) bags with the dimension of 3.0 cm × 2.0 cm were used.

2.2 Preparation of micro-encapsulated shrimp oil (MSO)

The shrimp oil was extracted from the digestive gland (hepatopancreas) of Pacific white shrimp (*Litopenaeus vannamei*) using a mixture of isopropanol and hexane as per the method of Takeungwongtrakul *et al.* [9]. Fish gelatin, sodium caseinate and glucose syrup were mixed at a ratio of 1:1:4 (w/w/w) in deionised water, to prepare the "wall materials" according to the method of Takeungwongtrakul and Benjakul [3]. Shrimp oil was added into the solution at a core/wall material ratio of 1:4 (v/v). The mixtures were homogenised at a pressure level of 4,000 psi for 4 passes. A SD-06A spray-dryer (LabPlant Ltd., Huddersfield, UK) was used to produce micro-encapsulated shrimp oil (MSO) following the method of Takeungwongtrakul *et al.*[9]. Four grams of MSO was packaged in the different bags as described above and heat-sealed.

2.3 Determination of initial and critical moisture content

Initial moisture content (IMC) was determined following the method of AOAC [10], and the critical moisture content (CMC) of MSO was determined by placing MSO into water desiccators at 30°C until MSO was assessed as being at an unacceptable level by 10 trained panelists. The panelists were trained to evaluate the appearance of MSO. Prior to the evaluation, the panelists were trained three times a week. They were trained with standards for two sessions using a scale of 0-15, where 0 and 15 represented dry and wet MSO, respectively. The appearance of MSO was the initial quality that was used to judge the acceptability of MSO.

MSO was evaluated for moisture content (% dry weight basis) every hour until MSO samples reached their CMC. The CMC was identified as the point at which the deterioration of MSO reached an unacceptable level. The CMC was reached when the MSO particles stuck together to a level at which it was be rejected by the panelists.

2.4 Moisture sorption characteristics

2.4.1 Moisture curve and rates

The sorption isotherm was determined by a static gravimetric technique, using different saturated salt solutions [11]. MSO was pre-dried in a desiccator over phosphorus pentoxide (P_2O_7) and kept in separated desiccators with different saturated salt solutions. The desiccators were then kept in an electric oven at 30±1°C, which gave equilibrated environments in the desiccators. All saturated salt solutions ($MgCl_2$, KI, KCl, NaCl and K_2NO_3) were selected to produce a broad range of a_w (0.324-0.923) [12]. The a_w of storage conditions were confirmed by means of a humidity meter (HM70, Vaisala, Helsinki, Finland). Weight of MSO was determined as a function of time. Moisture content (g H_2O/100 g dry sample) of MSO was determined by placing samples in an oven at 105°C for 3 h. Water activity (a_w) was measured using an AquaLab Series 3 water activity meter (Decagon Devices, Inc., Pullman, WA, USA). The moisture adsorption data were fitted into Peleg's equation [12]:

$$M_t = M_0 + t / (k_1 + k_2.t) \quad (1)$$

where M_t represents the moisture content after designated time of storage (% dry weight), M_0 represents the initial moisture content (% dry weight), k_1 represents the constant of Peleg rate (h/g water/g solids) and k_2 represents the constant of Peleg capacity (g solids/g water).

2.4.2 Sorption isotherm

The moisture sorption isotherm of MSO was determined for the a_w range of 0.113-0.923 using different saturated salt solutions in the desiccators as described above The salt solutions were K_2NO_3, KCl, NaCl, KI, $Mg(NO_3)_2$ and $MgCl_2$ at the following a_w of 0.923, 0.836, 0.751, 0.679, 0.514 and 0.324, respectively. Sample weights, as a function of time, were determined until they reached equilibrium. The moisture contents (% dry weight basis) in the samples were measured at 105 °C for 3 h using an oven. The a_w was evaluated using an Aqua-Lab Water Activity Meter. The percentage of the equilibrium moisture content (EMC) in the samples at each specific a_w was determined using equation 2 [13]:

$$\% \text{ EMC} = (W_e / W_i) \times (M_i + 1) - 1 \quad (2)$$

where W_e represents total weight (dry weight + water) at equilibrium (g), W_i represents the initial weight (g) and M_i represents the initial moisture content (g/g). Thus, a sorption isotherm was constructed by plotting between a_w and % EMC.

2.4.3 Moisture sorption isotherm modeling of MSO

The models of the sorption isotherms of MSO were expressed as shown in Table 1. The model parameters were derived from the experimental results. The root mean square error (RMSE) is the predicting capability of a model in relation to the number of data points. The RMSE was calculated using equation 9.

Table 1. Moisture sorption models for fitting experimental data

Model names	Model equations	Equation number	References
BET	$M = m_o C a_w/[(1 - a_w) + (C - 1)(1 - a_w)\, a_w]$	3	Brunauer *et al*. [14]
GAB	$M = m_o C k a_w/[(1 - k a_w)(1- k a_w + C k a_w)]$	4	Van Den Berg [15]
Peleg	$M = a.a_w^b + c.a_w^d$	5	Peleg [16]
Lewicki	$M = [F/(1 - a_w)^G] - [F/(1 + a_w^H)]$	6	Lewicki [17]
Oswin	$M = k[a_w/(1 - a_w)]^c$	7	Oswin [18]
Smith	$M = C_1 + C_2.\ln(1 - a_w)$	8	Smith [19]

$$RMSE = \sqrt{\frac{\Sigma[(M_{exp} - M_{cal})/M_{exp}]^2}{N}} \times 100 \quad (9)$$

where M_{exp} is the equilibrium moisture content determined from the experimental data and M_{cal} is the equilibrium moisture content predicted using the mathematical models; N is the number of experimental points.

2.5 Shelf -life prediction of the packaged MSO

MSO (4 g) was packed in the different plastic bags (PP, Nylon/LLDPE and metalized PET) and the bags were heat sealed. MSO is very sensitive to changes in % RH. A small change in RH affects the amount of water in MSO. Thus, the packaged MSO samples were stored at 30°C and either 75% RH or 80% RH for 13 weeks in an environmental chamber (WTB Binder, Tuttlingen, Germany). Water vapour permeability (WVP) was determined by a modified ASTM method [20] and calculated using equation 10:

$$\text{WVP (g m}^{-1}\text{ s}^{-1}\text{ Pa}^{-1}) = w\, l\, A^{-1} t^{-1} (P_2 - P_1)^{-1} \quad (10)$$

where w is the weight gain of the cup (g); l is the film thickness (m), A is the exposed area of film (m^2), t is the time of gain (s), (P_2-P_1) is the vapor pressure difference across the film (Pa). Three films were used for WVP testing.

The simplest shelf-life calculation is when the isotherm is treated as a linear function in the linear model [21]. The model can be calculated using equation 11:

$$t = [(l.w_d.\ \beta) / (P.A.p_s)].\ln [(a_{wo} - a_{w\ t=0}) / (a_{wo} - a_{w\ t=t})] \quad (11)$$

where w_d (g) represents the dry weight in the packaged food, l (cm) represents the film thickness, P represents the water vapour permeability coefficient (g water.cm/cm^2.sec.cm Hg), A represents the package surface area (cm^2), p_s represents the saturated vapour pressure of water at the storage temperature (cm Hg), a_{wo} represents the water activity of storage conditions, $a_{w\ t=0}$ represents the initial water activity of product, $a_{w\ t=t}$ represents the critical water activity of product at time = t, and β represents a slope of the moisture sorption isotherm.

Shelf-life calculation was verified by using the actual package product storage test. The MSO in a bag rendering the highest shelf-life was studied. The samples were regularly examined for the EMC. The value of RMSE was used to evaluate the goodness of fit between the experimental and the predicted moisture content. The lower value of RMSE indicates the better fit.

2.6 Statistical analysis

Experiments were performed in triplicate and data were subjected to analysis of variance (ANOVA) and mean comparison by Duncan's Multiple Range Test using the Statistical Package for Social Science (SPSS for windows, SPSS Inc, Chicago, IL, USA). The parameters of the different models were obtained from the regression curves by statistical tools using Microsoft Excel 2007.

3. Results and Discussion

3.1 Moisture content and a_w of MSO

MSO initially had a moisture content of 1.13 ± 0.01% (dry basis), which corresponded to a low a_w (0.35 ± 0.01). The CMC and critical a_w were 7.78 ± 0.07% (dry basis) and 0.71 ± 0.02, respectively. MSO readily adsorbs moisture from the surrounding atmosphere and is considered a moisture sensitive food. A high a_w in food products leads to shorter storage life because high free water promotes biochemical reactions that are associated with deterioration [22].

3.2 Moisture sorption kinetics

The sorption kinetic curves of MSO are given in Figure 1. Moisture sorption rapidly increased during the initial stage, while less moisture was adsorbed as storage time progressed and moisture content plateaued, which was likely to be the equilibration a_w in each condition. At a_w of 0.324-0.923, the time required to reach each equilibrium a_w increased when the a_w increased. Baucour and Daudin [23] reported that mass transfer is very slow at high a_w, indicating that it is difficult to reach equilibrium at a_w of 0.9-1.0. The sorption kinetics of MSO under experimental situation were predicted with Peleg's equation (Eq. 1), which is associated with the hydration behavior of MSO. The coefficient of determination was found to be high in all cases (R^2> 0.99), which represented a desired fit to the experimental data. The constants of Peleg's rate (k_1) and capacity

(k_2) are shown in Table 2. Food products generally kept at a higher RH have lower values of k_1 and k_2 [24]. Turhan *et al.* [25] reported that k_1 is associated with mass transfer. It means that a lower value of k_1 indicates a higher degree of initial moisture adsorption rate. The results indicated that a decrease in k_1 of MSO was also found when a_w was increased, which was in accordance with an increase in initial rate of water adsorption. The k_2 is associated with the maximum water adsorption capacity. The lower constant value of k_2 indicates a higher adsorption capacity [25]. The k_2 decreased as a_w increased from 0.324 to 0.923, meaning that the water adsorption capacity of MSO increased with increasing a_w. From the values of k_1 and k_2, MSO adsorbed faster and kept more water when stored in an environment with high a_w.

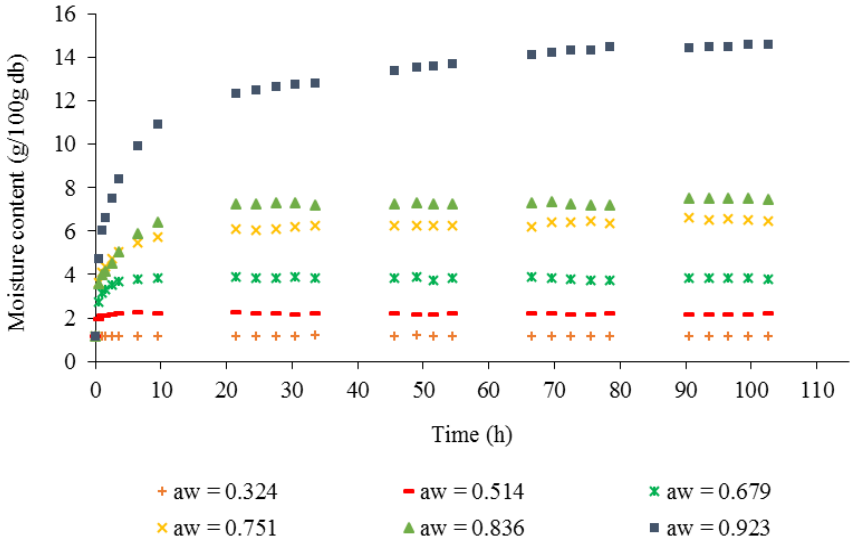

Figure 1. Moisture sorption curves of MSO at various water activities as a function of time

Table 2. Sorption kinetic model constants and coefficients of determination for MSO

Water activity	MSO		
	k_1	k_2	R^2
0.324	7.1160	18.1690	0.9917
0.679	0.4091	0.9534	0.9996
0.751	0.3255	0.1875	0.9998
0.836	0.2546	0.1537	0.9991
0.923	0.2217	0.0728	0.9996

k_1 = the constant of Peleg rate, k_2 = the constant of Peleg capacity, R^2 = the coefficient of determination

3.3 Moisture sorption isotherm of MSO

Changes in the experimental and predicted equilibrium moisture content (EMC) of MSO for a_w range of 0.324-0.923 are given in Figure 2. The results indicated that the EMC increased at higher a_w. The sorption isotherms obtained in this study are type III isotherms (J shape). Foods containing high-sugar levels typically show Type III isotherms and they adsorb relatively small amounts of water at low a_w and large amounts of water at high a_w, especially beyond 0.60 [26]. In this study, the EMC of MSO dramatically soared above a_w = 0.514. For a_w lower than 0.514, a slight increase in EMC with increasing a_w was found. This might have been due to the fact that adsorption of water occurs only at the surface of wall materials. For higher a_w (a_w > 0.514), the dissolution of soluble wall materials caused the increase in moisture content. Water molecules penetrate the pore structure of wall materials and are mechanically trapped in the void spaces of wall materials, especially at higher a_w. The uptake of water at higher a_w could be affected by the stability of the microporous structure [27]. The results reconfirmed that the constituents of the wall materials (fish gelatin, sodium caseinate and glucose syrup) were able to interact directly with the water molecules, which affected the moisture sorption ability of MSO. Type of isotherm (Type III isotherm) was similar to that reported for beta glucan-rich biscuits [28] and freeze-dried and spray-dried passion fruit pulp powders [29]. Panjagari *et al.* [28] reported that the sugar component of the product rendered the most significant influence on the sorption behavior. Barreiro *et al.* [30] reported that the EMC of barley malt sharply increased at higher a_w of 0.5, mainly at higher amounts of sugars.

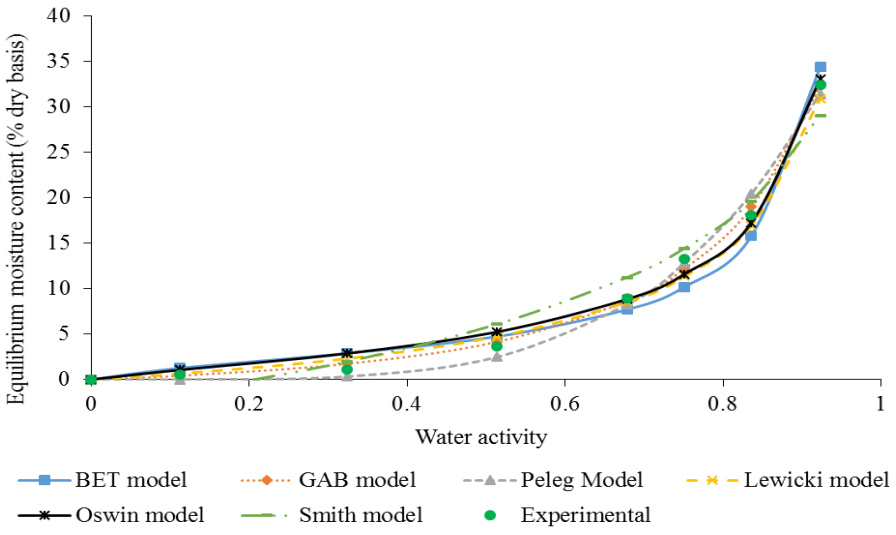

Figure 2. Experimental data and empirical models of moisture sorption isotherms for MSO at 30°C

3.4 Modeling of sorption isotherms

Various researchers have described the sorption isotherms for several products. Some of these models were empirical, others were semi-empirical. In this study, the sorption isotherms were predicted using six sorption isotherm equations: the BET, GAB, Peleg, Lewicki, Oswin and Smith models. These common mathematical equations used for describing moisture sorption in food

products are shown in Table 1. Those models have the parameters of two, three and four partial regression coefficients. The researchers correlated the experimental data with these moisture sorption isotherm models in order to identify the model of best fit. The isotherm models, equation constants and root mean square error (RMSE) for each model of MSO are summarized in Table 3. The goodness-of-fit of the experimental and predicted EMC values for the different sorption isotherm models were evaluated by RMSE, and lower values of RMSE pointed to the most suitable fit. The results showed that all investigated models gave a good description of the isotherms of spray-dried MSO at 30°C. However, the GAB model had the minimum values for RMSE (0.64) when used to predict the moisture sorption isotherm of spray-dried MSO at 30°C. It was found that the GAB model presented a good fitting for the spray-dried MSO at 30°C, and it was followed in increasing RMSE order of best-fit by the Smith, BET, Lewicki, Peleg and Oswin models, respectively. A similar result was reported by Zammouri *et al.* [31] for Hypril (pharmaceutical compounds). Zammouri *et al*. [31] found that the GAB model, Peleg model and the Oswin model fitted well with the water sorption isotherms of Hypril, but it was the Oswin model that presented the best fit. Referring to the GAB model, the monolayer moisture content (m_o) value of that model shows the amount of adsorbed water in a monolayer on the adsorbent surface. It is also used to measure the availability of active sorption sites in spray-dried MSO. It is a crucial quality factor for the planning of optimal storage conditions. The parameter *C* represents the stronger bonds between water molecules at monolayer and the binding sites on the adsorbent surface. In addition, *k* is a correction parameter for multilayer molecules relative to the bulk liquid. When *k* is 1, the molecules beyond the monolayer act as pure water [32]. Heat evaporation of the multilayer molecules is the same as pure water (k = 1) [33]. Sormoli and Langrish [34] used the GAB model to fit the moisture sorption data and found it to be more effective than other models they tried. The ability of the GAB model to fit the moisture sorption isotherms of foods with a wide range of a_w values was documented by Timmermann [35]. The monolayer moisture content shows the maximum shelf-life of dried products with a_w values between 0.2 and 0.3, and lipid oxidation can be initiated above this moisture content [36]. Changes in factors such as stickiness and caking of food powders occurred at a range of a_w from 0.35 to 0.5 [34]. However, the CMC of

Table 3. Sorption isotherm model constants and percentage of root mean square error for MSO

Sorption isotherm model	Constant	RMSE
BET	m_o = 2.69 C = 5.53	1.14
GAB	m_0 = 12.14 k = 0.87 C = 0.29	0.64
Peleg	a = 22.24 b = 4.34 c = 22.24 d = 4.34	1.90
Lewicki	F = 52.29 G = 0.38 H = 0.28	1.21
Oswin	k = 5.01 c = 0.76	2.12
Smith	C_1 = -2.93 C_2 = -12.47	0.90

food products in regard to microbial growth terms was recommended to be 0.6 [36]. Thus, the appropriate a_w for MSO should be at $a_w < 0.3$ and moisture content of lower than 12.14% (on a dry weight basis) was recommended (Table 3). The Smith model provides the sorption isotherm of biological substances. Al-Muhtaseb *et al.* [37] reported that the Smith model gave a good fit for experimental data between a_w values of 0.35 and 0.9. The BET model could be used to estimate the monolayer water content in the food products, and it was applicable at a_w range of 0.1-0.5 [38]. Nevertheless, the Lewicki model was applicable to a high range of a_w levels. The water content goes to infinity as a_w reaches 1.0 [17]. The Oswin model expressed the moisture isotherms throughout all the ranges of a_w [18]. Nonetheless, this present study found that the maximum RMSE value was predicted by the Oswin model. The non-sigmoidal and sigmoidal isotherms could be predicted using the Peleg model [16], since this model could predict the moisture sorption isotherm more effectively than the GAB model. The RMSE data from the Peleg model were higher than those of GAB model. The experimental and the predicted models of moisture sorption isotherms for spray-dried MSO are shown in Figure 3. Validation of the established model was evaluated by comparing the predicted moisture contents with their experimental counterparts in spray-dried MSO. The predicted data usually banded around the straight line, suggesting the mathematical model was suitable to describe the sorption behavior of spray-dried MSO.

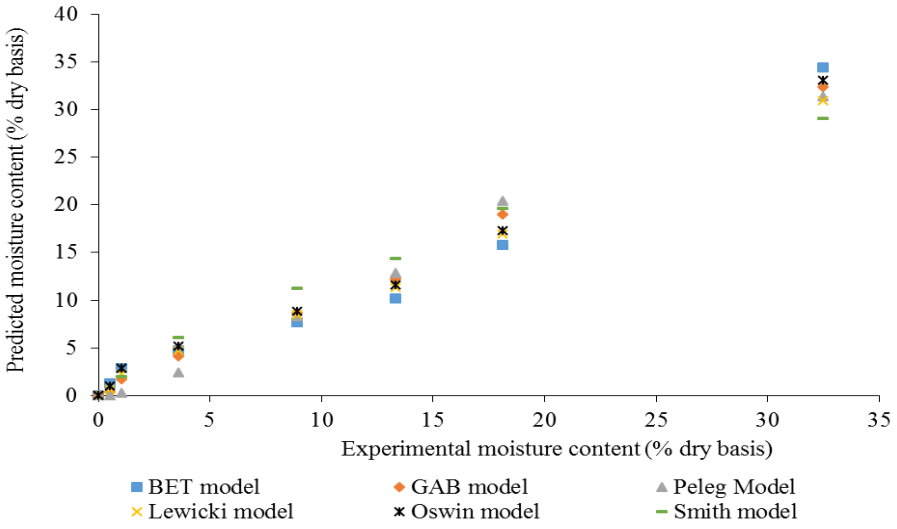

Figure 3. Comparison between experimental moisture content and predicted moisture content of MSO by various sorption models

3.5 Shelf-life prediction of MSO

The predicted shelf-life of spray-dried MSO in different commercial bags (PP, Nylon/LLDPE and metalized PET bags) at 30°C and either 75% or 80% RH is shown in Table 4. The shelf-life of

Table 4. Thickness and permeability coefficients of films and predicted shelf-life of MSO in different bags under 75% and 80% RH at 30°C

Package	Thickness (μm)	WVP ($\times 10^{-3}$ g.mm/day.m^2.mmHg)	Relative humidity (%)	Predicted shelf-life (days)
PP	89 ± 0.00a	6.09 ± 0.37b	75	722
			80	505
Nylon/LLDPE	82 ± 1.59c	9.31 ± 1.60a	75	436
			80	305
Metalized PET	85 ± 0.00b	5.76 ± 1.06b	75	725
			80	507

Data are expressed as mean ± SD (n=3), Lowercase letters in the same column indicate significant difference ($p < 0.05$).

spray-dried MSO is limited by moisture uptake. It is important to study the water vapour barrier properties of packaging materials. The moisture uptake of food products during storage depends on the moisture sorption isotherms of foods, the WVP of packaging film and environmental RH [12]. Table 4 shows that the average thickness of the PP, Nylon/LLDPE and metalized PET bags was found to be 89±0.00, 82±1.59 and 85±0.00 μm, respectively. The WVP of the bags was 6.09, 9.31 and 5.76 $\times 10^{-3}$ g.mm/day.m^2.mmHg for PP, Nylon/LLDPE and metalized PET bags, respectively. The results indicated that Nylon/LLDPE film bags had a higher WVP than the other two ($p < 0.05$), which was more likely related to its lower thickness (Table 4). However, there was no significant difference ($p > 0.05$) in WVP between metalized PET and PP films. Therefore, the commercial metalized PET and PP films had better water vapour barrier properties than the commercial Nylon/LLDPE film. The water vapour barrier property of film is an important parameter for estimating the shelf-life of food products. In this study, the shelf-life estimation of MSO depends on the critical limits used, initial product conditions, packaging conditions and environmental conditions. Equation 11 was used to calculate and predict the shelf-life of MSO in different commercial bags. The results indicated that the commercial metalized PET and PP bags extended the shelf-life of MSO more efficiently than the commercial Nylon/LLDPE bag at all % RH. However, the shelf-life of MSO packaged in PP bags was slightly lower than that of metalized PET bags. So, the different bags showed the influence of WVP on the shelf-life of MSO. For the same packaging, the results showed that MSO stored at 75% RH had longer shelf-life than MSO stored at 80% RH because of a lower moisture gradient between the storage environment and inside the packaging. The water molecules passed through the packaging film and the moisture content of dry food increased at a rapid pace at this early stage. Afterwards, the gradient of RH between the inside and outside of the packaging gradually decreased while the moisture content of dry food increased slowly until equilibrium with environmental RH was reached [39]. In conclusion, the longer shelf-life of MSO was calculated as 507 days in 80% RH and 725 days in 75% RH when MSO was packaged in metalized PET bags. The verification of shelf-life for MSO packaged in metalized PET bag was performed at 30°C, 75% RH. The EMC of MSO from the experiment was compared with the EMC from the prediction. The RMSE value of this equation was 0.711. In a similar study, Pisuchpen [40] reported that the experimental and the predicted values of EMC for hot curry cube in Nylon/LLDPE bags had the RMSE values of 1.88. The low RMSE value indicates the small difference between the experimental and predicted values. Thus, this equation could be considered a reliable tool for predicting the EMC with time for MSO in the metalized PET bag.

4. Conclusions

The isotherms and moisture sorption kinetics of MSO were made using various types of saturated salt solutions. The initial stage of the moisture sorption kinetics of MSO occurred relatively rapidly. However, the amount of water adsorbed decreased with time. MSO exhibited a Type III isotherm. The GAB model was the most appropriate model to estimate the moisture sorption isotherms of MSO. The predicted shelf-life of packaged MSO in metalized PET bags at 30°C and 75% RH fitted well with the actual shelf-life. Thus, the empirical models can be useful for predicting the shelf-life of MSO.

5. Acknowledgements

This work was financially supported by KMITL Research Fund under Researcher Development Grant for Project No. KREF016103 to Dr. SirimaTakeungwongtrakul. The authors would also like to thank Professor Dr. Anthony Keith Thompson, Visiting Professor at Faculty of Agro-Industry under Visiting Research Scholar granted by KMITL Research Fund, for invaluable suggestions and corrections.

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